Responses to Referee # 2

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 #2

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General:

The authors investigate organic nitrates formed from the oxidation of isoprene with NO3 radicals, illustrate the formation mechanisms of these organic nitrates (including HOM monomer, dimer, and trimers), their yield, and their contribution to SOA yield. The study is well designed and the data are well presented. If the authors can address my points and questions below, I would recommend the publication of the manuscript in Atmospheric Chemistry and Physics.

Specific:

1. Line 46 – It's a bit ambiguous for "both nucleation and growth of SOA". HOM are im-portant in nucleation of gaseous vapours, and they contribute to the growth of aerosol particles. Maybe a bit better to say e.g. HOM play a pivotal role in the atmospheric nucleation and also particle growth on pre-existing particles (secondary organic aerosol, SOA).

Response:

Accepted.

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

"HOM play a pivotal role in the atmospheric nucleation and also particle growth of pre-existing particles thus contributing to secondary organic aerosol (SOA)."

2. Line 92 – How about the chemical lifetime of the reaction of isoprene with NO3?

Response:

The chemical lifetime of isoprene with respect to NO_3 is calculated to be ~1.6 h and ~600 s using a NO_3 concentration of 10 ppt and 100 ppt, respectively. We did not intend to compare the lifetime of isoprene with respect to OH and NO_3. In the revised manuscript, we have revised this sentence as follows.

"Although isoprene from plants are mainly emitted under light conditions, i.e., in the daytime, isoprene can remain high after sunset in significant concentrations (Starn et al., 1998; Stroud et al., 2002; Brown et al., 2009) because of the reduced consumption by OH and is found to decay rapidly."

3. Line 94 – How significant is the reaction of isoprene with NO3 contributing to NO3 loss at night? Is it dominating in isoprene-dominated region? How about in monoterpene-dominated region? And during the day, how does it compare to the isoprene oxidation with OH?

Response:

The contribution of the reaction of isoprene with NO_3 to NO_3 loss depends on VOC composition. According to a number of field studies, the reaction of isoprene can be the dominant NO_3 loss channel in isoprene-dominated region, e.g. in Northeast US (Brown et al., 2009). In the monoterpene-dominated regions such as boreal forests, the reaction of isoprene with NO_3 may be not the dominant loss of NO_3. During most of the day, the reaction of isoprene with NO_3 cannot compete with its reaction with OH due to the fast reaction of NO_3 with NO and fast photolysis of NO_3. In the late afternoon, the isoprene oxidation by NO_3 can be comparable to that by OH under certain conditions e.g. reduced solar radiation and lower NO concentrations, and thus reduced OH concentration and reduced NO_3 loss rate (Ayres et al., 2015; Hamilton et al., 2021).

In the revised manuscript, we have modified this sentence to further define the significance of the reaction of isoprene with NO_3 as follows.

“Regarding the budget of NO_3, the reaction of isoprene with NO_3 can contribute to a significant or even dominant fraction of NO_3 loss at night in regions where VOC is dominated by isoprene such as Northeast US (Brown et al., 2009). Under some circumstances, the reaction of isoprene with NO_3 can contribute to a significant fraction during the afternoon and afterwards (Ayres et al., 2015; Hamilton et al., 2021).”

4. Line 143 – Please add the RH to describe how dry the condition was, e.g. RH<2% or lower. Also add the temperature inside the SAPHIR chamber somewhere in the Experimental part, e.g. line 125.

Response:

Accepted.

In the revised manuscript, we have added the description of temperature and RH.

“Experiments were conducted under dry condition (RH<2 %) and temperature was at 302±3 K.”

5. Line 158 – With a mass resolution of 4000, I am a bit curious is it difficult to distinguish different compounds at the same m/z (especially with one dominating compound), such as CHON1 from CHON3,5 compounds, CHON2 from CHON4 compounds, 1N-radicals from 2N-compounds, etc? It would be nicer to show a few masses of peak-fitting results of some organic nitrates in the supplement.

Response:

Accepted. We agree that it is not always easy to distinguish different compounds at the same m/z as their peaks overlap. Our approach is to “toggle” the inclusion of one peak and check the changes in the residue of peak fitting. If the toggling of inclusion of the peak does not significantly change the residue, we tend to not include the peak. Additionally, we also consider the double bond equivalence in the formula. For example, to distinguish $CHON_2$ from $CHON_4$, as in $C_{10}H_{16}N_2O_{12}$, and $C_{10}H_{20}N_4O_{10}$ at the same unit m/z, we consider 1) it is less likely to form H_20 in the reactions of isoprene with NO_3; 2) it is unlikely to form a compound with N4O10 considering the functionality (e.g. a NO_3- and a NO- group). When these peaks could not be distinguished, we did not include them in the peak list even if they may be actually present.

