The reviews of our paper are thorough and well-considered. We would like to thank the referees for their obvious effort in reading, understanding and critically reviewing our manuscript and for the improvements that they have helped us make.
All referees requested some simplification, clarification and shortening of the manuscript to some extent and we have taken this on board in the revision, with restructuring, shortening and modification in the focus.
We'd like to thank Referee 2 for their positive comments and to respond to the general and detailed clarifications, criticisms and suggestions as follows (reviewer comments in italics, line and Figure numbers refer to the original discussion manuscript for clarity):

Review of "Chamber investigation of the formation and transformation of secondary organic aerosol in mixtures of biogenic and anthropogenic volatile organic compounds" by Voliotis et al.

This paper reports on the initial findings of a series of laboratory experiments conducted in the Manchester Aerosol Chamber to examine the behavior of oxidation of mixed biogenic and anthropogenic VOCs to determine deviations from expectations of the yields of secondary organic aerosols, and possible reasons for such deviations. The paper presents the experimental design and selected results in a logical sequence that is generally easy to follow and understand. This reviewer found the topic interesting with cleverly designed experiments and insightful interpretation of the wide array of measurements. This paper represents a valuable contribution to our understanding of the details of oxidation of VOCs as related to production of aerosols under various conditions. The McFiggans group has led the community in the interesting area of mixed biogenic-anthropogenic VOC oxidation and production of SOA. This paper, and the ones to follow about these experiments, continues that legacy.

On the organization of the paper, after the introductory material, the figures are discussed along with aspects of the study (starting on line 375). The last figure (13) is referenced on line 650. Then in the discussion, the figures are discussed again (starting on line 674) in order. This reviewer suggests that the authors say everything to be said about the figures in the first pass, and then shorten the discussion to only the key points of the study. This is only a suggestion for the authors to consider.

We thank the reviewer for this suggestion and agree that the organisation could benefit from attention. We have partly taken the referee's suggestion on board but have more radically changed the paper to present all the results from the companion papers along with the associated Figures, into the supplementary information. This leaves them only to be referred to in the discussion section along with some discussion of the expansion of the interpretations in each paper. This is because of the progress made in the reviews and publication of the companion papers and the requirement to maintain the focus on the key new points in the current paper.

General comments.

While this review found the paper overall well-written and organized, there were some instances of jargon usage and awkward wording. These are pointed out in the specific comments. In the discussion and conclusions, the interference of VOCs in the measurement of ozone was noted. Had this been recognized earlier, an alternative method of measurement could have been employed (e.g. reverse NO chemiluminescence). Perhaps this should be briefly mentioned. It is also important to note that the Thermo 42i NOx analyzer has significant interference from reactive nitrogen compounds other than NO_2 in the NOx *mode due to the converter employed in this instrument. This should also be mentioned, perhaps in a footnote of Table 2.*

These are well-noted points. We became aware of the ozone interference during the measurement campaign and hurriedly attempted to deploy an old LOZ-2 ozone analyser using chemiluminescence with eosin-Y dye for all *o*-cresol containing experiments. Unfortunately the instrument suffered problems and useful data were not recoverable. Concerning the NOy interference, we have noted this in the revised text.

Specific comments.

Line 60. In the first mention of VOC:NO_x ratio, indicate that it is a ratio of mixing ratios (in other words, a molar ratio). This is important because is could be a ratio by mass.

We have clarified this and included ppb/ppb in the text.

Line 61. It is not clear what is meant by "less mechanistically". Perhaps use different terminology.

We mean that the exact chemical components acting as precursors and the mechanistic interactions in their degradation chemistry influencing the components partitioning to the particles are seldom considered. We have tightened up the language here.

Line 66. What is meant by "numerous representations of atmospheric SOA"? Consider different wording.

Rephrased to "numerous ways of representing the formation and transformation of atmospheric SOA"

Line 129. Suggest "use these metrics to quantify interactions in the oxidation of VOC mixtures leading to changes in SOA formation compared to those expected based on..."

Agree – changed.

Line 132. Suggest "use a suite of online and offline measurements...and the resulting properties of potential atmospheric significance."

Agree - changed

Line 141. Perhaps reword or add additional text to explain "...makes a comprehensive programme intractable."

This has been both reworded and expanded a little.

Line 151-2. Suggest "...study of a random mixture is expected to yield novel, but complex results. Thus, care is required to fully interpret information from such studies." ...or something similar.

Yes – this sentence has been simplified.

Line 159-160. This reviewer is not convinced that it is established how NO_x *affects SOA formation yields dramatically. While some studies have shown this, others are less convincing. It is not even obvious what the slope of the yield versus* NO_x *function is. Suggest softening this assertion.*

The NOx dependence is indeed ambiguous and we have recognised the contradictions in the literature in the revision. The mechanistic pathways during VOC oxidation during which SOA particles are formed are undoubtedly influenced and this is really all we were saying. It was in the light of this that we were contextualising our choice of VOC:NOx ratio. The assertion has been softened.

