REVIEWER COMMENT #2:

The paper by Xie et al. investigates the SOA formed from aPinene and dodecane, alone, and then in mixtures. The radical budget was altered by addition of CO, which increased the HO2 concentrations. The experiments utilized the FIGAERO-CIMS to determine the chemical composition of the SOA formed under the different conditions. The authors present their results clearly, and there are some questions about determination of OH concentration and their ability to achieve iso-reactivity. Though, there are serious questions about the interpretation of their data with respect to radical chemistry. The discussion then attempts to relate the chemical composition observed in the SOA to understand the radical chemistry taking place within the chamber. The discussion of the radical chemistry suffers greatly by only considering RO2 + RO2 chemistry. The authors state that the formation of specific molecules exclusively forms via RO2 + RO2 reactions, when they do not consider other radical pathways (e.g. alkoxy radicals). The other aspects of the paper are relatively well put together, but the leg on which the paper stands is being able to connect their FIGAERO-MS data to the radical chemistry in the chamber. At the moment, I don't see that clear connection because of my concerns about the radical chemistry discussion.

Major Comments:
Lines 539 – 562: The discussion here focuses purely on the RO2 + RO2 reaction pathway, and does not present a holistic understanding the radical pathways present in the reactions of α-pinene + O_3 or OH. This involves the alkoxy radical pathway, which is important part of both the RO2 + RO2 and RO2 + HO2 reaction schemes. This limitation is serious with this paper specifically because on lines 551-553 the authors state that the C10H14Ox can only be formed via RO2 + RO2. Molteni et al (2019) presents clear pathways to the same proposed products that do not invoke RO2 + RO2 (see R2 and R3a/R3B + R5). Because of the weight on these specific molecules (C10H14Ox) and their corresponding products from dodecane being used as the specific proof of the change of the RO2 + RO2 radical reaction pathways, it is crucial for the authors to change their discussion.
The discussion begins to diverge on lines 525-537: I do not understand what the authors mean by "fragment derived RO2 radicals". My understanding of fragmentation is associated with the alkoxy radical pathway. (Molteni et al. 2019).
More related with the gas-phase reaction pathways:
Section 4.3.1: It appears that the discussion here focuses on RO2 reactions, with the 3 pathways being RO2 + RO2, RO2 + HO2, RO2 + NO, or RO2 (autoxidation). What do the authors expect for the lifetimes of RO2 radicals in the chamber for the different experiments toward the 4 different pathways? (or 3 different pathways if it is not possible to discuss autoxidation) The general increase in N-containing products is surprising in the CO containing experiments. Lines 517-520: What would specifically cause the increase in the RO2 + NO pathway? It seems counter intuitive based on the lower NO levels when CO is present.
Considering the FIGAERO-CIMS can result in the degradation of molecules on the filter, here the authors only present molecular formula measurements (D'Ambro et al, 2017). Do the authors believe there is any serious degradation taking place? No thermograms have been presented, so it is difficult to understand if these are likely intact molecules or fragments.
Minor comments:
Lines 50-62: There is also rich literature about mixtures of VOCs and their impact on new particle formation.
Line 71-73: I understand this is a statement from the Baker paper, but it is simply not true as it stands. The SOA yield of a mixture with a dominant RO2 + RO2 pathway is the SOA yield for those specific conditions. The unspoken aspect of this sentence is that the HO2/RO2 ratio is not environmentally relevant for RO2 + RO2 dominant studies, meaning using a RO2 + RO2 dominant yield when the reality is that the RO2 + HO2 pathway is dominant would create an overestimate of the yields in whatever model you choose to use.
Lines 108 – 110: What is the total spectrum of UV light look like? What is the jNO2? Who is the supplier for the UVC lamp? (since the lights are slightly different with the addition of the 254nm lights compared to the Shao et al. publication)
Line 110-112: were the injections performed with a syringe?
Lines 122 – 125: What was the order of the seed injection and humidification? At the moment it is unclear to me what the phase state of the seed is.

Lines 169-181: Is it wise to heat the PTFE filter over 260 °C? There can be degradation of PTFE and the release of fumes from the filter above that temperature. (Sajid et al. 2017)
Section 2.2: how was OH radical concentration determined in Figure S5? I don't see something in the methods section that describes this, and with the presence of O3 does this complicate the determination of O3 when using aPinene as an OH tracer? I see this is mentioned briefly in section 4.1, but it warrants a clear explanation in the methods section. Since this is a batch mode experiment, how does dilution in the chamber impact the depletion of CO? Why is dodecane not included in mixture of Figure S5?
Section 2.3.2: Is the VOCUS run with a GC column? If so please provide the relevant details. I suspect that there is a GC column because of the mention of a chromatography cycle on line 198.
Section 2, what types of blank measurements were performed with the chamber?
Lines 202-204: how did you verify that the injected concentrations are what you think they were?
Lines 204-205, what fragments were used with this method?
Lines 205 – 206: were calibration performed similar to Figure S2 to verify the robustness of using $C_{10}H_{21}^+$
Section 2.3.3: was a dryer used with the AMS? If not how was it verified that the collection efficiency was the same between the experiment and the calibration? I ask because there was likely different RH conditions between the experiment and the calibration.
Figure 1: because of the presence of O3 what is the difference in the OH vs O3 reactivity in the different experiments? The caption should provide information about what fragments mean. Also, how does the OH produced by aPinene ozonolysis impact the iso-reactivity calculations?
(Continuing with Figure S12) In Figure S12, it is not clear if each bar corresponds to the integrated OH/O3 reactivity or is it for that specific unit time? The y-axis label should be changed, at the moment it appears to indicate a ratio of OH / O3, which isn't what the figure is showing.
Line 465-467: This doesn't appear to be true for the dodecane case because the OH never 'recovered'.
Line 470 – 475: I do not understand this discussion. It would appear to be true at face value if isoreactivity was achieved, but it clearly wasn't perfectly achieved in Figure S5. So aren't the changes in OH concentrations purely able to describe these results?
Figure 5 and section 4.2: I am a bit confused by the purported ~50% difference in the yield with vs. without CO for aPinene. Based on Figure 5 (left panel) the yield should be effectively the same with vs. without CO. Can the authors comment on the apparent discrepancy in the text and Table 1 with the Figure?
Lines 543 -545: the way the percentages are talked about are misleading. Perhaps the authors should talk about the percentage reduction of specific molecular cases e.g. 2% reduction for $C_{10}H_{14}O_x$ is a reduction from ~11% à 9% (Figure 6), which is a reduction of ~20%

References:

Sajid, M., Ilyas, M. PTFE-coated non-stick cookware and toxicity concerns: a perspective. Environ Sci Pollut Res 24, 23436–23440 (2017). https://doi.org/10.1007/s11356-017-0095-y

Ugo Molteni, Mario Simon, Martin Heinritzi, Christopher R. Hoyle, Anne-Kathrin Bernhammer, Federico Bianchi, Martin Breitenlechner, Sophia Brilke, António Dias, Jonathan Duplissy, Carla Frege, Hamish Gordon, Claudia Heyn, Tuija Jokinen, Andreas Kürten, Katrianne Lehtipalo, Vladimir Makhmutov, Tuukka Petäjä, Simone M. Pieber, Arnaud P. Praplan, Siegfried Schobesberger, Gerhard Steiner, Yuri Stozhkov, António Tomé, Jasmin Tröstl, Andrea C. Wagner, Robert Wagner, Christina Williamson, Chao Yan, Urs Baltensperger, Joachim Curtius, Neil M. Donahue, Armin Hansel, Jasper Kirkby, Markku Kulmala, Douglas R. Worsnop, and Josef Dommen, ACS Earth and Space Chemistry 2019 3 (5), 873-883, DOI: 10.1021/acsearthspacechem.9b00035

D'Ambro, E. L., Lee, B. H., Liu, J., Shilling, J. E., Gaston, C. J., Lopez-Hilfiker, F. D., Schobesberger, S., Zaveri, R. A., Mohr, C., Lutz, A., Zhang, Z., Gold, A., Surratt, J. D., Rivera-Rios, J. C., Keutsch, F. N., and Thornton, J. A.: Molecular composition and volatility of isoprene photochemical oxidation secondary organic aerosol under low- and high-NOx conditions, Atmos. Chem. Phys., 17, 159–174, https://doi.org/10.5194/acp-17-159-2017, 2017.