In the revised manuscript, we have shown some examples of peak-fitting results in the supplement.

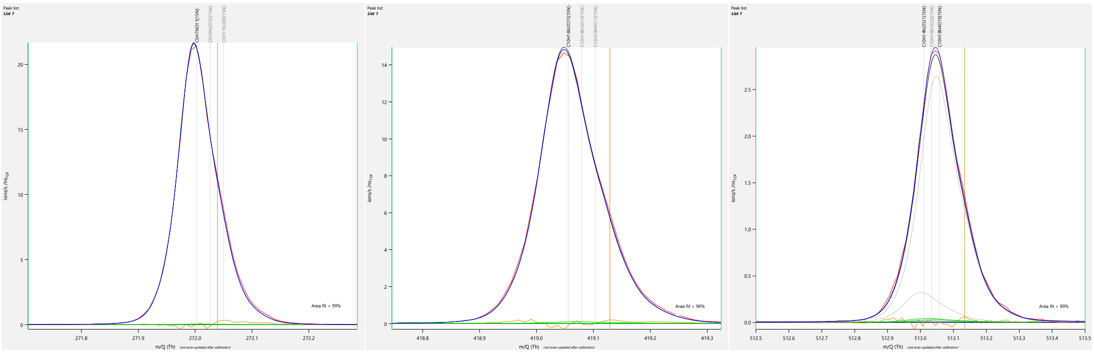

Figure S3. Examples of peak fitting. Formula in grey indicate compounds that have no noticeable effect on fitting residues and thus not included in the peak list.

6. Line 163 – Is the wall loss rate the same at different temperature in the SAPHIR chamber? Did you have the same chamber temperature as in Zhao et al., 2018?

Response:

The wall loss rate might be influenced by temperature in our chamber. It affects gas-wall equilibrium by changing evaporation rates and affects the condensation by changing diffusion in the boundary layer of the chamber. Because the reaction of NO_3 with isoprene cannot be stopped instantaneously as easy as photo-oxidation reactions by switching off illumination, we cannot directly determine the vapor wall loss rate from the experiments themselves. The temperature in this study 302±3 K is not the same as but within the range of our previous photo-oxidation experiments (298-314 K) (Zhao et al., 2018). Moreover, HOM yield is not sensitive to the vapor wall loss rate. An increase of wall loss rate by 100% or a decrease by 50% only changes the HOM yield by 11% and -6%, respectively. Therefore, we used the vapor loss rate determined in the photo-oxidation experiments. In the revised manuscript, we have discussed the influence of vapor wall loss rate as follows.

"Although the wall loss rate of vapors in this study might not be exactly the same as in our previous photo-oxidation experiments (Zhao et al., 2018), HOM yield is not sensitive to the vapor wall loss rate. An increase of vapor wall loss rate by 100% or a decrease by 50% only changes the HOM yield by 11% and -6%, respectively."

7. Line 195-197 – Why are the 2N-monomers dominating over 1N-monomers? Is it (partly) due to the reaction of NO3 radical with the remaining double bond of 1Nmonomer (forming 2N-monomers) being more reactive compared to the reaction of NO3 radical with the first double bond (C1) of isoprene (forming 1N-monomers)?

Response:

According to kinetic data of the reaction of NO_3 with isoprene and with the first-generation products, isoprene hydroxy nitrate, isoprene carbonyl nitrate, and isoprene peroxy nitrate, the reaction of NO_3 with the remaining double is slower than its reaction with isoprene (Wennberg et al., 2018). The higher abundance of HOM 2N-monomers than 1N-monomers is likely because HOM production rate via the autoxidation of 1N-RO_2 following the reaction of isoprene with NO_3 may be slower than that of the reaction of 1N-monomers (including both HOM and non-HOM monomers) with NO_3. We would like to note that some less oxygenated 1N-monomers such as $C_5H_9NO_{4/5}$ and $C_5H_7NO_4$ may have high abundance but are not detected by NO_3^--CIMS and are not HOM and thus not included in HOM 1N-monomers.

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

"The higher abundance of HOM 2N-monomers than 1N-monomers is likely because HOM production rate via the autoxidation of 1N-RO_2 following the reaction of isoprene with NO_3 may be slower than that of the reaction of 1N-monomers (including both HOM and non-HOM monomers) with NO_3. We would like to note that some less oxygenated 1N-monomers such as $C_5H_9NO_{4/5}$ and $C_5H_7NO_4$ may have high abundance but are not detected by NO_3^--CIMS and are not HOM and thus not included in HOM 1N-monomers."