Line 162. The sentence that begins "Truly low NO_x *regimes do not occur widely in the ambient atmosphere..." could certainly elicit discussion. It depends on the definition of "low* NO_x*" and "widely", which have not been presented here. There are certainly remote regions where the* NO_x *is very low (10s of pptv), but it probably is hard to argue that such locations are widespread. Suggest softening this assertion. Since the use of low and moderate* NO_x *levels are used in the paper, perhaps somewhere consider quantifying what is meant by these descriptors.*

We have balanced and softened the assertion.

Line 182. Suggest changing "...sort of atmosphere that..." to something else that is clearer, such as "...level and mixture of atmospheric components that are employed to approximately represent a given atmospheric situation."

Clarified our sloppy language.

Line 188. Several places in this paper use future tense, when present tense is more appropriate. This is one of those places. Suggest "...of the current approach is of considerable interest."

We have checked our tenses throughout.

Line 196. The use of ammonium sulfate particles is mentioned and justified, but the reason for this particular choice is not mentioned. Perhaps include a bit more information.

We're not completely sure of the additional information that the referee is asking for, but we have added a little more justification in terms of ammonium sulphate being one of the most abundant electrolytes in the atmosphere and a component most easily and reliably determined by online instrumentation such as the AMS (to contrast with, for example, sodium chloride).

Line 203. Suggest "Ammonium sulphate solutions are nebulized into...".

Agree – changed.

Line 210. Table 2 is referenced here, before Table 1. Suggest renumbering and reordering the tables. It is stated that the measurements are "state-of-the-science". While this is true for some of the instruments, it is not universally true. Suggest rewording this sentence.

We have removed reference to table 2 at this point and the statement of the instrumentation being "state-of-the-science …throughout".

Line 211. Time resolution of the measurements is mentioned. Perhaps include the time resolution or integration time in the instrument table.

Table 2 has been updated with the time resolution as follows:

Table 2: List of instrumentation employed over the course of the study

Instrument	**Model**	**Measured parameter**	**LOD/ range**	**Time resolution**
Dew point hygrometer	Edgetech (DM-C1-DS2-MH-13)	Dew point	-20 – 90 ± 0.2 °C	1s
NOx analyser	Thermo 42i	NO, NO_2	0.5 to 1000 ppb	10s
O_3 analyser	Thermo 49C	O_3	0-0.05 to 200 ppm	10s
Water-based condensation particle counter	TSI 3786	Particle number	$<10^7$ p/cc	1s
Differential mobility particle sizer	Custom-built[a]	Particle size	40-600 nm	600s
Filter collector	Custom-built[b]	Particle collection for offline analysis		n.a.
Condensation particle counter	TSI 3776	Particle number	$<10^7$ p/cc	n.a.
Scanning mobility particle sizer	TSI 3081	Particle size	10-1000 nm	120s
High-resolution aerosol mass spectrometer	Aerodyne	PM_1 non-refractory particle composition	>0.05 µg m^{-3}	60s
Iodide chemical ionisation mass spectrometer	Aerodyne/Tofware	Oxygenated VOC	LOD >60 ppt; Mass resolution 4000 Th/Th	0.25s
Filter Inlet for Gases and AEROsols	Aerodyne/Tofware	Particle composition	$>10^2$ ng	n.a.
Semi-continuous gas-chromatograph mass spectrometer	6850 and 5975C Agilent	VOC concentration	>0.4 ppb	1200s
Liquid chromatograph – orbitrap mass spectrometery	Dionex 3000, Orbitrap QExactive, ThermoFisher Scientific	Particle composition		n.a.
Hygroscopicity tandem differential mobility analyser	Custom-built[c]	Hygroscopicity	20-350 nm	600s
Cloud condensation nuclei counter	Droplet measurement Tech (model CCN-100)	CCN activity	$>6 \times 10^3$ particles cm^{-3} at SS:0.2%	600s
Thermal denuder	Custom-built[d]	Volatility	Temperature range: ambient – 200°C	n.a.
Three arm bounce impactor	Custom-built[e]	Particle bounce	20-500 nm, $< 10^4$ particles cm^3	90s

[a] Alfarra et al. (2012)
[b] Hamilton et al. (2011)
[c] Good et al. (2010)
[d] Voliotis et al. (2021)
[e] Liu et al. (2017)

Line 215. It is a little unclear what is meant by "collection of sufficient mass" in this context. Perhaps add a few words to indicate that the detection limits for aerosol components are enhanced by maximized the total mass of aerosol collected.

Added. This is indeed the point.

Line 234. It is not clear what is meant by "...throughout the photochemistry...". Suggest rewording.

Reworded to "as it transforms throughout the experiment".

Line 236. Suggest rewording "...characterization of the transformations of the oxidation products...". The oxidation products are the result of oxidant attack on the primary VOCs, perhaps in multiple generations, so oxidation products can be oxidized, but it is not clear what the point is here.