ANSWER TO REVIEWER #2:

We would like to sincerely thank the referee for carefully reviewing our manuscript and for the constructive feedback provided. The reviewer's comments are presented in **bold blue**, the authors' responses in black, any revised manuscript text is shown in *italicised red font,* and unchanged original text is shown in *italicised black font*.

In addition to the revisions made in response to the reviewers' comments, several further changes were made to improve the overall readability of the manuscript. These changes are listed at the end of the response to Reviewer 1.

Major Comments:

Lines 539 – 562: The discussion here focuses purely on the RO2 + RO2 reaction pathway, and does not present a holistic understanding the radical pathways present in the reactions of α-pinene + O_3 or OH. This involves the alkoxy radical pathway, which is important part of both the RO2 + RO2 and RO2 + HO2 reaction schemes. This limitation is serious with this paper specifically because on lines 551-553 the authors state that the C10H14Ox can only be formed via RO2 + RO2. Molteni et al (2019) presents clear pathways to the same proposed products that do not invoke RO2 + RO2 (see R2 and R3a/R3B + R5). Because of the weight on these specific molecules (C10H14Ox) and their corresponding products from dodecane being used as the specific proof of the change of the RO2 + RO2 radical reaction pathways, it is crucial for the authors to change their discussion.

Thanks to the reviewer for raising these crucial points.

We acknowledge that the original manuscript did not adequately discuss alkoxy radical pathways, unimolecular termination, or ozonolysis products, all of which potentially contribute to the formation $C_{10}H_{14}O_n$ and $C_{12}H_{24}O_n$ carbonyls. Here, we provide a detailed discussion of these reaction pathways.

(1) Carbonyls by unimolecular termination channel
$RO_2 \rightarrow QOOH$; $QOOH + O_2 \rightarrow O_2QOOH$; $O_2QOOH \rightarrow HO_2Q{=}O + OH$

In principle, RO_2 radicals formed from α-pinene oxidation ($C_{10}H_{15}O_x$) can produce $C_{10}H_{14}O_n$ carbonyls through such a series of isomerisation, oxidation, and unimolecular decomposition reactions. Similarly, RO_2 radicals derived from *n*-dodecane ($C_{12}H_{25}O_x$) can lead to the formation of $C_{12}H_{24}O_n$ carbonyls.

However, under ambient-temperature conditions and in the presence of NO_x, unimolecular termination pathways are not expected to be dominant. Goldman et al. (2021) showed that, at a pressure of 0.5 bar, temperatures below 300 K, and NO concentrations ranging from 1 ppb to 1 ppm, reactions of n-propyl and γ-isobutanol RO_2 radicals are dominated by RO radical formation, and that increasing NO concentrations shift the onset of unimolecular termination to higher temperatures. On this basis, we assume that the contribution of carbonyl compounds formed via unimolecular termination pathways is negligible in this study.

(2) Alkoxy-O_2 channel
$RO + O_2 \rightarrow R'CHO + HO_2$

RO radicals derived from $C_{10}H_{15}O_x$ can form $C_{10}H_{14}O_n$ carbonyls via this pathway, and RO radicals derived from $C_{12}H_{25}O_x$ yield $C_{12}H_{24}O_n$ carbonyls.

Previous studies have shown that C-C bond scission of RO radicals derived from α-pinene has a very low energy barrier, with reaction rates far exceeding those of reactions with O_2 (Dibble, 2001). For linear RO radicals formed from alkanes, isomerisation generally dominates over reactions with O_2 and unimolecular decomposition. For

example, for 2-pentoxy and 2-hexoxy radicals, isomerisation ≫ reaction with O_2 ≈ decomposition (Ziemann and Atkinson, 2012). On this basis, the contribution of the RO + O_2 pathway is expected to be minor and is therefore not explicitly considered in this study.

(3) Ozonolysis
$C_{10}H_{15}O_x$ RO_2 radicals originating from a-pinene ozonolysis form $C_{10}H_{14}O_n$ carbonyls exclusively via RO_2 + RO_2 termination pathways. whereas $C_{10}H_{16}O_n$ products can originate from both RO_2 + HO_2 and RO_2 + RO_2 reactions. Consequently, variations in the relative abundance of $C_{10}H_{14}O_n$ products can be used as an indicator of changes in the RO_2 + RO_2 pathway.

Overall, under certain conditions, alkoxy-O_2 reactions or unimolecular termination channels may contribute with substantial branching ratios. However, in this study, their contributions are expected to be minor. These channels are discussed in the newly added Section 2.1, as outlined in response to reviewer 1.

Line 162-169:
Theoretically, $C_{10}H_{14}O_n$ and $C_{12}H_{25}O_n$ carbonyls can be formed via multiple pathways, including RO_2 + RO_2 reactions (R2), unimolecular termination of RO_2 radicals (R6), and reaction of RO radicals with O_2 (R11). However, previous studies have demonstrated that, under ambient-temperature conditions and in the presence of NO_x, unimolecular termination pathways are not expected to be dominant in RO_2 chemistry (Goldman et al., 2021; Goss et al., 2025). In addition, RO radicals derived from α-pinene generally favour fragmentation owing to the low energy barrier for C-C bond scission (Dibble, 2001). For linear RO radicals formed from long-chain alkanes, isomerisation dominates over reactions with O_2 (Atkinson, 2007; Ziemann and Atkinson, 2012). On this basis, both unimolecular termination and RO + O_2 reactions are expected to make only minor contributions and are therefore not explicitly considered in this study.

The discussion begins to diverge on lines 525-537: I do not understand what the authors mean by "fragment derived RO2 radicals". My understanding of fragmentation is associated with the alkoxy radical pathway. (Molteni et al. 2019).

We thank the reviewer for pointing this out and apologise for the ambiguity in the original manuscript.

We have removed the original statement and clarified the origin of these species in the revised manuscript. The formation of RO_2 radicals containing fewer than 10 carbon atoms necessarily involves fragmentation of RO radicals; therefore, these species are more appropriately described as fragmented RO_2 radicals (Kang et al., 2025).

