SOA is important to both air quality and climate. Most previous studies predicted the SOA mass based on the SOA yield method from a signal precursor. However, VOCs and their oxidation products in the real atmosphere are a complex mixture (Nie et al., 2022). The interactions among different VOC oxidation processes can potentially influence the SOA yield but are largely understudied. This study investigated the influence of isoprene on the SOA formation from a-pinene, o-cresol, and their mixtures. The results did provide useful information and fit the scope of ACP. However, I have a few comments before this manuscript can be published.

We'd like to thank Referee 1 for their positive comments and to respond to the general and detailed comments as follows (reviewer comments in black and our responses in blue; the line numbers referred throughout are referring to the original manuscript).

1. **Reliability of measurement technology:** Thermal denuder and FIGAERO-CIMS need to heat the sample before detection. This may induce interferences via thermal decomposition, as well as chemical reactions occurred during the heating. For example, in the α-pinene/isoprene mixture system, there were no detected product molecules with nC=15 (Fig. 4a), which should be one crucial group of products from the cross-reactions between C10-RO2 and C5-RO2. In addition, most detected molecules by FIGAERO-CIMS were less oxidized (lower O:C) than those observed in the gas phase. What's the possible reason? And how to evaluate these possible instrument-induced interferences?

We thank the reviewer for his/her suggestion to discuss in more detail the potential effect of thermal decomposition in our manuscript. This has been also raised from the reviewer #2 and we acknowledge that it warrants some additional discussion. Indeed, thermal decomposition is undoubtedly affecting our measurements to an unknown extent (Stark et al., 2017). To reflect on this, we have added some discussion in the revised manuscript to acknowledge the thermal decomposition as a potential measurement artefact.

Regarding C_{15} compounds, indeed, one would expect to be potentially important group of compounds and their absence may be related to the thermal decomposition. Alternatively, these products might had very low signal, so we couldn't identify them reliably. This has been explicitly mentioned in the revised manuscript.

In response to "*most detected molecules by FIGAERO-CIMS were less oxidized (lower O:C) than those observed in the gas phase*"; in this work, we did not show any comparative O:C results between the gas and particle phase measurements. However, in our previous companion manuscript (see Voliotis et al., 2021), we contrasted gas and particle phase measurements from a subset of the experiments presented here and we found higher sum signal fractions of compounds with high O:C (O:C>0.7) in the particle than in the gas phase in the o-cresol system (24 and 9%, respectively). However, the opposite was also true for the more volatile α-pinene system (12 and 25%, respectively), while their mixture was in-between (i.e., α-pinene/o-cresol; 23 and 24% respectively), as was its particle volatility. Therefore, based on our observations, we believe that the split of O:C between the gas and particle phase is largely attributable to the volatility of the systems.

We see however the point that the reviewer is trying to make, i.e., that thermal decomposition of the products in the particle phase mode might result in lower observed O:C in the particle phase than in the gas phase, however in our case, volatility seemed to be a stronger driver for this. Again, as it was mentioned above, the potential effect of thermal decomposition to our results has been added as potential explanation in the discussion of the revised manuscript.

2. **Calculation method of partitioning coefficient (Line 230)**: This method is only valid when a compound is in equilibrium between the gas and particle phases and cannot be used to assess the volatility of very condensable compounds (< LVOC). Moreover, considering the large number of potential gas- and particle-phase reactions, it is difficult to ensure that compounds with the same molecular formula in both phases are indeed the same molecule, i.e., have the same molecular structure. The authors need to provide more results to clarify the accuracy, uncertainty and applicability of the method. Given the use of FIGAERO-CIMS, the authors may be able to infer volatility based on thermograms for individual molecular components (Thornton et al., 2020), and may be able to obtain information by comparing the results of the thermal-desorption-based approach with the results of the equilibrium method used in the text.

A similar point have been raised from the reviewer #2 (see comment #3), so we acknowledge that more information is required about this. In the revised manuscript we have added more information about our reasoning for selecting this method, along with its potential limitations.

Indeed, the partitioning calculations have an inherent assumption of gas/particle equilibrium. As we further show in Voliotis et al., (2021), Du et al. (2021) and also previously shown by Stark et al. (2017), this approach has some limitations in quantifying compounds with very low or high volatility (see L286-291). Previously, we have contrasted the partitioning approach with the thermogram-based using explicit T_{max}-V_p calibrations as well as the TD-AMS measurements for a subset of the systems (see Voliotis et al., 2021). We found that the volatility derived from partitioning calculations were more realistic and broadly comparable with the concurrent TD-AMS measurements opposed to the thermal desorption/calibration based approach. Hence that exercise informed our decision to select the partitioning approach here.

