

General Comments

This study investigates how systematic changes in both the magnitude and composition of the bimolecular $\text{RO}_2\cdot$ reaction sink, through varying contributions from NO , $\text{HO}_2\cdot$, and $\text{RO}_2\cdot$, affect HOM composition and SOA formation while maintaining an approximately constant α pinene + OH turnover rate in the SAPHIR STAR chamber. The work builds directly on Baker et al. (2024), extending the previously explored $\text{HO}_2\cdot/\text{RO}_2\cdot$ dimension to include NO containing and mixed $\text{NO}/\text{HO}_2\cdot$ chemical regimes.

Several aspects of the study are particularly noteworthy. First, the experimental design provides strong control over the entry flux into α pinene oxidation while systematically modifying the downstream $\text{RO}_2\cdot$ reaction environment, allowing chemical regime effects to be investigated more cleanly than in many conventional chamber experiments. Second, the analysis in Sect. 4.4 is particularly interesting. The inferred shift between the $\text{C}_{10}\text{H}_{15}\text{O}_x\cdot$ and $\text{C}_{10}\text{H}_{17}\text{O}_x\cdot$ precursor families is examined using three complementary molecular signatures, including non ON monomers, C_{20} accretion products, and organic nitrates. Together, these observations provide a persuasive mechanistic picture. Third, the authors are generally transparent about the limitations of their framework and appropriately report observations that cannot be fully explained by the proposed mechanisms, such as the persistence of the $\text{C}_{10}\text{H}_{14}\text{O}_z$ family.

Overall, the qualitative conclusions are well supported by multiple lines of evidence. In particular, the data consistently indicate substantial changes in HOM composition and a decrease in SOA formation potential as the system moves away from the $\text{RO}_2\cdot$ dominated regime. The seed addition experiments also support the conclusion that ON HOMs partition less efficiently to the particle phase than non ON products of similar molar mass under the experimental conditions employed here. However, several of the more quantitative and mechanistic conclusions require further refinement, because the central chemical regime parameters rely on modeled radical concentrations and generic reaction rate coefficients. Most of the concerns raised below appear addressable through additional analysis of existing data, clearer quantification of uncertainties, and expanded discussion rather than through new experiments.

Specific Comments

1. Sensitivity to generic rate coefficients

Please provide a table in the Supplementary Information showing how S_{RO_2} and the relative contributions of the reactions change when $k(\text{RO}_2+\text{RO}_2)$ and $k(\text{RO}_2+\text{NO})$ are varied within a plausible range, and clarify whether the relative order of the regimes

is maintained. In part, the framework relies on a single value: $k(\text{HOM-RO}_2\cdot + \text{R}'\text{O}_2\cdot) = 5.0 \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$. Because the relative contributions of $\text{RO}_2\cdot + \text{RO}_2\cdot$, $\text{RO}_2\cdot + \text{NO}$, and $\text{RO}_2\cdot + \text{HO}_2\cdot$ differ among conditions, uncertainty in this rate coefficient will not affect all conditions equally. Demonstrating that the regime ordering is robust to plausible variations in these rate coefficients would substantially strengthen the quantitative use of the framework.

2. α -pinene ozonolysis

Lines 212–213: Authors mention that at least 75% of α -pinene turnover was driven by the reaction with OH radical. However, since the O_3 inflow was adjusted to maintain a nearly constant OH $\cdot$ concentration, it may be possible that the relative contribution of ozonolysis varied depending on the CO and NO perturbation conditions. If the ozonolysis fraction changes alongside the imposed chemical regime, it could be an additional variable rather than a constant background factor. Please provide further details on this point in the main text.

3. Scope of conclusion

This experiment addresses only sub-ppbv NO conditions, with a maximum reported steady-state NO concentration ($[\text{NO}]_{\text{ss}}$) of 0.5 ppbv. Therefore, the broad reference to "urban" conditions in lines 645–646 falls outside the scope of this study, and a qualifying statement or explanation regarding this should be added. Similarly, regarding the statement in lines 636–637 that similar effects are expected for "all monoterpenes," it must be specified that the quantitative significance of RO_2 isomerization, alkoxy chemistry, nitrate formation, and accretion reactions varies depending on the type of precursor.

4. Minor details

- Abstract, line 20: "An important source ... are" should be corrected to ensure subject-verb agreement.
- Abstract, line 25: "Steady -state" should be corrected to "steady-state".
- Introduction, lines 38–39: "One important source of SOA are" should be corrected to ensure subject-verb agreement.
- Table 1: The extra closing parenthesis after "Saunders et al., 2003))" should be removed.
- Line 250: "lesser oxidized products" should be corrected to "less oxidized products."

- Line 375: “The good correlation ... show” should be corrected to “The good correlation ... shows”.
- Line 417: HOM-Acc is presented in Fig. 5e, not Fig. 5c.
- Fig. 6 caption: “ $C_{20}H_{23}O_z$ ” appears to be a typo for “ $C_{20}H_{32}O_z$,” which is used in the main text and the figure; verification is required.
- Line 612: “extend of formation” should be corrected to “extent of formation.”