8. Line 254-255 – If the fast loss of C5H9NO10 after isoprene addition is due to faster wall loss, why did the compound decay slower after 21:40 PM (which I think is partly due to wall loss) compared to those after isoprene addition? Maybe some other rea-sons are more important for its fast loss. Maybe it is similar to C5H9N2On radicals, that both its reactions and additionally wall loss contribute.

Response:

We meant the faster decay $C_5H_9NO_{10}$ relative to $C_5H_9NO_6$. After 21:40 PM, $C_5H_9NO_{10}$ showed a decay while $C_5H_9NO_6$ did not.

In the revised manuscript, we have modified this sentence to avoid ambiguity.

"The faster loss of $C_5H_9NO_{10}$ than $C_5H_9NO_6$ may result from the faster wall loss due to its lower volatility."

9. Line 264-265 – If the peak intensity of C5H8NO7 radicals in Fig. S4 is plotted in log scale, maybe it's more obvious that it increased during isoprene addition? As far as in the current figure, I cannot see that clearly and can only see it continuously increasing over time. But it's more clear for C5H8NO8 radicals that is responding to isoprene addition.

Response:

The description of $C_5H_8NO_7$• and $C_5H_8NO_8$• was swapped by mistake and it is $C_5H_8NO_7$• that showed more mostly a time profile of second-generation products instead. In the revised manuscript, we have corrected this error and re-organized this paragraph as follows.

"The second-generation products may be different isomers formed in pathways other than Scheme 1. Second-generation $C_5H_9NO_6$ can be formed via $C_5H_8NO_7$•, which can also be formed by the reaction of NO_3 and O_2 with

$C_5H_8O_2$ as mentioned above (Scheme S2b), or by the reaction of OH with $C_5H_7NO_4$ (Scheme S2a). The time profiles of $C_5H_8NO_7$• did show more contribution of second-generation processes because it continuously increased with time in general. If the pathways via the reaction of NO_3 and O_2 with $C_5H_8O_2$ and the reaction of OH with $C_5H_7NO_4$ contribute most to $C_5H_9NO_6$, $C_5H_9NO_6$ would show mostly a time profile of second-generation products. Similarly, second-generation $C_5H_9NO_7$ can be formed via $C_5H_8NO_7$• or $C_5H_8NO_8$•. The time series of $C_5H_8NO_8$• showed the contribution of both the first- and second-generation processes, which generally increased with time while also responding to isoprene addition (Fig. S4). Similar to $C_5H_9NO_6$, the second-generation pathway for $C_5H_9NO_7$, $C_5H_9NO_9$, and $C_5H_9NO_{10}$ are shown in Scheme S1, S3, S4. For the RO_2 in $C_5H_8NO_n$• series other than $C_5H_8NO_{7/8}$•, the peak of $C_5H_8NO_n$• overlaps with $C_5H_{10}N_2O_n$ in the mass spectra, which is a much larger peak, and thus cannot be easily differentiated from $C_5H_{10}N_2O_n$. Therefore, it is not possible to obtain reliable separate time profiles in order to differentiate their major sources. It is worth noting that nitrate CIMS may not be able to sensitively detect all isomers of $C_5H_9NO_6$ due to the sensitivity limitation. Therefore, we cannot exclude the possibility that the absence of some first-generation isomers of $C_5H_9NO_6$ was due to the low sensitivity of these isomers."

10. Line 267-270 – For the RO2 in C5H8NOn radicals series, do you mean the radicals with n>=9 cannot be distinguished from 2N-monomers, but C5H8NO7 radicals and C5H8NO8 radicals (you shown in Fig. S4) can be distinguished because they don't have interference compounds nearby them?

Response:

Yes. To make it more clear, in the revised manuscript, we have modified this sentence as follows.

"For the RO_2 in $C_5H_8NO_n$• series other than $C_5H_8NO_{7/8}$•, the peak of $C_5H_8NO_n$• overlaps with $C_5H_{10}N_2O_n$ in the mass spectra, which is a much larger peak, and thus cannot be differentiated from $C_5H_{10}N_2O_n$."

11. Line 303 – Other than SOAS, C5 organic nitrates (C5H7-11NO4-9) were also observed in both gas phase and particle phase with FIGAERO-CIMS in a rural area in Germany (Huang et al., EST, 2019), although their measurement site was not an isoprene-dominating region.

Response:

We thank the reviewer's reminder. In the revised manuscript, we have added this citation as follows.

"$C_5H_xNO_{4\text{-}9}$ and $C_5H_xNO_{4\text{-}10}$ have been observed in the gas phase and particle, respectively, in a rural area in southwest Germany (Huang et al., 2019)."