Agree – we quantify the changing concentrations of oxidation products that may partition between phases.

Line 237. It is unusual to use the term "payload" for instruments used in a ground based laboratory or field study. It is usually reserved for aircraft studies. Suggest using "suite" or some other term.

Changed to "suite"

Line 251. Suggest "The biogenic VOCs that were chosen were α-pinene and isoprene..."

Changed

Line 256. Suggest "This means that the initial amounts are added in a α-pinene : isoprene : o-cresol ratio of 309 : 164: 400 based...".

Changed

Line 266-7. Suggest rewording this sentence. This appears to be a disjoint collection of sentence fragments. It might be easily fixed by a couple of transition words.

Tried to make it a bit more fluid

Line 273. Change to present tense: "non-linearity leads to changes in the...". Line 277. Could eliminate the word "Ambient...".

Tense changed

Line 287. Suggest "...mimic a specific atmospheric region, but more to explore...".

We use "atmospheric chemical regime" to capture not only geographical region, but seasonal / diurnal temporal variability and are not sure that this suggestion captures what we intended.

Line 289. I'm not sure what is mean by "neutral seed experiments". Suggest rewording.

Changed to “non-acidic”.

Line 290. Suggest “...particularly acid catalysed reactions.”

We are not sure that this is correct so have revised to “…condensed phase reactions, particularly those that are acid catalysed”

Line 298. Suggest replacing “space blanket” with “Mylar film”. Also suggest “...homogenise the light intensity throughout the chamber.”

Changed

Line 302. Suggest “...transmit light up to 100% above 305 nm.”

Changed

Line 318. Suggest “...is conducted using full illumination without the UV filter on the arc lamps and using ppm levels...”

Changed

Line 322. It is not obvious why the term “stock solution” is used and why the concentration is given, since the sentence indicates that the concentration is changed to control the seed concentration. Suggest rewording.

Reworded

Line 325. Suggest rewording the phrase to read “...by irradiating the chamber mixture for the selected conditions of VOC and NO_x.

Reworded

Line 327 and 329. It appears these are almost the same sentence. Suggest eliminating one of them. It is possible that all the text in lines 327-330 could be replaced by “The correction for the interference by o-cresol is determined by the ozone instrument signal measured before the experiment began and using the change in o-cresol concentration determined by the CIMS instrument.”

Deleted the repetition

Line 345. Suggest beginning this sentence with “The instrumentation includes: a high-resolution...”

Reframed the sentence as suggested

Line 351. Suggest “...of the online instrumentation was changed after several hours of reaction to cycle...”

Changed

Line 353. Suggest "...Table 2 provides the list of instrumentation...". Also change table number as per previous suggestion.

Removed the earlier reference rather than renumbering

Line 357. Suggest adding more detail about the "actinometry and off-gassing experiments". At least say what parameters were determined, and maybe briefly describe how they are done. Also include references as appropriate.

We've referenced Shao et al. (2022a) and briefly expanded the sentence.

Line 362. Suggest "...in the MAC forms O_3 *which rapidly establishes the photostationary state...".*

We've subsumed much of the text into a methodology section which is now in section 2 and this sentence has been revised in this section.

Line 367. It seems that the phrase "at a given OH" could be removed. If there is isoreactivity, then it doesn't matter what the OH is, correct?

True. What we had meant to state was "… at *any* given OH concentration" – it does not state that the OH will be the same between systems.

Line 369. Suggest "...concentration), in practice the loss rate of each VOC..."

Changed

Line 371-3. The formation of HONO on chamber walls and release to the gas phase is well-known and has been studied for a long time. Suggest a more recent reference and perhaps a bit more discussion for those readers that might not be familiar with this issue.

A briefly expanded discussion has been provided in the revision

Line 379. Figure 1 x-axis label. Suggest something like "Illumination time (h)".

We agree that "lights on" is a bit parochial, so have taken this suggestion

Lines 382-395. With the discussion of the photostationary state and the Leighton ratio, in principle you could calculate the HO_2*+*RO_2 *concentration that explains the observed ratio. Not discussed were the instances where the ratio is observed to be less that unity. Suggest adding some text to expand this discussion, or alternatively to eliminate the topic completely.*

We include some discussion of this from line 682 in the original manuscript which we have expanded in the revision. The values below unity imply a local source of NO. This may arise from a wall NOx source, but must be the subject of further investigation.

Line 420. Suggest "...not all VOC were consumed...". This is because VOC is plural. This reviewer prefers VOC for singular and VOCs for plural, but this is not widely accepted.

We are in agreement with the reviewer in preferring pluralisation to VOCs, but will defer to convention. We have changed all occurrences to reflect this.

Line 431. Suggest "The same colour scheme as in Figure 2 is used."

