

We thank the reviewer for the comments on our manuscript. The comments and suggestions are greatly appreciated. All the comments have been addressed and we believe that the revisions based on these comments improved the quality of our manuscript. Below 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.

*This study provided a detailed analysis of the HOM formation from limonene reacting with NO<sub>3</sub> radical. These HOMs are potentially important in forming new particles and secondary organic aerosol. In addition, this study provides observational evidence on the formation of HOM-trimer from NO<sub>3</sub> oxidation of limonene, to my best knowledge, for the 1st time. However, I have a few comments to be addressed before this manuscript can be published in ACP.*

- 1. The concentration issue: The monoterpene concentrations are still much higher than in most regions. This could be why the dimer/monomer ratio is so high, and why can you observe trimer? However, this is far from the real atmosphere, where dimers concentration is usually too low to react with the other oxidant before condensing to particles. As shown in Fig.5 and Fig. 6, some aerosol exists universally in the atmosphere; they can scavenge HOM dimers so effectively that the dimers have no time to react with oxidants again to form a trimer.*

**Response:**

We agree that the higher monoterpene concentrations may favor trimer formation. Although monoterpene concentrations in this study (0-0.92 ppbv) are higher than in most ambient regions, they are still in the range of ambient concentrations (~0.01-1 ppbv) (e.g. Coggon et al., 2021; Wang et al., 2022), especially for forested regions (e.g. Xu et al., 2015; Kontkanen et al., 2016; Janson, 1992).

Regarding the influence of aerosols, the relative loss of dimers by condensations on aerosols and by gas-phase reaction depends on the concentrations of aerosol and oxidants. Assuming that dimers react with NO<sub>3</sub> at a rate constant similar to limonene and have a condensation sink similar to H<sub>2</sub>SO<sub>4</sub> ( $10^{-3}$  to  $10^{-1}$  s<sup>-1</sup>) (Dada et al., 2020) at a NO<sub>3</sub> concentration of 5-300 ppt, the lifetime with respect to NO<sub>3</sub> and to condensation on particles are ~0.1-10 min and ~0.1-20 min, respectively. Therefore, although aerosols may scavenge HOM dimers hindering the formation of trimers, dimers can still react with oxidant, at least with NO<sub>3</sub> at nighttime, forming trimers. Such reactions are particularly important when the ambient aerosol concentration is low (similar to the situation of the early stage of our experiment).

In the revised manuscript, we have added the following discussion regarding the VOC concentrations:

“Since our experiment of NO<sub>3</sub> oxidation of limonene was performed under near atmospheric conditions, such NPF events induced by the oxidation of limonene by NO<sub>3</sub> could also occur in the ambient atmosphere. Although monoterpene concentrations in this study (0-0.92 ppbv) are higher than in most ambient regions, they are still in the range of ambient concentrations (~0.01-1 ppbv) (e.g. Coggon et al., 2021; Wang et al., 2022), especially for forested regions (e.g. Xu et al., 2015; Kontkanen et al., 2016; Janson, 1992). Assuming that dimers react with NO<sub>3</sub> at a rate similar to limonene, and that they have a condensation sink similar to H<sub>2</sub>SO<sub>4</sub> (10<sup>-3</sup> to 10<sup>-1</sup> s<sup>-1</sup>) (Dada et al., 2020), the lifetime with respect to NO<sub>3</sub> at an NO<sub>3</sub> concentration of 5-300 ppt and to condensation on particles are ~0.1-10 min and ~0.1-20 min, respectively. Therefore, although aerosols may scavenge HOM dimers in the ambient atmosphere, dimers can still react with NO<sub>3</sub> at nighttime, forming trimers. Such reactions are particularly important when the ambient aerosol concentration is low.”