12. Line 349-350 – "C5H9N2On radicals M2b were formed from C5H9NO4 followed by an alkoxy-peroxy step" is from Scheme 3b, not from Scheme 2.

Response:

Corrected.

13. Line 356 – It's not the case for C5H10N2On and C5H8N2On with n=7. With increasing oxygen number, they increased and then decreased.

Response:

We apologize that Fig. 3b was not updated. In the revised manuscript, we have updated the legend with $C_5H_{10}N_2O_7$ omitted as we are not confident with their assignment. For $C_5H_8N_2O_n$, we have modified this sentence as follows.

"The intensity of $C_5H_8N_2O_n$ first increased and then decreased with oxygen number while $C_5H_{10}N_2O_n$ decreased with oxygen number with the $C_5H_{10}N_2O_8$ and $C_5H_8N_2O_8$ being the most abundant within their respective series."

14. Line 506 – For these 1N-dimers, have you checked the monoterpene concentration from PTR in the SAPHIR chamber? Is the monoterpene concentration in the chamber low enough not to produce any C10H16NOn molecules to interference/contaminate the results?

Response:

The monoterpene concentration in the chamber during the study is below the limit of detection, which is ~50 ppt (3σ). Therefore, there is unlikely interference to $C_{10}H_{16}NO_n$ from monoterpenes. In the revised manuscript, we have added the following note:

"We would like to note that there is unlikely interference to C_{10}-HOM from monoterpenes, which has been reported previously (Bernhammer et al., 2018), as the concentration of monoterpenes in the chamber during this study was below the limit of detection, which was ~50 ppt (3σ)."

15. Line 523-524 – Why only C5H8NOn radicals with n>8 can react with isoprene? The C5H8NOn radicals detected (in Table 1) has an n range of 7-12, and the n range for C10H16NOn are expected to be between 9 and 14, based on R13. But in Table 2, the n range for C10H16NOn is between 10 and 16. Could you infer whether the smaller n (n<=8) for C5H8NOn radicals can work or not to form C10H16NOn from R13? If not, could you give a possible explanation why the smaller n cannot work? And also how was C10H16NO16 formed?

Response:

We have further checked our data and the n range for $C_{10}H_{16}NO_n$ should be 10-14. In the revised manuscript, we have corrected this. We expected the n range of $C_{10}H_{16}NO_n$• to be 7-14 because besides the $C_5H_8NO_n$• detected by our NO_3^--CIMS, there should be $C_5H_8NO_n$(n=5,6) according to the reaction mechanism. Among these $C_{10}H_{16}NO_n$• compounds, $C_{10}H_{16}NO_{7\text{-}9}$• are expected to be detectable by NO_3^--CIMS. The absence of these compounds in the mass spectra is likely attributed to their low concentration, which might result from low precursor concentration, low reaction rate with isoprene, and/or fast reaction with other radicals. In addition, we would like to note that the $C_{10}H_{16}NO_n$• series has low signal in the mass spectra and their assignment and thus range of n may be subject to uncertainties. In the revised, we have added this note and revised this part as follows.

"Only $C_{10}H_{16}NO_n$• with n≥10 were detected, while according to the mechanism of self-reaction between $C_5H_8NO_n$•, the n range of $C_{10}H_{16}NO_n$• is expected to be 7-14. The absence of $C_{10}H_{16}NO_{n(n<10)}$• is likely attributed to their low abundance, which might result from low precursor concentrations, low reaction rates with isoprene, and/or faster reactive losses with other radicals."

"We note that due to their low signals in the mass spectra, their assignment and thus range of n may be subject to uncertainties."

16. Line 568-571 – Can you also check the sesquiterpene concentration in the chamber to exclude the contamination of its products?

Response:

The sesquiterpene concentration in the chamber during this study was below the limit of detection, which was ~50 ppt (3σ). Therefore, there is unlikely contamination from sesquiterpene.

17. Line 603-604 – From the results, it's suggesting whether they were first-generation products or second-generation products, or a mix/combination of both, if I didn't misunderstand.

Response:

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

"The time profiles provide additional constraints on their formation mechanism beside the molecular formula, suggesting whether they were first-generation products, second-generation products or a combination of both."

Technical:

1. Line 98 – "initials". Do you mean "initial"?

Response:

In the revised manuscript, we have changed it to "initial".

2. Line 116 – Change to "in the isoprene+NO3 system" throughout the manuscript.

Response:

In the revised manuscript, we have changed the phrase to "in the isoprene+NO_3 reaction" throughout the manuscript.