Line 539-551:
AMS measurements showed a decrease in SOA particle mass concentrations in the presence of CO (Fig. 1c). Besides OH scavenging, another important factor is that CO introduces competition between RO_2 + RO_2 and RO_2 + HO_2 reactions, thereby reducing the formation of accretion products (Baker et al., 2024; McFiggans et al., 2019; Peräkylä et al., 2023). Despite this reduction, CO did not significantly alter the overall fraction of accretion products. However, the relative contribution of C_{16}–C_{24} species decreased (Fig. 2c and 4c), accompanied by an increase in C_{11}–C_{15} species in the α-pinene system and C_{13}–C_{14} species in the n-dodecane system. ***Accretion products with lower carbon numbers are expected to form via pathways that involve fragmentation of RO radicals*** *(Kang et al., 2025), and their increased relative contribution is consistent with the elevated fraction of fragment products discussed above. In contrast, longer-chain accretion products are more likely to arise from RO_2 + RO_2 reactions involving non-fragmented C_{10}/C_{12} RO_2 radicals, including reactions between* ***non-fragmented RO_2 radicals and fragmented RO_2 radicals ($<C_{10}$)****, or between two non-fragmented RO_2 radicals, yielding C_{20} and C_{24} accretion products in the α-pinene and n-dodecane systems, respectively. Combined with the reduced fractions of $C_{10}H_{14}O_n$ and $C_{12}H_{24}O_n$ families (Fig. 3), these observations indicate that CO preferentially suppressed RO_2 + RO_2 chemistry, particularly pathways forming longer-chain accretion products.*

More related with the gas-phase reaction pathways:

Section 4.3.1: It appears that the discussion here focuses on RO2 reactions, with the 3 pathways being RO2 + RO2, RO2 + HO2, RO2 + NO, or RO2 (autoxidation). What do the authors expect for the lifetimes of RO2 radicals in the chamber for the different experiments toward the 4 different pathways? (or 3 different pathways if it is not possible to discuss autoxidation) The general increase in N-containing products is surprising in the CO containing experiments. Lines 517-520: What would specifically cause the increase in the RO2 + NO pathway? It seems counter intuitive based on the lower NO levels when CO is present.

We respond to the reviewer's comments point by point below.

(1) What do the authors expect for the lifetimes of RO2 radicals in the chamber for the different experiments toward the 4 different pathways? (or 3 different pathways if it is not possible to discuss autoxidation)

Compared with experiments conducted without CO, the presence of CO is expected to modify the RO_2 fate as follows.
1. CO reacts with OH to form HO_2, increasing HO_2 concentrations.
2. Elevated HO_2 enhances the HO_2 + NO reaction, resulting in lower NO concentrations.
3. HO_2 competes with RO_2 and NO for reaction with RO_2, and together with the reduced NO concentrations decreases the relative importance of RO_2 + RO_2 and RO_2 + NO pathways.

Consequently, in the presence of CO, the lifetime of RO_2 radicals with respect to reactions with NO and other RO_2 radicals is expected to increase, whereas their lifetime towards reaction with HO_2 is expected to decrease.

(2) The general increase in N-containing products is surprising in the CO containing experiments. Lines 517-520: What would specifically cause the increase in the RO2 + NO pathway? It seems counter intuitive based on the lower NO levels when CO is present.

We thank the reviewer for pointing this out.

In the single-precursor systems, the presence of CO did not enhance the RO_2 + NO pathway. Instead, only its **relative contribution** increased, while the absolute abundance was significantly suppressed. Evidence for this behaviour is provided by both AMS and CIMS measurements. (Figure S13 has been added to the Supplementary Information)

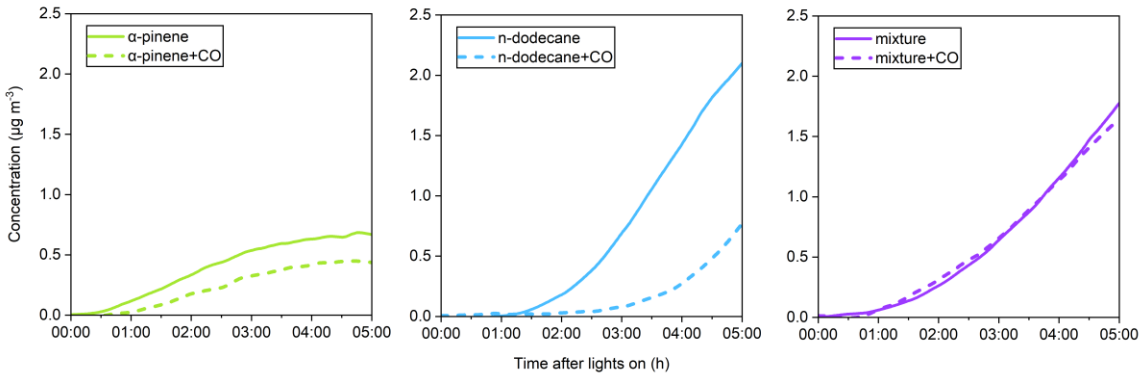
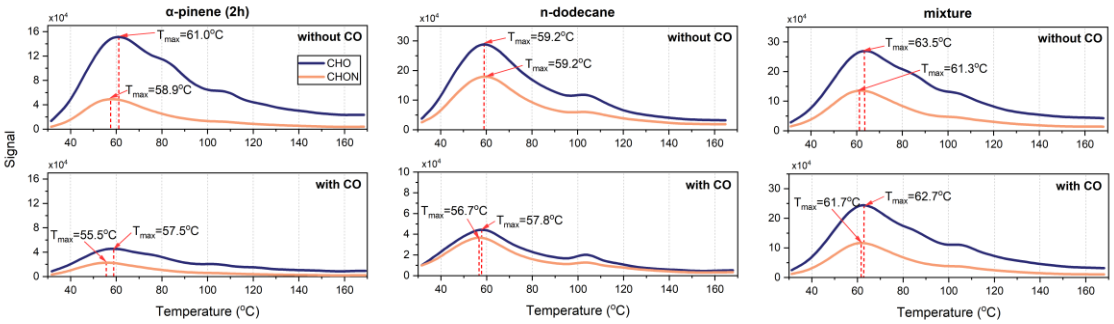


Figure S13: The concentration of organic nitrates estimated from AMS measurements.

In the single-precursor systems, the presence of CO led to a pronounced decrease in organic nitrate concentrations, suggesting a reduced likelihood of RO_2 reacting with NO. The relative contribution of CHON and fragment products increased in the presence of CO (Fig. 2 and 4). CHON products are formed through the RO_2 + NO → $RONO_2$ channel, while fragment species originate from the fragmentation of RO radicals. Owing to the rapid reaction of RO_2 with NO and the high branching toward RO formation, reactions of RO_2 with NO represent an

important source of RO radicals under NO_x conditions. These observations therefore indicates that, in the presence of CO, the contribution of the RO_2 + NO reactions were decreased, but to a lesser extent than the competing RO_2 termination pathways.

Consequently, the overall decrease in CHO mass exceeded the reduction in CHON mass, resulting in an apparent increase in the **normalised** contribution of CHON compounds.



Thermograms of CHO and CHON compounds.

Moreover, the thermograms derived from FIGAERO-CIMS showed that, in the single-precursor experiments, CHON compounds exhibited slightly lower T_{max} values in the presence of CO than in its absence, indicating higher volatility during thermal desorption.

In the revised manuscript, this behaviour is interpreted by considering both the concentrations of organic nitrates estimated from AMS measurements and the changes in the fraction of CHON and fragment products.