In more detail, as we state in our response to the reviewer's 2 comment #3: "*The volatility distributions obtained from the* T_{max} *calibrations were unrealistically high with the vast majority of the signal being accumulated in* $C^*>100$ *µg* m^{-3} *at SOA particle mass loadings* >200 *µg* m^{-3}*, inconsistent with the absorptive partitioning theory. We attributed this to the method we selected to introduce the calibrants in the instrument (syringe method) that was more recently shown to have an effect on the* V_p-T_{max} *relation (Ylisirniö et al., 2021), as well as to unquantified matrix effects during the desorption (Schobesberger et al., 2018). On the other hand, the volatility distributions obtained from the partitioning calculations were realistic and broadly comparable with the concurrent TD-AMS measurements. Importantly, despite the challenges in the volatility quantification from the FIGAERO-CIMS, we showed that our method is able to capture the volatility changes between the systems. Therefore, in this companion and follow-up paper, we have selected the partitioning method to illustrate the volatility changes and their effect in the volatility predictions as a result of the mixing of the precursors. The application of this method to our data is detailed in section 2.5 of the original manuscript.*"

A brief version of the above has been added in the revised manuscript in the methods section.

3. FIGAERO-CIMS can also measure the gas-phase oxidation products. Why not add some discussions of gas-phase chemistry to support the interpretation of the aerosol phase observation?

Indeed, FIGAERO-CIMS can provide detailed information both for the gas and particle phase products. Owing to our selected method to calculate the particle volatility that requires both gas and particle phase measurements to calculate the partitioning coefficient of each product(s) (see L227-234), we have used one common peaklist for both the gas and particle phase measurements and the only thing varying between them is their contributions in either phase. Therefore, the oxidation products that are shown here (Fig. 1, 2, 4 and 5) are broadly reflecting both the gas and particle phase composition, and each systems' volatility is by extent providing some information about their ratio. Consequently, the interpretation of our systems based on the volatility from the partitioning approach as well as an illustration of the particle composition should be adequate for the purposes of this work, which is to explore the volatility changes in mixed systems from the addition of isoprene and to assess whether additivity can predict those changes.

Nonetheless, we agree with the reviewer that the results that can be provided by the gas and particle phase measurements from the FIGAERO-CIMS can be insightful in understanding the systems' behaviour. In a subsequent manuscript (Du et al., in prep), we apply the methodology developed in Du et al. (2021) to all systems and by utilising dimensionality reduction techniques, we are able to identify the drivers of the SOA formation from the gas and particle phase time-series and link them with the results presented here.

4. **To Line 298-300**: The carbon number is not shown in Fig. 1c and Fig. S1.

We thank the reviewer for pointing this out. This has been corrected in the revised manuscript.

5. Table 1: Exp. 1-4: why the SOA yield change from 0.32 to 0.15?

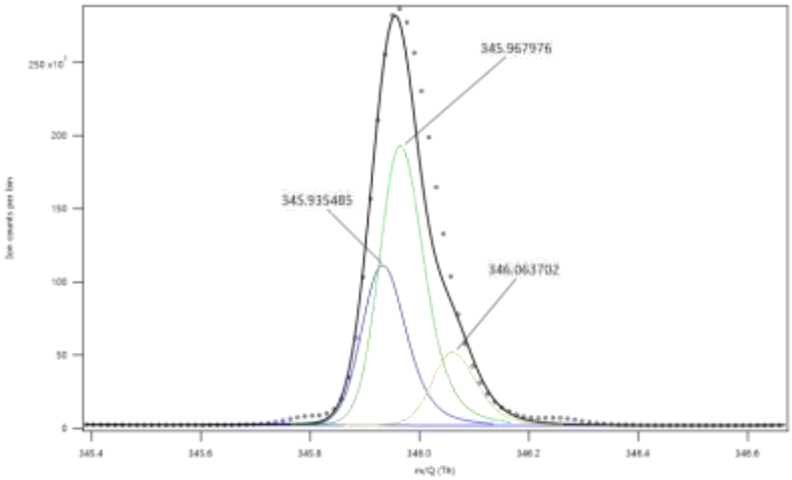
The SOA particle yield is reducing as the initial VOC concentration reduces (see column 4 of table 1); experiments at full initial reactivity (i.e., 309 ppb; Exp. 1-2) have higher SOA particle yield (Y=0.32) than those at 1/3 initial reactivity (i.e., 103 ppb; Y=0.15; Exp. 4). According to previous works, this is an expected behaviour, e.g., see Alfarra et al., (2012) and Chen et al., (2019), and references therein.

6. **To Line 357, 380, 408-409**: Based on the assigned elemental formulas, many compounds seem to be not closed-shell molecules, e.g. $C_{10}H_{10}NO_4$ and $C_{12}H_{11}O_4$, etc. Can the authors supply the results of HR peak fitting (including the measured signal, the fitted peak and the residual)?