Changed

Lines 440-1 and 443. The terms "particle mass" and "SOA particle mass" are used interchangeably in the first sentence, but this is not strictly correct. It is not really necessary to include "particle" with SOA, since SOA includes "aerosol". "SOA mass" should suffice in this discussion. Note that "particle mass" and "SOA particle mass" are used incorrectly (meant to mean SOA mass, but strictly meaning total aerosol mass) throughout page 18 (and Figure 4 caption), and perhaps elsewhere in the paper. Please search for this term and correct its usage.

We agree with the first part of the referee's comment – particle mass is not SOA particle mass, since it will include inorganic. However, the particle yield does not include the gas phase components, so the yield should be SOA particle mass / VOC consumed. It should not be Total SOA mass (including gas mass) / VOC consumed. We have checked all usage and made it consistent with this definition, which we contend is correct (and consistent with previous usage).

Lines 443-444. It is difficult to see that that this sentence is a list of possible alternatives for calculation of yields. Perhaps number (1, 2, 3) of add a letter (a, b, c) to the various options, or change the wording of the first part to indicate that various options are coming in the rest of the sentence, for example "...the yield is reported as a single number that could be calculated from data at various times in an experiment, including at maximum SO2 mass, ..."

We have broken down the sentence to make it clearer

Line 480. The term "referenced" is used, but perhaps "compared" would be better. This applies at many places in the paper.

We had used the term "referenced" since it is intended to specify a reference baseline. We agree that compare is more precise, and use "compared to the baseline mixture without isoprene" etc…

Line 481-481. The nomenclature for the various yield calculations can make the sentence grammar confusing. This sentence is an example. The use of "yield at maximum mass" might be better with a symbol (such as Y_{MM}) which is defined somewhere and then used in the discussion. Likewise with the other types of yield calculations (e.g. Y_{MVC}).

We agree and have adopted this suggestion

Line 482. Suggest "The uncertainties in SOA mass yields were calculated by...". Also, this implies the uncertainties shown in Table 3 are 1σ, but this is not explicitly stated.

We disagree with this, since SOA does not include gaseous mass, so retain “SOA particle mass yield”

Table 3. In an earlier table, a “-“ indicated missing data. What does it mean in this table?

The ‘-’ also indicated missing data in Table 3. Table 3 has been replaced by the figure as shown in Response to Referee 1.

Line 485. In this sentence “organic mass” is used to mean “SOA mass”. Suggest being consistent in the terminology throughout the paper.

Yes, the terminology should be consistent throughout the whole paper with “SOA particle mass”.

Also, this sentence is confusing, because it apparently refers to equation (4), but equation (3) is shown first. Suggest reordering the text and equations to make things clearer. The symbols in both equations need to be defined.

Equation 3 has been moved ahead of the Figure that replaces table 3, immediately after the introduction of equation 3 and all terms have been defined.

The term “additively combining” means simply that the predicted yield is the mass of SOA for each component determined from the single VOC experiments at the same amount of VOC reacted divided by the sum of the amounts of each VOC that reacted. It may be that the sentence in line 485-6 is meant to say this, but it is a bit confusing. Consider rewording.

The referee is correct – it has been reworded to clarify “Figure 5 shows “predicted” yields for the mixtures, based on the sum of the SOA mass determined from the single VOC experiments divided by the sum of the amounts of each VOC that reacted according to eq. 4”

Line 490-3. This sentence is also confusing. Are the two-product fits referred to from equation 3? Perhaps more information is needed to clarify this sentence. Also, the term “particle mass” is used again.

The sentence has been rephrased to: “The parameters (alpha1, alpha2, Kp1, Kp2) from the two-product fit by eq.3 from the single VOC half- and third-reactivity experiments were used to generate a yield-mass-ΔVOC look-up-table. This look-up table is then used to predict the SOA particle mass from each VOC with known VOC consumption in the mixed systems.”

Line 528. It is not clear what is meant by “decremental decay” in this context.

This means the stepwise decrease, time interval by time interval – in the same way that an incremental build up would be used to describe a stepwise increase.

Page 23. There are several instances of “particle mass” on this page.

Now all SOA particle mass

Line 539. Suggest “Measured (15% error) and reconstructed decays of (a) isoprene and (b) α-pinene in...”.

Changed

Line 540. Suggest "In each case, the initial decay of the VOC due to reaction with ozone was calculated based on the initial concentrations of O_3 *and the VOC along with the appropriate reaction rate coefficient."*

Sentence rewritten to clarify

Line 548-549. Suggest "...yields owing to the differences in the tendencies of oxidation products from reaction with different oxidants to condense."

Agree - changed

Line 551. Suggest "...AMS total signal at m/z values of 44 (f_{44}*) and 43 (*f_{43}*) to represent more or less oxygenated contributions, respectively, to the SOA mass."*

From this point onwards to line 660, all Figures and results have been moved to the Supplementary Information, with much of the text subsumed into extended Figure captions. We have responded to the comments here, but most of this is now in the SI.

Line 554. Suggest "...in all systems explored in this study."

Agree. Included in expanded caption

Line 564. It is not clear what is meant by "...more oxidized and higher...". Suggest rewording.