Line 525-537:

Organic nitrate concentrations were estimated from AMS measurements using the method described by Kiendler-Scharr et al. (2016). ***The results show that, in the single-precursor systems, the presence of CO led to a pronounced reduction in organic nitrate concentrations*** *(Fig. S13). This reduction can be attributed to two main factors. First, CO competes with SOA precursors for available OH (Fig 1b and S6). Second, CO enhances HO_2 formation, introducing an additional competing sink for RO_2 and thereby altering RO_2 reaction branching. In addition, lower NO concentrations were observed in the presence of CO (Fig. S5), consistent with enhanced conversion of NO to NO_2 by HO_2. The increase in HO_2 and decrease in NO reduces the likelihood of RO_2 reacting with NO. Despite this absolute reduction, FIGAERO-CIMS results showed that* ***the relative contribution of CHON and fragment products increased in the presence of CO*** *(Fig. 2 and 4). CHON products are formed through the RO_2 + NO → $RONO_2$ channel, while fragment species originate from the fragmentation of RO radicals (Atkinson, 2000; Ziemann and Atkinson, 2012). Owing to the rapid reaction of RO_2 with NO and the high branching toward RO formation, reactions of RO_2 with NO represent an important source of RO radicals under NO_x conditions (Orlando et al., 2003; Ziemann and Atkinson, 2012).* ***These observations therefore indicates that, in the presence of CO, the contribution of the RO_2 + NO reactions were decreased, but to a lesser extent than the competing RO_2 termination pathways.***

Line 553-555:

Overall, in the single-precursor systems, CO reduced the contributions of both RO_2 + RO_2 and RO_2 + NO reactions. ***However, reactions of RO_2 with NO decreased to a lesser extent than competing RO_2 termination pathways****, and the reduction in RO_2 + RO_2 termination was more pronounced for longer-chain accretion products than for shorter-chain ones.*

Considering the FIGAERO-CIMS can result in the degradation of molecules on the filter, here the authors only present molecular formula measurements (D'Ambro et al, 2017). Do the authors believe there is any

serious degradation taking place? No thermograms have been presented, so it is difficult to understand if these are likely intact molecules or fragments.

We acknowledge that minor thermal decomposition is present in the FIGAERO-CIMS measurements in this study. Several compounds with relatively low carbon numbers were found to exhibit comparatively high $\overline{OSc}$ values and elevated T_{max}. Nevertheless, these species together accounted for less than 10% of the total signal, indicating that the impact of thermal decomposition on the chemical composition was limited. (Figure S2 has been added to the Supplementary Information)

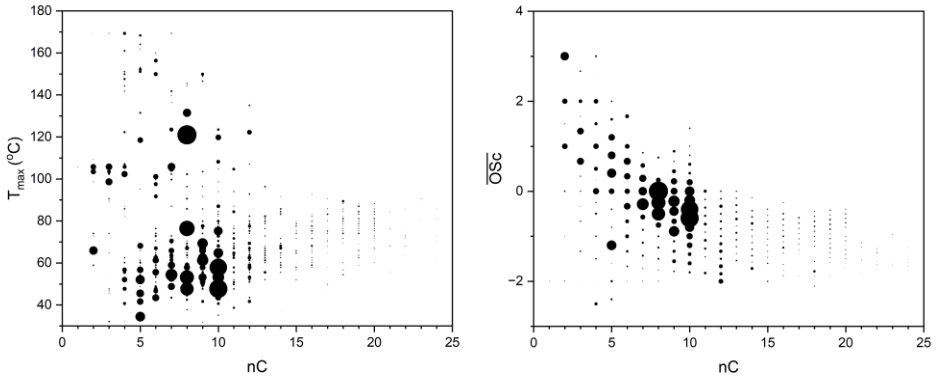


Figure S2. (Left panel) Maximum desorption temperature (T_{max}) against carbon number (nC) and (right panel) average carbon oxidation states ($\overline{OSc}$) against nC for all the particle-phase products for α-Pinene system.

Line 262-266:

Additional uncertainties may arise from the thermal decomposition in the FIGAERO. As shown in Fig. S2, several compounds with relatively low carbon numbers exhibit comparatively high average carbon oxidation state ($\overline{OSc}$) values and elevated maximum desorption temperature (T_{max}). Nevertheless, these species together accounted for less than 10 % of the total signal, indicating that the impact of thermal decomposition on the chemical composition was limited.

Minor comments:

Lines 50-62: There is also rich literature about mixtures of VOCs and their impact on new particle formation.

We thank the reviewer for this suggestion.

We agree that many studies have investigated the SOA formation from the multi-precursor systems. The original sentence has been revised to clarify that many chamber studies are conducted under simplified conditions or with a single SOA precursor to address specific research questions, and corresponding modifications have also been made in the Abstract. We have also added a statement noting that an increasing number of studies have focused on multi-precursor systems.

Line 13-18:

The ambient atmosphere comprises a complex mixture of biogenic and anthropogenic emissions. Atmospheric simulation chambers are powerful tools for investigating atmospheric processes and form the basis for model parameterisations. Ensuring the atmospheric relevance of experimental conditions is crucial for understanding and predicting the impacts of secondary organic aerosols (SOA) on air quality and climate. However, many chamber studies are conducted under simplified conditions or with a single SOA precursor to address specific research questions, which may limit their applicability to real-world scenarios.

Line 43-49:

The ambient atmosphere comprises a complex mixture of biogenic and anthropogenic emissions, including a wide range of gas-phase organic compounds and inorganic trace gases (Gu et al., 2021; Guenther et al., 1995). Field measurements have provided evidence that anthropogenic emissions can modulate SOA formed from biogenic precursors (Budisulistiorini et al., 2015; Shilling et al., 2013; Xu et al., 2015). However, many laboratory experiments are conducted under simplified conditions or with a single SOA precursor to address specific research questions, which may introduce uncertainties when extrapolating these results to atmospheric models (Kenagy et al., 2024; Shrivastava et al., 2017; Tsigaridis et al., 2014).

Line 61:

An increasing number of studies have focused on mixtures of multiple precursors.

Line 71-73: I understand this is a statement from the Baker paper, but it is simply not true as it stands. The SOA yield of a mixture with a dominant RO2 + RO2 pathway is the SOA yield for those specific conditions. The unspoken aspect of this sentence is that the HO2/RO2 ratio is not environmentally relevant for RO2 + RO2 dominant studies, meaning using a RO2 + RO2 dominant yield when the reality is that the RO2 + HO2 pathway is dominant would create an overestimate of the yields in whatever model you choose to use.

We thank the reviewer for this suggestion.

We have added a discussion comparing RO_2 reaction pathways under atmospherically relevant and laboratory conditions, emphasising that the elevated RO_2 levels typically present in laboratory experiments can lead to an overestimation of SOA particle mass yields.

Line 77-88:

In laboratory experiments, SOA precursor concentrations are often higher than those typically observed in the ambient atmosphere for practical reasons (Ziemann and Atkinson, 2012). This can lead to substantially elevated RO_2 radical concentrations relative to atmospheric levels, favouring RO_2 + RO_2 reactions over RO_2 + HO_2 reactions (Ziemann and Atkinson, 2012). The former forms accretion products, which may have extremely low volatility and are expected to contribute to new particle formation, potentially leading to an overestimation of SOA particle mass yields *(Kenagy et al., 2024; Peräkylä et al., 2023; Ziemann and Atkinson, 2012). The presence of CO can directly consume OH and produce HO_2 radicals, thereby shifting the HO_2/RO_2 ratio and increasing the importance of the RO_2 termination via HO_2 (Lu and Khalil, 1993). Previous studies have quantified the effect of CO on SOA production. McFiggans et al. (2019) showed that CO suppressed α-pinene dimer (containing 17 to 20 carbon atoms) formation by a factor of two, while the amounts of HOMs were suppressed by factors of 4 to 5.* ***Baker et al. (2024) further demonstrated that, under constant OH conditions, the addition of CO increased the HO_2/RO_2 ratio from approximately 1/100 to about 1/1, leading to a ~ 60 % reduction in the abundance of HOM-accretion products and a ~ 30 % decrease in the SOA formation potential of HOMs***.

Lines 108 – 110: What is the total spectrum of UV light look like? What is the jNO2? Who is the supplier for the UVC lamp? (since the lights are slightly different with the addition of the 254nm lights compared to the Shao et al. publication)

We apologise that the spectrum of the UVC lamp was not measured in this study.

The actinic flux spectrum of the built-in light sources (two xenon arc lamps and a series of halogen lamps) in the MAC is shown in Shao et al. (2022). An additional UVC lamp was installed to promote OH radical production, with more than 90 % of its length masked to prevent excessive irradiation.