2. *Self-termination of HOM-RO<sub>2</sub>: One of this work's major conclusions is that the HOM-RO<sub>2</sub> self-termination is more important than the previously understood. I believe more evidence is needed to support this point. For example, is it possible C<sub>10</sub>H<sub>15</sub>NO<sub>x</sub> is formed from C<sub>10</sub>H<sub>15</sub>NO<sub>x</sub>+NO<sub>3</sub>? In addition, what's the potential influence of the differences in instrument sensitivity on detecting carbonyl compounds and hydroxyl compounds?*

**Response:**

We observed much more carbonylnitrates than hydroxynitrates (or hydroperoxynitrates) in the gas-phase products in the limonene + NO<sub>3</sub> reaction system. As discussed in our previous study by Shen et al. (2021), this higher abundance of carbonylnitrates is not likely to be explained by the reaction of alkoxy RO• + O<sub>2</sub> forming carbonyls and HO<sub>2</sub>•, decomposition of β-nitrooxyperoxynitrate or self-reactions of RO<sub>2</sub>• via the Bennett and Summers mechanism forming carbonyls and H<sub>2</sub>O<sub>2</sub>. Reactions between RO<sub>2</sub>• in general produce overall equal amounts of carbonyl and hydroxyl compounds. The decomposition of β-nitrooxyperoxynitrate is slow in the gas-phase. The reaction of alkoxy RO• with O<sub>2</sub> for large RO• is generally slower than isomerization and decomposition. Thus, the higher abundance of carbonylnitrates than hydroxynitrates may be attributed to unimolecular termination of HOM-RO<sub>2</sub>•. In addition, isomerization of RO• forming carbonyl compounds may also contribute to this finding.

A C<sub>10</sub>H<sub>15</sub>NO<sub>x</sub> compound could be formed via the reaction of C<sub>10</sub>H<sub>15</sub>NO<sub>x-2</sub> with NO<sub>3</sub> and O<sub>2</sub> followed by the conversion of RO<sub>2</sub>• to RO• and the dissociation forming NO<sub>2</sub>. However, this reaction does not have an impact on the total amount of C<sub>10</sub>H<sub>15</sub>NO<sub>x</sub>, which still need a source. Also, main products of C<sub>10</sub>H<sub>15</sub>NO<sub>x</sub>+NO<sub>3</sub> are compounds containing 2 N atoms.

According to Hyttinen et al. (2015), for nitrate CI-API-TOF, HOM containing two hydrogen bond donors

(such as -OOH and -OH group) have strong binding energy with  $\text{NO}_3^-$ . Additional hydrogen bond donors only enhance the binding energy marginally. If we compare HOM carbonyl product (such as  $\text{C}_{10}\text{H}_{15}\text{NO}_{10}$ ) with the corresponding hydroxy product ( $\text{C}_{10}\text{H}_{17}\text{NO}_{10}$ ), they only differ in one functional group. As both are highly functionalized, it is likely that carbonyl HOM have a quite similar sensitivity with hydroxyl HOM. If the sensitivity of carbonyl HOM were lower, this would result in even more dominance of carbonyl HOM over hydroxyl HOM. Thus, we conclude that carbonylnitrates are more abundant than hydroxynitrates or hydroperoxynitrates.

In the revised manuscript, we have added discussion on this point as follows.

“As discussed in our previous study by Shen et al. (2021), this higher abundance of carbonylnitrates is not likely to be explained by the reaction of alkoxy  $\text{RO}\cdot + \text{O}_2$  forming carbonyls and  $\text{HO}_2\cdot$ , decomposition of  $\beta$ -nitrooxyperoxynitrate or self-reactions of  $\text{RO}_2\cdot$  via the Bennett and Summers mechanism forming carbonyls and  $\text{H}_2\text{O}_2$ . Reactions between  $\text{RO}_2\cdot$  in general should produce overall equal amounts of carbonyl and hydroxyl compounds. The decomposition of  $\beta$ -nitrooxyperoxynitrate is slow in the gas-phase. The reaction of alkoxy  $\text{RO}\cdot$  with  $\text{O}_2$  for large  $\text{RO}\cdot$  is generally slower than isomerization and decomposition (Vereecken and Peeters, 2009, 2010). Thus, the higher abundance of carbonylnitrates compared to hydroxynitrates may be attributed to unimolecular termination of  $\text{HOM-RO}_2\cdot$ . In addition, isomerization of  $\text{RO}\cdot$  forming carbonyl compounds may also contribute to this finding.”