Agree, now "…more oxidised products with higher f_{44}"

Line 576. Suggest "A more thorough study of the aerosol composition using analysis of the high resolution AMS is data is the...". Not all readers will be familiar with what "high resolution" means in this context, so somewhere define that it is high mass resolution (as opposed to high time resolution or some other type).

Agree – now state "high mass spectral resolution" at first use.

Line 587. Suggest "...and mixed α-pinene / o-cresol systems...".

Changed

Line 599. Suggest defining "-ve" and "+ve" ionization modes here or elsewhere in the paper.

-ve and +ve replaced with negative and positive throughout

Line 601. Suggest "...phase reactions cannot be determined from these...".

Changed

Line 606. It may be obvious but suggest defining the term "unique-to-mixture" compounds somewhere.

These compounds are now solely referred to by the description “compounds uniquely found in the mixtures”

Line 616. Suggest “Panel (a): Amounts of $C_7H_7NO_4$ *isomers products in o-cresol systems as measured by...of each experiment in this study; and (b) time series of total aerosol phase* $C_7H_7NO_4$ *(all isomers) from...”. Also, do these plots indicate inconsistency between the two measurements? If so, this should be discussed in the text.*

Changed to ‘Panel (a): Amounts of $C_7H_7NO_4$ isomers products in *o*-cresol systems as measured by LC-Orbitrap MS analysis of each experiment in this study; and (b) time series of total aerosol phase $C_7H_7NO_4$ (all isomers) from FIGAERO-CIMS. The peak area of each compound/isomer was normalised to the corresponding total peak area of all detected compounds from LC-Orbitrap MS and FIGAERO-CIMS, respectively’. The figure is shown in Figure S10.
The reasons behind the different chemical compositions between the two instruments are discussed in Du et al., (2022): ‘there are a number of factors that could result in the differences in the composition, including thermal desorption of SOA in the filters or the selectivity and sensitivity of instrumental ionisation methods toward compounds (Stark et al., 2017; Mehra et al., 2020; Voliotis et al., 2021) and possibly differences in the limits of detection of the instruments as suggested by the reviewer. It is not possible to attribute the differences in composition to a specific cause in our experiments, but this should be the focus of future work.’

Line 625. Suggest “...found predominately in the aerosol phase (i.e. exhibiting low volatilities).”

Changed

Line 632-3. It is not clear why the data from the end of each experiment was presented in Figure 11. Would it not be better to either show time series or to show data at the peak of the SOA yield. Suggest adding some explanation in the caption or the text why this approach was taken.

The time evolution of the O:C and nC (and perhaps the OSc) is indeed interesting and is described in detail in an upcoming publication (Shao et al., 2022; in preparation). Towards the end of each experiment, most of the systems tended to be more stable, i.e., the formation rate of SOA particle mass < wall loss rate, indicating less rapidly changing chemistry (see Fig. 3). Therefore, the data from the final cycle of the FIGAERO-CIMS in each experiment were chosen as a reference. The caption has been extended in the SI

Line 656. Suggest “The evolution of these properties for all the systems in this study are discussed in detail...”

Changed

Line 666. Suggest “The control of photochemical conditions in the VOC mixtures studied here is challenging.”

Changed

Lines 666-673. In the discussion of the challenges with different reactivities of OH and O_3 *for the various VOCs, you might want to discuss the philosophy of adding oxidant scavengers. For example, a carefully selected alkane could be used to scavenge OH and then* O_3 *reactions could be studied in isolation. The (as stated elsewhere in the paper) an alternative OH source that doesn’t make ozone could be used to study*

OH reactions separately. Good understand of single oxidant systems could then be followed by those with multiple oxidants. Clearly, in a study such as this, one must be careful with such approaches, but it would be helpful for the reader if the topic is discussed.

This opens a philosophically interesting set of discussions. Careful selection of an alkane can indeed be used to scavenge OH. However, the peroxy radical product will act as a potential termination partner for peroxy radicals formed from SOA particle precursor oxidation. This involvement of the products of scavenger oxidation in the formation of SOA particles makes the impacts on their formation in mixtures quite challenging. Additionally, the reference baseline for the mixture yield would need to be established – is the VOC consumption included in the yield. This is precisely the discussion we opened with respect to isoprene in the systems we have reported. We are sure that there is insight to be gained by isolation of the oxidants, but are not sure of the best way to do this. We have included a brief section further raising these points in the amended discussion.

Lines 681-2. Need to define "+ve" and "φ".