The J_{NO_2} was 1.38×10^{-3} s^{-1}.

The supplier for the UVC lamp is Philips (TUV 130W XPT SE UNP/20).

Line 179-184:

The irradiation source, consisting of two xenon arc lamps (XBO 6000W/HSLA OFR, Osram) and a series of halogen lamps (50W/4700K MR16, Solux), is mounted inside the chamber and generates irradiation over the wavelength range of 290–800 nm to mimic the atmospheric radiation spectrum. The corresponding actinic flux spectrum is presented in Shao et al. (2022). The photolysis rate of NO_2 (J_{NO_2}) was 1.38×10^{-3} s^{-1}. To promote OH radical production, an additional UVC lamp (TUV 130W XPT SE UNP/20, Philips) was installed, with more than 90 % of its length masked to prevent excessive irradiation.

Line 110-112: were the injections performed with a syringe?

Yes, the injections were performed using a syringe. The required volume of liquid precursor was calculated based on its density, the target concentration, and the chamber volume. The liquid sample was then injected into a pre-heated VOC bulb using a syringe and subsequently introduced into the chamber by flushing with N_2.

Line 184-186:

*The liquid precursors (α-pinene, analytical standard, Sigma-Aldrich; n-dodecane, anhydrous, ≥ 99.0 %, Sigma-Aldrich) were initially injected **via syringe** into a heated glass bulb to facilitate vaporisation, after which the vapours were carried into the chamber by electronic capture device-grade nitrogen (ECD N_2).*

Lines 122 – 125: What was the order of the seed injection and humidification? At the moment it is unclear to me what the phase state of the seed is.

Compressed air was passed through the humidifier when flushing the seed into the chamber, hence ensuring the deliquescence of the seed as they were generated.

Line 189-193:

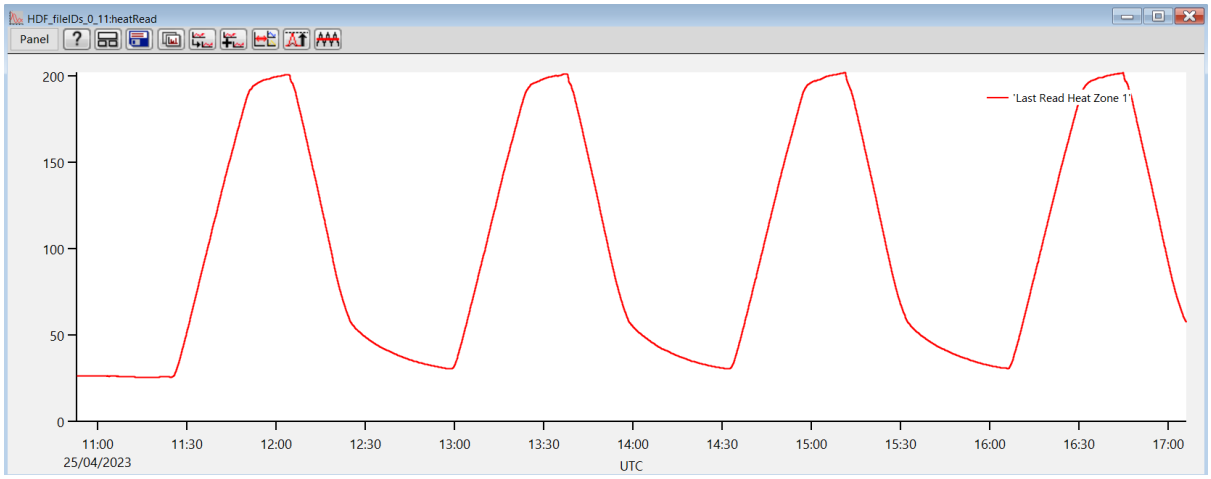
*Seed particles with a mass concentration of 40.2 ± 8.0 μg m^{-3} were generated by nebulising aqueous ammonium sulfate solutions ($(NH_4)_2SO_4$, ACS reagent, ≥ 99.0 %, Sigma-Aldrich) using an aerosol generator (ATM 230, Topas). **During seed injection, the carrier air was passed through the humidifier, ensuring the deliquescence of the seeds as they were generated**. These particles provided a condensation surface for the oxidation products, thereby reducing wall losses and suppressing nucleation (Nah et al., 2017).*

Lines 169-181: Is it wise to heat the PTFE filter over 260 °C? There can be degradation of PTFE and the release of fumes from the filter above that temperature. (Sajid et al. 2017)

We thank the reviewer for pointing this out.

The temperature of 310 °C refers to the set value of the heating unit. As shown in the figure below, the actual temperature experienced by the PTFE filter did not exceed 200 °C throughout the desorption process.

We have corrected the temperature reported in the manuscript and added a clarification that the filters were pre-heated to 200 °C to remove potential contaminants.



Line 240-247:

(i) 30 min of gas-phase sampling and simultaneous particle collection onto a PTFE filter (2.0 µm pore size, Zefluor; filters were pre-heated to 200 °C to remove potential contaminants) both at 1 L min^{-1}. During this step, the instrument was flushed with N_2 for 0.5 min every 4.5 min to obtain the gas-phase instrument background signal.

(ii) 25 min of temperature-programmed thermal desorption of the collected particles, with the temperature ramped from ambient to 200 °C.

(iii) 15 min of isothermal soaking at 200 °C.

(iv) 20 min of cooling from 200 °C to ambient temperature.

(v) 2 min of N_2 flushing to clean the instrument.

Section 2.2: how was OH radical concentration determined in Figure S5? I don't see something in the methods section that describes this, and with the presence of O3 does this complicate the determination of O3 when using aPinene as an OH tracer? I see this is mentioned briefly in section 4.1, but it warrants a clear explanation in the methods section. Since this is a batch mode experiment, how does dilution in the chamber impact the depletion of CO? Why is dodecane not included in mixture of Figure S5?

We respond to the reviewer's comments point by point below.

(1) How was OH radical concentration determined in Figure S5?

OH concentrations were estimated from the evolution of O_3 and the consumption of precursors, or alternatively, from the depletion of CO.

Based on the consumption of α-pinene or n-dodecane:

$$[OH] = \frac{\frac{[VOC]_i - [VOC]_{i+1}}{\Delta t} - k_{VOC+O_3}[VOC]_i[O_3]}{k_{VOC+OH}[VOC]_i}$$

Based on the decay of CO:

$$[OH] = \frac{[CO]_i - [CO]_{i+1}}{k_{CO+OH}[CO]_i \Delta t}$$

These equations have been added to the Supplementary Information.

(2) With the presence of O3 does this complicate the determination of O3 when using aPinene as an OH tracer?

No. When deriving OH radical concentrations using α-pinene as a tracer, the loss of α-pinene via ozonolysis was explicitly included in the calculation. As a result, the contribution of O_3 chemistry was accounted for in the calculated OH concentrations.

No action

(3) Since this is a batch mode experiment, how does dilution in the chamber impact the depletion of CO?

The upper and lower frames of the chamber can move freely, allowing the chamber volume to expand or collapse when sample air is extracted. Therefore, dilution does not influence the depletion of CO.

No action

(4) Why is dodecane not included in mixture of Figure S5?

We apologise for the ambiguity in the original figure. As the n-dodecane concentration was not quantified, we preferred to present OH concentrations estimated based on α-pinene and CO for the mixed-precursor system. In the revised figure, we have now additionally included the OH concentrations estimated from the decay of n-dodecane for completeness. The OH concentrations derived from n-dodecane show good agreement with those calculated from α-pinene and CO.