3. Line 123 – The first letter "s" in the word "system" is in Italic.

Response: Corrected.

4. Line 148 – Can you plot the Fig. S1 similar to Fig. S2 so that easier to compare with Fig. S2? Or just label those additions directly in Fig. S2? Because there is no x axis in the SAPHIR box in the figure, but it seems the x axis is time.

Response:

Accepted. In the revised manuscript, we have added further labels in Fig. S2 to show the times of additions of isoprene, NO_2, and O_3.

5. Line 186 – Please specify in the Figure 1 caption that the m/z has included the reagent ion (15NO3-).

Response: Accepted.

6. Line 204 – (a) the labelled species in the figure 2 caption at m/z 351 and 353 should be C5H8N2O8 and C5H10N2O8 with H[15N]2O6-. Also in the figure itself, they are labeled as C5H8N2O14H[15N]2O6- and C5H10N2O14H[15N]2O6-. Please double check them and correct. (b) I suppose the number on the upper left corner of each compound is the m/z including the reagent ion (15NO3-). Please also specify that in the Fig. 2 caption. (c) What's the blue circle in the figure at m/z around 390?

Response: We thank the reviewer for pointing out this typo. We have corrected the captions and labeling in the figure, and also added the note of the m/z.

The blue circle is $C_6F_{11}HO_3^-$, coming from chamber wall. In the revised manuscript, we chose to not plot this compound in the figure to avoid confusion.

7. Line 211 –In Scheme 1 caption, it says (b) is for n = 8, 10, 12. But in the Scheme 1b itself, it's showing compounds with n = 4, 6, 8. Complete the Scheme 1b to show the compounds with n = 10 and 12.

Response:

Accepted. In the revised manuscript, we have added the pathway to form $C_5H_9NO_{10}$•. The peak of $C_5H_9NO_{12}$• overlaps with $C_5H_9NO_8$•$H(^{15}NO_3)_2^-$. After further checking the spectra, to be cautious we decided to not assign it.

8. Line 235 – Remove the first "in".

Response: accepted.

9. Line 310 – n = 9, 11 are odd number of oxygen atoms, instead of even number.

Response:

We have corrected this error in the revised manuscript.

10. Line 339 – In Scheme 2a, -ONO2 is missed from the 2nd molecule.

Response:

We have corrected this mislabeling in the revised manuscript.

11. Line 345 – In Scheme 3b, -ONO2 is missed from the 2nd molecule.

Response:

We have corrected this mislabeling in the revised manuscript.

12. Line 372 – Change "VOCs" to "VOC". You have been using "VOC" previously in the manuscript. So make it consistent.

Response:

Accepted.

13. Line 385 – Could you separate these 3N-monomers in Fig. S8 into two panels, or use log scale? It's not obvious to see their time profile, especially for compounds with oxygen number bigger than 12, which were overlapping on top of each other.

Response:

Accepted. We use two panels to show them in the revised manuscript.

14. Line 390 – Change "Such an mechanism" to "Such a mechanism".

Response: Accepted.

15. Line 438-439 – Could you visualize the mass defect plots in Fig. 4 in a better way? Compound series were so close to each other to see clearly which compound belongs to which line. And also it's better to label the number of oxygen atoms to guide audience since you are discussing a lot of oxygen numbers. Both for Fig. 4 and Fig. 2. For Fig. S11, compound series were not so many and therefore not so difficult to distinguish.

Response: Accepted. In the revised manuscript, we have added the number of oxygen atoms. Also we have adjusted the scale of y axis, removed the grid line, and colored the HOM series lines in different colors to show the data points more clearly.

16. Line 443 – Since you have mentioned the compounds clustered with $H[^{15}N]_2O_6^-$ were labelled in grey in Figure 2 caption. Do the same for Figure 4 caption for the compound in grey ($C_{10}H_{17}N_3O_{12}$-$14H[^{15}N]_2O_6^-$).

Response: Accepted.

17. Line 551-552 – Most abundant trimers were $C_{15}H_{24}N_4O_n$ (n=17-23), but in Fig. S11, n=16-22. Double check. Also double check all the n ranges for each compound series in the figures, tables, and text throughout the manuscript and supplement.

Response: We thank the reviewer for pointing this inconsistency. In the revised manuscript, we have corrected the numbers. Also we have checked throughout the manuscript for all series and fixed the inconsistency in the n ranges.

18. Line 556 – The formation pathways of dimer RO2 $C_{10}H_{16}N_3O_n$ (n=14-20) and $C_{10}H_{17}N_2O_n$ are shown in R10 and R11.

Response:

In the revised manuscript, we have corrected this error.

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