+ve replaced with "positive", "ϕ" now "Leighton ratio, ϕ"

Line 690-694. In this discussion, it is clear that the chemistry of SOA formation needs to be understood over the range of oxidants studied: from pure OH to pure O_3*. With such understanding, studies in specific systems of OH :* O_3 *can potentially be compared. It is complex, but as with other chemical systems, the detailed mechanistic understanding must be developed before results from complex systems can be interpreted. Did you consider trying to develop a detailed model (using MCM with specific additions, for example) to try and constrain the mechanism responsible for the observations?*

We have thought hard about the means to constrain the mechanisms. It became apparent throughout the campaign that the differences in the oxidants were substantial from one mixture to another. The experimental design to move from one oxidant to another is quite different and out of scope of our study. We agree that the detailed mechanistic understanding must be developed before results from complex systems can be interpreted, but it has never been demonstrated that this is the case – for example, that yields in mixtures are not additive, or composition changes when combining precursors, for example. This is the starting point of our study and the mechanistic interpretation will necessarily follow from subsequent experimental campaigns. This is going to be a long programme of study…

Line 700. Suggest "...derived RO_2 *must be considered."*

Changed

Line 702. Suggest "...reaction with NO_2 *can be quite complex."*

Changed

Page 33 and 34. In the discussion of the complexity of such systems as in this study involving multiple oxidants with reactivities of VOCs toward the oxidants changing with time, it should be recognized and pointed out that oxidants can be measured. Indeed, you have direct measurements of O_3 *(albeit with the o-cresol interference) and direct OH measurements are possible. Such measurements seem critical to the success of studies such as the one presented in this paper. There are alternative methods (such as decay* *of a VOC that only reacts with OH; VOC decay mentioned in line 719) to determine oxidant levels. Also, OH levels can be minimized by addition of compounds that do not affect aerosol yields, such as CO. Thus, it is conceivable that many of the complex issues discussed on these pages could be managed by careful design of experiments, including the addition of further experiments. Additionally, it is conceivable that compounds other than those selected might be better choices for a mixed oxidant system. For example, selection of an aromatic compound with an unsaturated side chain might be a better choice than o-cresol. Perhaps one of the other terpenes with different relative reactivities toward OH and* O_3 *might be more suitable.*

We thank the reviewer for these suggestions. We have continually turned over these ideas in the planning of these, subsequent and now ongoing experiments. In our previous study (McFiggans et al., 2019) we used CO and CH4 which both acted as OH scavengers, but also showed that their oxidation products influenced the a-pinene yield. Addition of reactive species as an OH probe cannot be guaranteed not to perturb the SOA production in the system. As mentioned in the response to referee 2, direct measurement of OH is desirable but very non-trivial. Choice of the VOCs is also nuanced. There does not appear to be a clear winner and there is always a trade-off. The fact that o-cresol does not react with ozone is useful in determination of OH-concentration from its decay in cresol-containing mixtures. An unsaturated side-chain would make this inaccessible. We are unsure about the relative merits of terpenes with differential oxidant reactivities, but have looked at such individual precursor systems previously (see Alfarra et al., 2014). These are all touched on in the discussion section.

Line 704. Suggest “...the oxidant regime is also complex.”

Changed

Lines 705-6. The first part of this sentence is not clear. Why does NO_x *level affect the reactivity of alkanes toward OH? Suggest rewording to make clearer.*

We agree this was confusing and conflated two unrelated points. We have clarified this sentence.

Line 712. Suggest “...since anthropogenic VOCs seldom exist...”

Changed

Lines 713-4. It is asserted that reduced NO_x *levels lead to reduced OH levels, but this is not necessarily true. It depends on the how high NO levels are in the first place. It could actually be the opposite that reduction in NO leads to an increase of OH. This argument needs to be reworded or removed. Also, remove the future tense in “This will inevitably lead...”. Change to present tense.*

Agree. Changed in both instances

Line 716-7. It is not clear what is meant by the last phrase that states that the VOCs were chosen for their reactivities which leads to their concentrations be of comparable magnitude for mixture reactivities that are comparable. First, “comparable magnitude” is a vague term that is not well defined. It is stated elsewhere that the ratios of VOCs at equivalent OH reactivity are 309:164:400, which implies the ratio of the highest to lowest amount is 2.4. Is this comparable? Second, what does comparable mixture reactivities mean? Does it mean that the sum of the VOC reactivities toward OH (1/k[VOC]) for the binary *and ternary experiments are similar to the reactivities for the single compound ones? In any case, suggest rewording the last part of this paragraph.*

We have used "Comparable magnitude" loosely to imply concentration values within the same order of magnitude (here 100s of ppb). As discussed in section 2.4, the total reactivity of all systems towards the OH was roughly the same and the reactivities between the single component and the mixture experiments were similar. The sentence has been reworded for clarity.

Line 721. It is not clear what is meant by "...in the system practice." Suggest rewording.

We do not know what it means either and it is a typo that slipped through. Sentence reworded.

Line 722. This sentence has "owing" twice. Suggest rewording. Also as stated earlier, recognize that other methods of ozone measurement exist and could be used for such studies.

Agree, changed. We did try, but our alternative method for ozone measurement failed.

Line 725. It is not clear that it has been demonstrated that the amount of OH in the chamber is adequately constrained. If true, this needs to be discussed in more detail to make the case more clearly. To begin, the term "adequately constrained" needs to be defined.