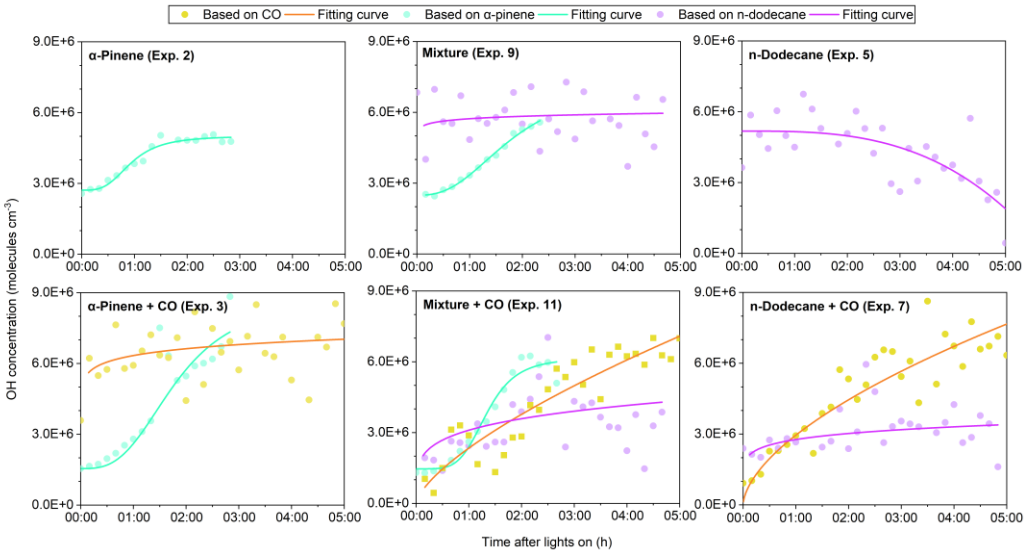


Figure S6: Estimated OH concentrations derived from the decay of precursors or CO. Fitting curves are shown as a visual guide. Data are from representative experiments.

Section 2.3.2: Is the VOCUS run with a GC column? If so please provide the relevant details. I suspect that there is a GC column because of the mention of a chromatography cycle on line 198.

We thank the referee for noting this point. The VOCUS was not operated with a GC column in this study. The reference to a "chromatography cycle" was a wording error and has been corrected to "sampling" in the revised manuscript.

Line 277-278:
Measurements were made on a 5 min cycle, consisting of 4 min of sampling followed by 1 min of instrumental background.

Section 2, what types of blank measurements were performed with the chamber?

Chamber background measurements were conducted weekly. All components (SOA precursors, seed particles, CO, and NO_x) were injected into the chamber under the same experimental conditions as the regular experiments, while the chamber was kept in the dark. CIMS data obtained during these background measurements were subtracted from both the gas- and particle-phase data acquired during the "experiment" phase.

Line 252-255:

To further correct for background species in the chamber, background measurements were conducted weekly. During these measurements, all components (SOA precursors, seed particles, CO, and NO_x) were injected into the chamber under the same conditions as the regular experiments, while the chamber was kept in the dark. Data obtained during these background measurements were subtracted from both the gas- and particle-phase data acquired during the "experiment" phase.

Lines 202-204: how did you verify that the injected concentrations are what you think they were?

The injection approach used in this study is reliable. The required volume of each precursor was calculated based on its density, the target concentration, and the chamber volume. Quantitative measurements of α-pinene confirm the reliability of this approach, as the injected concentrations deviated from the target values by less than 25% in the majority of experiments.

Moreover, this study focuses on the relative differences in SOA yields between the CO-present and CO-absent conditions. Therefore, any minor deviations in the absolute injected concentrations are unlikely to substantially affect the conclusions.

No action

Lines 204-205, what fragments were used with this method?

Fragment ions originating from *n*-dodecane generally exhibit the formula $C_nH_{2n+1}^+$. $C_{10}H_{21}^+$ fragment ion was used to infer the relative consumption of n-dodecane.

Line 281-283:

Therefore, alternative approaches were adopted for its quantification: (i) the initial concentrations were taken as the target values (160 ppb in the single-precursor system and 80 ppb in the mixed-precursor system), and (ii) the relative consumption of n-dodecane was inferred from the temporal evolution of the ***$C_{10}H_{21}^+$*** *fragment ion (Fig. S4).*

Lines 205 – 206: were calibration performed similar to Figure S2 to verify the robustness of using C10H21+

No, calibration was not performed similar to α-pinene because the absence of an n-dodecane calibration standard.

We estimated the expected n-dodecane concentration using the OH concentration derived from CO decay, and this estimate was compared with the measured $C_{10}H_{21}^+$ signal (Fig. S4). As shown in this figure, the predicted and measured values agree well during the first three hours of reaction, whereas deviations occur at later times, likely due to interference from other oxidation products or fragments. The SOA particle mass yields of n-dodecane and the mixture may be overestimated by up to ~30 % in this study. Nevertheless, this uncertainty does not affect the overall trends and relative differences in yields.

We have clarified this limitation in the manuscript.

Line 283-285:

However, contributions from other oxidation products or fragments remain unavoidable, which could result in an overestimation of SOA particle mass yields. Nevertheless, this uncertainty does not affect the overall trends and relative differences in yields.

Section 2.3.3: was a dryer used with the AMS? If not how was it verified that the collection efficiency was the same between the experiment and the calibration? I ask because there was likely different RH conditions between the experiment and the calibration.

Yes, a dryer was installed upstream of the AMS inlet.

No action

Figure 1: because of the presence of O3 what is the difference in the OH vs O3 reactivity in the different experiments? The caption should provide information about what fragments mean. Also, how does the OH produced by aPinene ozonolysis impact the iso-reactivity calculations?

We respond to the reviewer's comments point by point below.

(1) because of the presence of O3 what is the difference in the OH vs O3 reactivity in the different experiments?

The OH and O_3 reactivities are defined by the following equations:

$$\text{OH reactivity } (s^{-1}) = \sum C_{precursor,i} \times k_{OH,i}$$

$$O_3 \text{ reactivity } (s^{-1}) = \sum C_{precursor,i} \times k_{O_3,i}$$

n-Dodecane dose not react with O_3; therefore, its O_3 reactivity is zero. For α-pinene, k_{O_3} (9.6×10^{-17} cm^3 $molecule^{-1}$ s^{-1}) is much smaller than k_{OH} (5.33×10^{-11} cm^3 $molecule^{-1}$ s^{-1}), so its O_3 reactivity is also very low relative to its OH reactivity and can be approximated as zero. Consequently, the discussion here mainly focuses on differences in OH reactivity.

O_3 can influence precursor decay as well as the formation of secondary oxidants, thereby affecting the OH reactivity.

At the beginning of the reaction, owing to the iso-reactivity condition, all systems exhibited comparable reactivity. As the reaction proceeded, differences emerged due to changes in precursor concentrations. Because α-pinene decayed faster in normalised terms than *n*-dodecane, the reactivity over the course of the experiment followed the order: n-dodecane > mixture > a-pinene.

Line 467-477:

Under idealised iso-reactivity conditions, all systems would exhibit comparable initial OH reactivity, and in the mixture each precursor molecule would initially have an equal probability of reacting with OH. ***In practice, however, O_3 also contributed to precursor oxidation, and the differing reactivities of individual precursors towards O_3 can modify the precursor decay and secondary oxidant formation, thereby influencing the reactivity. n-Dodecane was oxidised exclusively by OH radicals.*** *For α-pinene, although OH remained the dominant photochemical sink in this study, the contribution of O_3 to its decay was not negligible. As shown in Fig. S13, the relative contributions of these oxidants evolved over time, with the role of O_3 becoming increasingly significant as the reaction proceeded. In the α-pinene single-precursor system, on average approximately 80 % of α-pinene decay was attributable to OH oxidation, while the remaining ~20 % was driven by ozonolysis. By comparison, the contribution of ozonolysis was slightly higher in the mixed-precursor system. Thus, fully comparable reactivity across different systems was difficult to maintain throughout the reaction when multiple oxidants were present.*

This reflects an inherent limitation of defining iso-reactivity with respect to a single oxidant in multi-oxidant systems.