“According to Hyttinen et al. (2015), for nitrate CI-API-TOF, HOM containing two hydrogen bond donors (such as -OOH and -OH group) have strong binding energy with  $\text{NO}_3^-$ . Additional hydrogen bond donors only enhance the binding energy marginally. If we compare HOM carbonyl product (such as  $\text{C}_{10}\text{H}_{15}\text{NO}_{10}$ ) with the corresponding hydroxy product ( $\text{C}_{10}\text{H}_{17}\text{NO}_{10}$ ), they only differ in one functional group. As both are highly functionalized, it is likely that HOM carbonyl have a quite similar sensitivity with hydroxyl HOM. If the sensitivity of carbonyl HOM were lower, this would result in even more dominance of carbonyl HOM over hydroxyl HOM. Thus, we conclude that carbonylnitrates are more abundant than hydroxynitrates or hydroperoxynitrates.”

3. *I would suggest adding more discussions on the potential influence of ozone oxidation of limonene in the system, as well as the potential role of HO forming via ozonolysis of monoterpene. As shown in Table 2,  $\text{C}_{20}\text{H}_{33}\text{NO}_x$  and  $\text{C}_{20}\text{H}_{34}\text{N}_4\text{O}_x$  are likely from OH oxidation. In addition, the abundance of  $\text{C}_{20}\text{H}_{31}\text{NO}_x$  ( $x=10-15$ ) is considerably high may also indicate the role of  $\text{O}_3$  chemistry.*

**Response:**

Accepted. We agree that  $\text{C}_{20}\text{H}_{33}\text{NO}_x$  may be formed via accretion reaction of  $\text{C}_{10}\text{H}_{16}\text{NO}_x\cdot$  with  $\text{C}_{10}\text{H}_{17}\text{O}_x\cdot$ , which is formed via OH oxidation of  $\text{C}_{10}\text{H}_{16}$ .  $\text{C}_{20}\text{H}_{34}\text{N}_4\text{O}_x$  may be formed via accretion reaction of two

$C_{10}H_{17}N_2O_x\bullet$ , which can be formed via OH oxidation of  $C_{10}H_{16}N_2O_x$ . Also, the considerably high abundance of  $C_{20}H_{31}NO_x$  ( $x=10-15$ ) may be partly attributed to the contribution of  $O_3$  chemistry. The revised texts are as follows:

“We cannot exclude that the formation pathway of  $C_{20}H_{33}NO_x$ ,  $C_{20}H_{34}N_4O_x$  and  $C_{19}H_{31}NO_x$  may also involve limonene oxidation by  $OH\bullet$  (Table 2), which can be formed in the ozonolysis of limonene as a minor pathway. In addition, the high abundance of  $C_{20}H_{31}NO_x$  ( $x=10-15$ ) among the dimers may be partly attributed to a contribution of the reaction of limonene with  $O_3$ .”

We have also updated Table 2 to reflect the potential contribution of OH oxidation as follows:

Table 2. Major dimer and trimer families and their possible formation pathways.

Dimer/Trimer family	Possible formation pathways
$C_{20}H_{32}N_2O_x$	$C_{10}H_{16}NO_x\bullet + C_{10}H_{16}NO_x\bullet$
$C_{20}H_{33}N_3O_x / C_{20}H_{31}N_3O_x$	$C_{20}H_{32}N_2O_x + NO_3 + HO_2\bullet/RO_2\bullet$
$C_{20}H_{31}NO_x$	$C_{10}H_{16}NO_x\bullet + C_{10}H_{15}O_x\bullet$
$C_{20}H_{33}NO_x$	$C_{10}H_{16} + OH\bullet + C_{10}H_{16}NO_x\bullet$
$C_{20}H_{34}N_4O_x$	$(C_{10}H_{16}N_2O_x + OH\bullet) + (C_{10}H_{16}N_2O_x + OH\bullet)$
$C_{19}H_{30}N_2O_x$	$C_{10}H_{16}NO_x\bullet + C_9H_{14}NO_x\bullet$
$C_{19}H_{31}N_3O_x$	$C_{19}H_{30}N_2O_x + NO_3 + HO_2\bullet/RO_2\bullet$
$C_{19}H_{29}NO_x$	$C_9H_{14}NO_x\bullet + C_{10}H_{15}O_x\bullet$
$C_{19}H_{31}NO_x$	$C_{10}H_{16} + OH\bullet + C_9H_{14}NO_x\bullet$
$C_{30}H_{48}N_4O_x$	$C_{20}H_{32}N_3O_x\bullet + C_{10}H_{16}NO_x\bullet$
$C_{30}H_{47}N_3O_x$	$C_{20}H_{31}N_2O_x\bullet + C_{10}H_{16}NO_x\bullet$