As shown in Figure 6 reasonable closure of the VOC decay (within uncertainty) can be obtained arriving at OH concentration by indirect calculation. We have changed the sentence to state that some constraint on OH concentration is possible.

Line 730. The limitations due to the offline filter collection and analysis has been mentioned several times. It constrains how low initial VOC levels can be, for example. Are there alternative analytical techniques that could take the place of this approach? Elimination of this constraint would certainly make the experiments easier to design.

Online mass spectrometric techniques are approaching the mass spectral resolution enabling similar capabilities as the offline analyses with useful detection limits, though are not quite there yet. Coupling of the LToF and more recently Orbitrap mass spectrometers to Chemical Ionisation or online Extractive Electrospray show promise and it should enable easier experimental design in the coming years.

Line 727-743. It seems that employing detailed chemical models might help in the interpretation and design of the conditions and evolution of these experiments. Rather than state something "would like" be the case, specific and quantitative statements could be made. Employing such models is still possible even in the post-analysis phase of these experiments, although they could also have been very useful in the initial design of the study.

Coupled models of aerosol microphysics and photochemistry such as PyCHAM (O'Meara et al. 2021) have employed gaseous photochemical mechanisms such as the MCM and should be able to use more explicit mechanisms such as GECKO-A. These can be coupled with extensions to consider autoxidation such as PRAM (Roldin et al., 2019), but the rate constants for reasonable constraint of O_3 and OH initiated autoxidation for multiple species in complex mixtures are as yet unavailable. At the current state-of-the-science, "would likely" is the most reasonable statement possible at line 740 , since the ability of bimolecular or unimolecular reactions to interrupt the various steps in the reaction pathways of the numerous potentially autoxidising species is dependent on too many unknown rate constants. A huge amount of activity is underway with experiments to unravel such mechanistic information in admittedly simpler systems, as yet, in a number of laboratories including our own.

Line 747. The word "straightforwardly" and elsewhere "straightforward" is used several times. Suggest rewording these sentences. Also, it is not clear how an equation can be easily applied but with consideration of oxidant regime. An equation has inputs and an output. How does this change with "consideration"? Suggest rewording.

We agree that the sentence makes too many assumptions of prior knowledge and "turns over too many pages". We have expanded with a simpler explanation.

Line 751. Suggest "...that the corrections accounting for the loss of particles...".

Changed

Line 759-60. Suggest "...since there are likely significant vapour pressure differences for the products of different chamber experiments and there are also likely dependencies of wall loss rates on species vapour pressures.

Changed

Line 750-764. This discussion is interesting and important for all chamber systems. Are there references to studies that could make this issue more quantitative? Does the EUROCHAMP consortium have information about differences in SOA yields from VOCs in chambers with differing surface-to-volume ratios and wall composition? It seems that this discussion could be more than it is "unquantifiable". Important information may exist that can be used in this study.

The referee makes an important point. There has been significant effort in the EUROCHAMP consortium to start unravelling these problems, though currently without full resolution. Once such information has been fully processed and considered, it may indeed be possible to revisit our results. The data are not complete in terms of the possible dependencies or wall materials, but should help the quantification. There are MAC data within the EUROCHAMP dataset and these are broadly in line with the other Teflon chambers. Unfortunately, there are few data across the consortium for components in the vapour pressure range of most interest and there is still significant scatter in the data that do exist. These are under active scrutiny at the moment.

Line 765. Need to define "single value yields".

This is used as opposed to full yield curves at a single value of total particle mass or VOC consumption. It is expanded in the text.

Line 768. Why is "predictions" in quotes?

Quotes removed

Line 770. Suggest "...that such predictions are valid." (use present tense) Line 771. Suggest "...the mixed yield depends on the question...".

Both changed

Line 775. Suggest "...initial isoprene concentration studied and hence...".

Changed

Line 779. Suggest changing "reference" in this context (the single VOC experiments) to something else (e.g. "base case" or "constraint"). The term "reference" could be confusing since it is used for previous related published papers.

OK, changed to "baseline comparator".

Line 780. Suggest removing the parenthesis in "...shown in Figure 5b, the..."

Changed

Line 781. Suggest coming up with a term to describe the SOA yield from α-pinene / isoprene experiments that does not include the change in isoprene concentration, since the normal approach to calculated yield is the ratio of the increase in SOA divided by the decrease in VOC. Developing terminology for this case is preferable to stating "excluding isoprene in the calculations". One could imagine using Y (for yield) with various superscripts or subscripts to indicate the various cases, for example $Y^{pin,isop}$ *and* $Y^{pin,isop}$ *could (1) indicate the SOA yield from a* $_{pin,isop\ pin}$ *binary α-pinene / isoprene system (superscript) using the loss of both compounds in the calculation (subscript), while (2) could indicate the SOA yield from a binary α-pinene / isoprene system (superscript) using the loss of α-pinene only in the calculation. Just a thought.*

This is interesting and in direct opposition to the contention of referee 1 who states that it is unimportant whether consumption of a VOC in a mixture is included or not if it doesn't contribute particle mass. We have adopted a similar formalism to that suggested in the revised manuscript to reflect these cases.