(2) Also, how does the OH produced by aPinene ozonolysis impact the iso-reactivity calculations?

OH reactivity is defined as the sum of the products of the concentrations of each SOA precursor and their reaction rate coefficients with OH.

The calculation of iso-reactivity does not involve the OH concentration and is therefore not affected by OH produced from α-pinene ozonolysis.

No action

(Continuing with Figure S12) In Figure S12, it is not clear if each bar corresponds to the integrated OH/O3 reactivity or is it for that specific unit time? The y-axis label should be changed, at the moment it appears to indicate a ratio of OH / O3, which isn't what the figure is showing.

We thank the reviewer for this suggestion and apologise for a minor error in the calculation of the fractional contributions shown in the original version of Fig. S11. This has now been corrected in the revised figure (now Fig. S12), and the correction does not affect the overall interpretation of the results.

The pink and purple portions of each bar represent the fractional contributions of ozonolysis and OH oxidation, respectively, to α-pinene decay. These contributions are calculated based on the relative magnitudes of $k_{VOC+O_3}[VOC]_i[O_3]$ and $k_{VOC+OH}[VOC]_i[OH]$.

Owing to differences in the time resolution of the instruments, all values were averaged over 10-min intervals.

The y-axis label has been revised to "Fractional contribution to α-pinene oxidation" to clarify the meaning of the plotted values.

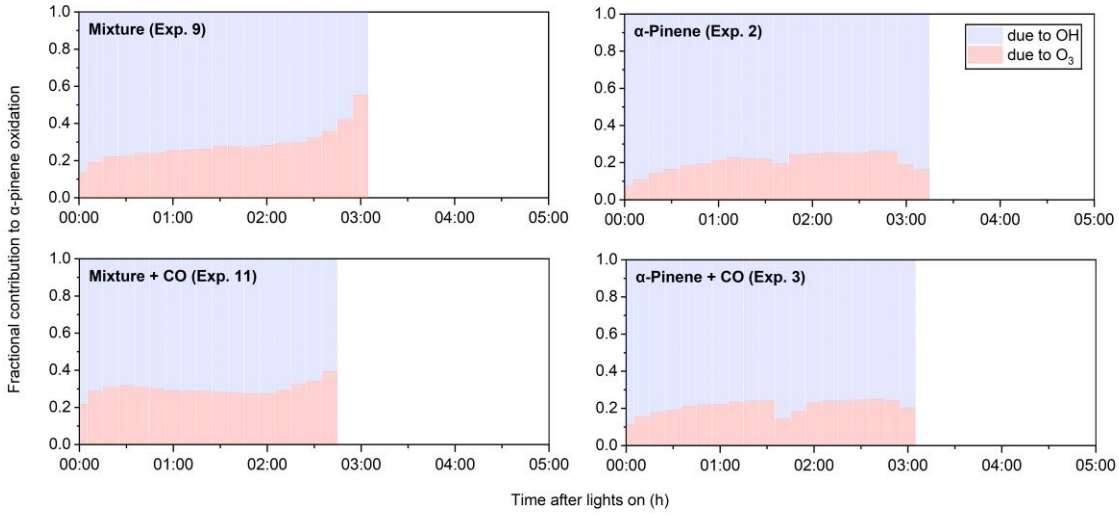


Figure S12: Relative contributions of O_3 and OH to α-pinene oxidation. These contributions are calculated based on the relative magnitudes of $k_{VOC+O_3}[VOC]_i[O_3]$ and $k_{VOC+OH}[VOC]_i[OH]$. Data are from representative experiments.

Line 465-467: This doesn't appear to be true for the dodecane case because the OH never 'recovered'.

We apologise for the ambiguity in the original manuscript.

In the n-dodecane system without CO, the OH concentrations gradually decreased as the reaction proceeded. This decline likely reflects the continuous consumption of OH and the lack of efficient OH regeneration pathways in the absence of CO.

Line 498-514:

The addition of CO further perturbed the photochemical processes, altering both oxidant levels and precursor decay rates. CO can consume OH radicals, preventing their reaction with SOA precursors (McFiggans et al., 2019). Based on the estimated OH concentrations, evidence for this oxidant scavenging effect was observed. During the initial stage of the reaction, CO reduced the OH concentrations by approximately 50 % to around 1.5 × 10^6 molecules cm^{-3} (Fig. S6). However, OH levels gradually recovered as the reaction progressed and eventually reached values comparable to those observed in the absence of CO ***(except for n-dodecane system). In the n-dodecane system without CO, OH concentrations gradually decreased as the reaction proceeded. This decline likely reflects the continuous consumption of OH by n-dodecane and the lack of efficient OH regeneration pathways.*** *By contrast, in both the α-pinene and mixture systems, OH concentrations continued to increase even in the absence of CO, indicating additional OH regeneration processes, such as OH formation during α-pinene ozonolysis.*

Line 470 - 475: I do not understand this discussion. It would appear to be true at face value if isoreactivity was achieved, but it clearly wasn't perfectly achieved in Figure S5. So aren't the changes in OH concentrations purely able to describe these results?

We respond to the reviewer's comments point by point below.

(1) It would appear to be true at face value if isoreactivity was achieved, but it clearly wasn't perfectly achieved in Figure S5

We thank the reviewer for raising this important point.

Iso-reactivity is defined by precursor concentrations and their respective reaction rate coefficients with OH, and is independent of the OH concentration itself. Therefore, the OH concentrations shown in Fig. S5 cannot be used to determine whether iso-reactivity was achieved. In addition, iso-reactivity ensures that different precursors have the same initial potential to react with OH; however, it does not guarantee identical oxidative conditions are maintained throughout the entire experiment.

We have clarified this point in the revised manuscript.

Line 542-546:

Under idealised iso-reactivity conditions, all systems would exhibit comparable initial OH reactivity, and in the mixture each precursor molecule would initially have an equal probability of reacting with OH. In practice, however, O_3 also contributed to precursor oxidation, and the differing reactivities of individual precursors towards O_3 can modify the precursor decay and secondary oxidant formation, thereby influencing the reactivity.

(2) So aren't the changes in OH concentrations purely able to describe these results?

We thank the reviewer for raising this important point.

These results can be described by the changes in OH and O_3 concentration.

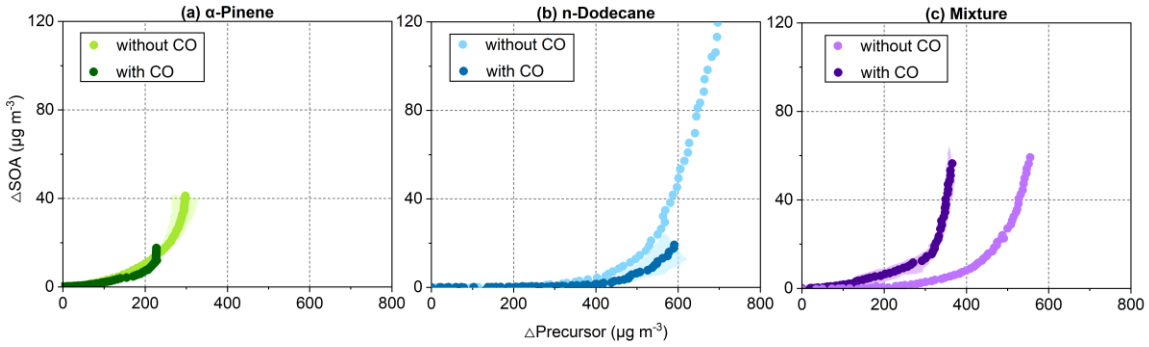
In addition, we have revised our previous statement regarding the OH scavenging effect of CO in the α-pinene system. In the original manuscript, we stated that "the effect of OH scavenging on the decay of α-pinene was

limited." We now clarify that an OH scavenging effect does occur, as evidenced by the reduced α-pinene decay rate during the early stage of the reaction. However, this initial suppression is later compensated by the regeneration of OH and the concurrent increase in O_3 concentration, such that the overall extent of α-pinene consumption is not substantially reduced. Accordingly, we now state that "CO did not significantly affect the overall extent of α-pinene consumption."