We thank the reviewer for pointing this out. Indeed, the observation of open-shell species is unlikely in the FIGAERO-CIMS. We have now conducted a thorough investigation in our peak assignment and in retrospect, we are confident for our assignments, as they were the most likely candidates from all the related suggestions within our accepted error range. For example, see the case of $C_{12}H_{11}O_4$ below, a compound detected in the *o*-cresol/isoprene system including measured signal, fitted peaks and residuals. What we didn't account for, was any potential secondary chemistry in the IMR (e.g., Zhang and Zhang, 2021) and/or potential adducts with $I.H_2O$, $I.O_3$, etc (e.g., Veres et al., 2020;Murschell et al., 2017). As the compounds shown in the original manuscript were randomly chosen to cover a range of nC and did not reflect their importance, in the revised manuscript we selected to show the unique compounds that had the highest signal contributions.

Case of $C_{12}H_{11}O_4$

The figure below shows the fitted peaks at the UMR of the related ion (i.e., $C_{12}H_{11}IO_4$; 346 m/z) directly exported from Igor Pro, using the Tofware v. 3.1.2 workflow. Three peaks were fitted in the UMR, shown in different colours and the total fit is shown in black. Here, we attempt to identify the largest peak (shown in green; exact m/Q: 345.96976). The table below shows all the suggested molecular formulae and their exact mass, sorted based on the fitting error (ppm). As can be seen, within our trusted error range (~6 ppm), the only two suggested formulae that can exist as molecules are the $C_7H_{11}IN_2O_6$ (5.2 ppm) and the $C_{12}H_{11}IO_4$ (6.4 ppm); both open-shell species. This remains to be the case even if we extend our trusted fitting error range to ~9 ppm. Based on these elemental formulae, the former, unsaturated molecule containing two nitrogen atoms, is probably unlikely in this system (*o*-cresol/isoprene). Therefore, our only viable option in this case would be to assign the $C_{12}H_{11}IO_4$.



Suggested Formula	Exact m/Q	Error (ppm)
$C_{10}H_4NO_{13}$	345.9682641881199	-0.83299005
$C_2H_8N_3O_{15}S$	345.9676127394399	1.0499843
$C_{17}H_2N_2O_5S$	345.96844234566	-1.3479427
$C_3H_{24}I_2S$	345.96881574792	-2.4272358
$C_{10}H_9IN_3O_3$	345.96886306207	-2.5639939
$C_4H_{10}O_{16}S$	345.9689554103001	-2.8309195
$CH_{17}INO_9S$	345.96687420311	3.184689
$C_4H_{15}IN_2O_6S$	345.96955428425	-4.5619168
$C_7H_{11}IN_2O_6$	345.9661829809299	5.1826425
$C_{14}H_4NO_8S$	345.96576226452	6.3987126
$C_{12}H_{11}INO_4$	345.97020573293	-6.44487
$C_7H_6O_{16}$	345.96558410698	6.9136734
$C_6H_{20}I_2$	345.9654444446	7.3173647
$C_{13}H_2N_2O_{10}$	345.97094426926	-8.5795336
$C_{20}N_3O_2S$	345.9711224268	-9.0944777

7. **To Line 449-450**: It seems that SOA particle mass yield of o-cresol was reduced by the presence of isoprene (from 0.11 to 0.06/0.05).

We thank the reviewer for pointing this out, indeed the yield was reduced in this mixed system compared to the single precursor experiment. We have corrected our statement in the revised manuscript.

8. **To Line 541**: As shown in the study of McFiggans et al. (2019) and Heinritzi et al. (2020), isoprene depletes OH radicals, preventing their reaction with monoterpenes, and the resulting isoprene peroxy radicals scavenge highly oxygenated monoterpene products. These effects ultimately suppress both particle number and mass of secondary organic aerosol. It can be simply inferred that the mass yield, composition, and volatility of SOA will be altered after mixing isoprene in the monoterpene system, but this was not the case in this study. Given that FIGAERO-CIMS can provide near-molecular information, it is appropriate for the authors to discuss the above situation.

Indeed, isoprene can compete for the available oxidants and/or scavenge the peroxy radicals of the other precursors involved in each system (e.g., *o*-cresol for the system described in L541). This can result a reduction of the SOA mass, which was observed here (see response to our comment #7 above and Table 1). Further, in all mixed systems we found evident changes in the chemical composition compared to the single precursor systems, which is discussed in the original manuscript submission in detail (e.g., see L430-439, 454-464, L510-519). We similarly found changes in the SOA particle volatility (see Sections 4.1 and 4.2), apart from the α-pinene/isoprene system, consistent with previous literature (Ylisirniö et al., 2020; see L428).

More specifically, in all mixed systems, we attributed a significant fraction of the signal in products that were uniquely found in mixtures (see L32). These products might either derive as RO_2-$R'O_2$ termination products (where R and R' radicals deriving from each individual precursor in each mixed system) and/or as products that were derived from the alteration in the oxidation pathways of each precursor due to the potential changes in the chemical regime (see Voliotis et al., 2022). We cannot distinguish which of the two is more important as FIGAERO-CIMS cannot provide molecular information, however the likelihood of both to affect our results is acknowledged in the original manuscript e.g., L451.

Given the above, we are unsure of what the reviewer is suggesting here.

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