4. *There are so many details in the study, which is good, but makes the manuscript not so easy to follow. I suggest adding some summary statement in each section.*

**Response:**

Accepted. We have added a summary statement at the end of each sub-section in the result section.

*Detailed comments:*

5. *There is growing evidence that monoterpene-OOMs are also important in urban regions. I suggest adding some discussion in the introduction part, i.e., Liu et al., 2021, ACP; Nie et al., 2022, Nat. Geosci.*

**Response:**

Accepted. The revised texts are as follows:

“Besides the observations at forested regions, monoterpene derived HOM via NO<sub>3</sub> oxidation also contribute to organic aerosols in urban regions. For example, Liu et al. (2021) and Nie et al. (2022) have found that HOM derived from monoterpene nighttime chemistry are important in megacities in China, especially during summertime.”

6. *Line 162: what's the concentration of isoprene-HOMs in the chamber? Can they influence the subsequent reactions?*

**Response:**

The concentration of isoprene-HOM in our chamber are less than 1 ppt. Besides, all the isoprene-HOM observed (C<sub>5</sub>H<sub>9</sub>NO<sub>7,10</sub>, C<sub>5</sub>H<sub>8</sub>N<sub>2</sub>O<sub>8-10</sub>, C<sub>5</sub>H<sub>10</sub>N<sub>2</sub>O<sub>8</sub>, C<sub>5</sub>H<sub>9</sub>N<sub>3</sub>O<sub>9,10</sub>) are saturated and do not contain C=C double bond. Therefore, the isoprene-HOM will not influence the reaction of limonene with NO<sub>3</sub> in this study.

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

“These isoprene oxidation products were likely formed in an isoprene + NO<sub>3</sub> experiment performed two days before (Zhao et al., 2021) and released slowly from chamber walls due to their semi-volatile character. Their total concentration is less than 1 ppt. All the isoprene-HOM observed (C<sub>5</sub>H<sub>9</sub>NO<sub>7,10</sub>, C<sub>5</sub>H<sub>8</sub>N<sub>2</sub>O<sub>8-10</sub>, C<sub>5</sub>H<sub>10</sub>N<sub>2</sub>O<sub>8</sub>, C<sub>5</sub>H<sub>9</sub>N<sub>3</sub>O<sub>9,10</sub>) are saturated and do not contain C=C double bond. The isoprene-HOM will not influence the reaction of limonene with NO<sub>3</sub> in this study. Therefore, they are not discussed as products from the limonene oxidation in our experiment.”

7. *Line 179-180: More discussion on the mass-independent transmission calibration rather than citing a reference.*

*Calibration issue: the authors quantify observed HOMs by calibrating H<sub>2</sub>SO<sub>4</sub> and assuming they have similar charging efficiency. However, besides charging efficiency, the transmission would influence the calibration coefficient either (Junninen et al., 2010), especially when referring to a bunch of the molecules covering a wide mass range. Like in this study, one can reasonably speculate the transmission between HOM-dimers and HOM-monomers is different. I may recommend reconsidering the calibration factors used in the current version.*

**Response:**

Accepted. We have added discussion about the dependence of transmission on m/z as follows:

“A mass-independent transmission efficiency was used according to our previous study, which causes an additional uncertainty of 14 % (Pullinen et al., 2020). In this previous study, the transmission efficiency curve of nitrate CI-API-TOF was determined and found to monotonously decrease with increasing mass of ions but only slightly depend on the mass range (14% change). As we used the same setting as our previous study, we have included the slight dependence of transmission on  $m/z$  in the uncertainties.”