Line 509-514:

Variations in oxidant concentrations resulted in changes in VOC decay rates (Fig 1b). In the absence of CO, α-pinene was almost completely consumed within 3 h. When CO was present, its decay was suppressed during the initial stage; however, after approximately 2 h the decay rate accelerated, owing to secondary OH production and elevated O_3 concentrations, such that α-pinene was nevertheless nearly fully consumed within 3 h. ***As a result, CO did not significantly affect the overall extent of α-pinene consumption****. **In contrast, for n-dodecane, CO not only slowed the oxidation rate but also reduced the overall consumption, with a substantial fraction of n-dodecane remaining unreacted by the end of the experiment**.*

Figure 5 and section 4.2: I am a bit confused by the purported ~50% difference in the yield with vs. without CO for aPinene. Based on Figure 5 (left panel) the yield should be effectively the same with vs. without CO. Can the authors comment on the apparent discrepancy in the text and Table 1 with the Figure?



We thank the reviewer for raising this important point and apologise that in the original figure the Δprecursor for α-pinene under CO-absent conditions was slightly shifted towards higher values. This has now been corrected, and the correction does not affect any of the results or conclusions.

The yields reported in the text refer to the <u>overall yield</u>, defined as $\triangle SOA_{max}/\triangle precursor_{max}$.

In the presence of CO, $\triangle SOA_{max}$ was approximately 18 μg m^{-3}, $\triangle precursor_{max}$ was approximately 230 μg m^{-3}, corresponding to an SOA yield of ~ 0.08.
In the absence of CO, $\triangle SOA_{max}$ was approximately 41 μg m^{-3}, $\triangle precursor_{max}$ was approximately 297 μg m^{-3}, corresponding to an SOA yield of ~0.14.

Compared with the CO-present case, the SOA particle mass yield in the absence of CO was therefore reduced by approximately 43%.

To address this potential confusion, we have revised the corresponding descriptions in both the Methodology and the yield discussion.

Line 304-305:

In this study, the SOA particle mass yield refers to the ***<u>overall yield</u>****, calculated from the total SOA formed and the precursor consumed at the end of the experiment.*

Line 589-601:

Figure 6 presents the SOA particle growth curves for each system. ***The slope of the curve represents the incremental SOA particle mass yield at a given stage of precursor consumption, while the final position of the curve reflects the overall yield achieved by the end of the experiment.*** *The induction period is defined as the amount of SOA precursor consumed before SOA particle formation begins (Zhou et al., 2019). Compared with the α-pinene system, the n-dodecane system exhibited a longer induction period, while that of the mixed-precursor system lay in between. In the presence of CO, the induction period was extended in the n-dodecane system but remained largely unchanged in the α-pinene system. Notably, the induction period in the mixture system was shortened in the presence of CO. These behaviours suggest a distinct influence of CO on the SOA particle mass yields across different systems.*

In the single-precursor systems, CO substantially reduced SOA formation, with a stronger effect for n-dodecane than for α-pinene. In the presence of CO, SOA particle mass concentrations and the ***overall yields*** *decreased by 83 % and 79 % for n-dodecane, and by 57 % and 43 % for α-pinene, respectively. In contrast, the mixed-precursor system exhibited only an 8 % decrease in SOA mass concentration, and the* ***overall yield*** *slightly increased, indicating a markedly weaker sensitivity to CO.*

Lines 543 -545: the way the percentages are talked about are misleading. Perhaps the authors should talk about the percentage reduction of specific molecular cases e.g. 2% reduction for C10H14Ox is a reduction from ~11% à 9% (Figure 6), which is a reduction of ~20%

We thank the reviewer for this suggestion. We agree that the original description of the percentages could be misleading.

We have revised the text to describe the fractions under CO-absent and CO-present conditions separately, without explicitly discussing the percentage reduction.

In addition, to improve the flow of the manuscript, these descriptions and Fig. 6 (now Fig. 3) have been moved to the Results section. As the variation in the fraction of $C_{12}H_{22}O_n$ was not relevant to the discussion of the results, the corresponding data has been removed.

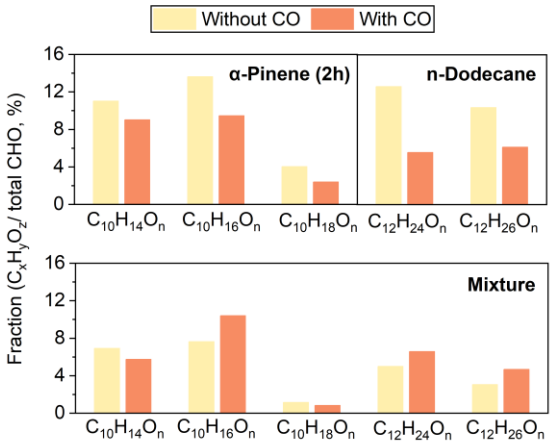


Figure 3: Relative contributions of $C_{10}H_{14}O_n$, $C_{10}H_{16}O_n$, $C_{10}H_{18}O_n$, $C_{12}H_{24}O_n$, and $C_{12}H_{26}O_n$ to the total CHO compounds in the α-pinene, n-dodecane, and mixture systems in the absence and presence of CO.

Line 365-366:

The main RO_2 radicals derived from α-pinene undergo R1 and R2 reactions to form the $C_{10}H_{14}O_n$, $C_{10}H_{16}O_n$, and $C_{10}H_{18}O_n$ families. As shown in Fig. 3, in the absence of CO, these compounds accounted for 11.0 %, 13.6 %, and 4.0 % of the CHO products, decreasing to 9.0 %, 9.5 %, and 2.4 % in the presence of CO.

Line 412-414:

The main RO_2 radicals derived from n-dodecane undergo R1 and R2 reactions to form the $C_{12}H_{24}O_n$ and $C_{12}H_{26}O_n$ families. As shown in Fig. 3, in the absence of CO, these compounds accounted for 12.6 % and 10.4 % of the CHO products, decreasing to 5.6 % and 6.1 % in the presence of CO.

Line 452-455:

The bottom panel of Fig. 3 shows the relative contributions of $C_{10}H_{14}O_n$, $C_{10}H_{16}O_n$, $C_{10}H_{18}O_n$, $C_{12}H_{24}O_n$, and $C_{12}H_{26}O_n$ to the CHO products in the mixture. In the presence of CO, the fractions of $C_{10}H_{14}O_n$ and $C_{10}H_{18}O_n$ decreased from 6.9% and 1.2% to 5.5% and 0.8%, respectively, whereas the fractions of $C_{10}H_{16}O_n$, $C_{12}H_{24}O_n$, and $C_{12}H_{26}O_n$ increased from 7.6%, 5.0%, and 3.1% to 10.4%, 6.6%, and 4.7%, respectively.

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