8. *How to calculate the wall loss of N<sub>2</sub>O<sub>5</sub> of the chamber?*

**Response:**

The wall loss rate constant of N<sub>2</sub>O<sub>5</sub> in the SAPHIR chamber is  $7.2 \times 10^{-5} \text{ s}^{-1}$  (Fry et al., 2009), and the lifetime of N<sub>2</sub>O<sub>5</sub> due to wall loss is about 4 h. As the HOM yield determination is based on the first 3 min, the wall loss of N<sub>2</sub>O<sub>5</sub> can be ignored compared with the loss via the reaction of NO<sub>3</sub> with limonene.

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

“The wall loss rate constant of N<sub>2</sub>O<sub>5</sub> in the SAPHIR chamber is  $7.2 \times 10^{-5} \text{ s}^{-1}$  (Fry et al., 2009). As the HOM yield determination is based on the first 3 min, the wall loss of N<sub>2</sub>O<sub>5</sub> can be ignored compared to the loss via the reaction of NO<sub>3</sub> with limonene.”

9. *Line 209-212: SVOCs can also contribute to SOA formation, especially in the case when SOA monotonic increases.*

**Response:**

We agree that SVOC can contribute to SOA formation, especially when particle concentrations are relatively high. However, according to our analysis (Sect. 3.6), the LVOC and ELVOC already constitute most of the SOA in this study and the fraction of SVOC in total SOA must be low. Therefore, the contribution of SVOCs to SOA formation is likely small in this study, especially during the early stage of particle formation.

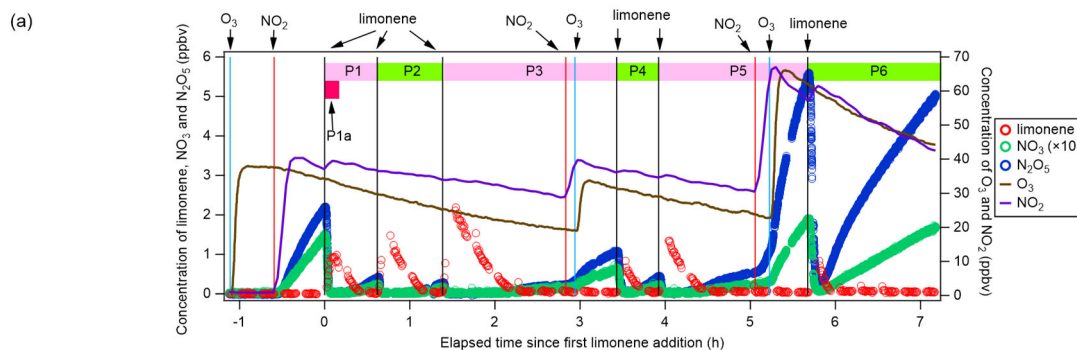
In the revised manuscript, we have modified this sentence as follows.

“In Scenario 2, only the irreversible uptake of LVOC and ULVOC/ELVOC compounds were considered to contribute to the growth of SOA particles in order to examine the role of LVOC and ELVOC while IVOC and SVOC were not included, although they may also contribute to SOA.”

10. Please mark clearly of P1 to P6 in Figure 1. The current version makes the statements in the text a bit hard to follow.

**Response:**

Accepted. In the revised manuscript, Fig. 1a has been modified in which P1 to P6 as well as P1a are marked clearly (also shown below).



11. Line 278-279: why there was only one peak of C<sub>10</sub>H<sub>15</sub>NO<sub>9</sub>?

**Response:**

As we pointed out in the text, we could not explain the behavior of C<sub>10</sub>H<sub>15</sub>NO<sub>9</sub> in section 3.2.2: “At this time, we do not have a reasonable explanation for the trend of C<sub>10</sub>H<sub>15</sub>NO<sub>9</sub>, though we should consider that there are many isomers at play, which may have very different chemical pathways (un)available.” We suspect that C<sub>10</sub>H<sub>15</sub>NO<sub>9</sub> may contain multiple isomers, which follow patterns of different generation products and together result in the general trend of only one peak with time. However, we cannot justify this speculation.

12. Line 296: why the pattern is 16 TH intervals other than 32 TH intervals?

**Response:**

According to autoxidation mechanism, the pattern should be 32 Th intervals. However, RO<sub>2</sub>• can be transformed to RO• via reactions with NO, other RO<sub>2</sub>•, or NO<sub>3</sub>, which leads to a pattern of 16 Th interval.

In the revised manuscript, we have further clarified this point as follows.

“Such a pattern is attributed to autoxidation of RO<sub>2</sub>• (with 32 Th interval for each O<sub>2</sub> addition) plus the alkoxy-peroxy pathway (shifted by 16 Th compared with exclusive autoxidation) as discussed below.”

13. Line 410: Can C<sub>10</sub>H<sub>14</sub>O<sub>x</sub> be formed from proposed NO<sub>3</sub> oxidation pathways?

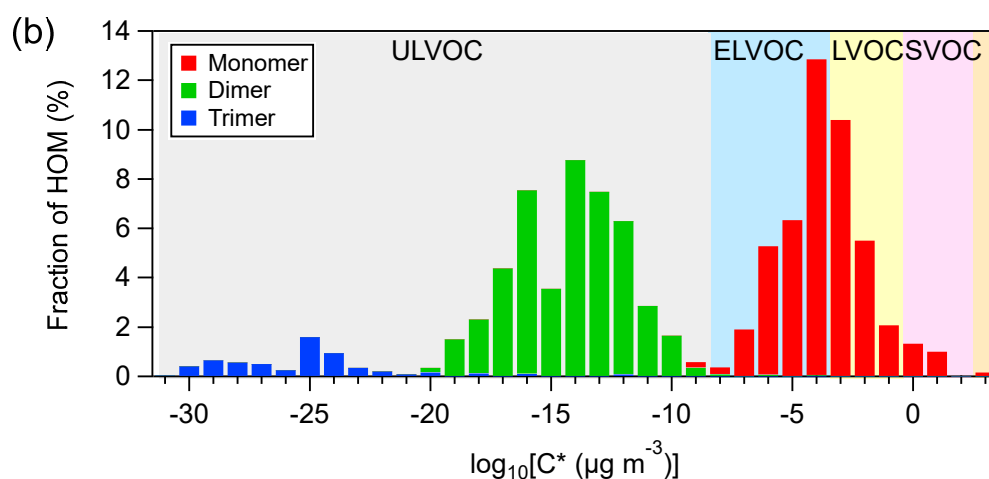
**Response:**

According to scheme S2, we can only assume that  $C_{10}H_{16}O_x$  may be formed via  $NO_3$  oxidation of limonene. We are not aware of pathways to form  $C_{10}H_{14}O_x$  via  $NO_3$  oxidation of limonene, to our knowledge.

14. Add ULVOC in Fig. 7, and explain why dimer cannot trigger NPF?

**Response:**

Accepted. We have added ULVOC in Fig. 7b (also shown below).



In this study, NPF was observed as well as HOM trimers. In contrast, in a previous study investigating the  $NO_3$ -initiated oxidation of  $\beta$ -pinene also conducted in the SAPHIR chamber under similar conditions, barely NPF was observed (Shen et al., 2021), as we discussed in our original manuscript. Also in that study, no trimers were observed. Therefore, NPF in our study was more likely attributed to HOM trimers since they have the strongest potential of initiating nucleation due to their much lower volatility compared to dimers. We did not intend to state that dimers cannot trigger nucleation. Under the same conditions, trimers are more likely to trigger NPF than dimers. In the revised manuscript, we have further clarified this point as follows. “Therefore, NPF in the current study can more likely be attributed to HOM trimers since they have the strongest potential of initiating nucleation, although we cannot rule out some contribution of dimers in the NPF.”



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