Line 813. The question of whether the inorganic aerosol mass should be somehow included in the yield is important. Has anyone done multiple experiments (say with α-pinene) under identical conditions with varying amounts and identities of inorganic seeds, perhaps also including different size distributions? This seems to be a critical part of performing such experiments in the laboratory chambers. If such work exists, suggest referring to it here and adjusting the discussion accordingly.

We are not aware of such experiments. It is not completely straightforward to capture all possible dependencies and there are measurement considerations for different components, but it should be possible.

Lines 819-20. Suggest "...composition is an important topic for a future study." Line 823. Suggest "...chemical composition would provide important...".

Both changed

Line 835. It Is not clear what is meant by the phrase "from the beginning of the experiment". Suggest removing it.

Clarified.

Line 840. In the discussion of which process could form the substances that are observed on the filters collected in ternary experiments, it should also be mentioned that there are known processes of oxidation of surface aerosol substances by gas-phase oxidants, and also condensed-phase chemical oxidation. Addition of one or more references here would be helpful to the reader.

Such processes have indeed been recognised for some time and we now refer the reader to Liu et al. (2014) and Turpin et al. (1994)

Line 843. Should the term "ternary" be added to "...only found in the ternary mixed system."?

No, they are found in a number of mixtures; the sentence has been revised for clarity.

Line 846. Remove right parenthesis on "Figure 10a".

These are now in the SI and referred to accordingly

Line 849. Suggest "...there are confounding differences..."

Changed

Lines 851-2. Are there composition data from the FIGAERO-CIMS during the experiments (not just at the end)? If so, consider a way to present these data, too.

The data shown are from FIGAERO-CIMS and panel b shows the different time-series behaviours of $C_7H_7NO_4$ between experiments. The measurements presented here are indicative and full time-resolved measurements of the FIGAERO-CIMS are presented in companion papers (Shao et al., 2022b, Du et al., 2022 and Du et al., 2022, in prep).

Line 861. Suggest "...which expresses the FIGAERO-CIMS...".

Changed

Line 862. Suggest "...in the mixture and in the single...".

Changed

Line 885. This reviewer does not think the word "certainly" is appropriate here. It is conceivable, possible, and even highly likely that scavengers could influence the oxidation product distribution, but it is not certain until the appropriate experiment has been performed. Suggest rewording.

Agree that this has yet to be proven, so changed

Line 886. Suggest "...secondary oxidant formation occurs in the real...". While this is a true statement, it does not necessarily justify that initial mixed VOC experiments must be conducted in mixed oxidant environments. It is better to conduct the mixed VOC experiments with single oxidants first and then graduate to mixed oxidant situations.

We would contend that this depends on the objectives of the experiment. A phenomenological demonstration of the importance of full complexity vs individual oxidant system is completely possible. We agree that resolution of a specific oxidation process requires the approach stated. There is value in both approaches and inability to conduct an "ideal" experiment should not preclude collection of useful information from less "clean" systems.

Line 888. It is not true that high peroxide concentrations are required in experiments that use peroxide as an OH precursor. The peroxide can be added constantly to maintain a relatively low steady-state level. This is actually preferred because OH reacts rapidly with hydrogen peroxide. Suggest rewording.

Thanks for the advice. Reworded.

Line 892. Suggest "...may increase the OH : O_3 ratio such that the OH reaction pathways dominate...".

Changed

Line 897. The statement "...maybe more suited to targeted laboratory studies than to chamber experiments" is confusing since chamber studies are target laboratory studies. Suggest different wording for "targeted laboratory studies" such as "flow tube kinetic studies" or something similar.

Point taken. Reworded.

Line 904. This sentence is confusing because it uses "resolution" and "temporally resolved" together. One needs to be clear whether resolution refers to temporal or mass resolution (as in mass spectrometry). Suggest rewording.

Again – sloppy language. Changed

Line 906. Suggest "...reactor experiments to study multiple steady states...". The authors should also consider more detail in what is meant by multiple steady states, along with references.

We were referring to the sorts of studies conducted in McFiggans et al. (2019). Changed

Line 909. Suggest "...thought it should be recognized that oxidation and SOA formation occur during...".

Changed

Line 910. It is not clear what is meant by "mixed night-time oxidation by NO_3". Also, suggest "...should be considered" instead of "should not be forgotten."

We should have stated "in mixtures" rather than "mixed". Changed.

Line 915. Suggest "...to their interpretation, several important observations..."

Changed

Line 917. It is not clear what is meant by "photochemical trajectory". Suggest defining somewhere with discussion to amplify its importance in the present study.

OK, this is referenced back to Figures 1 and S1 and expanded.

Line 925. Suggest "...existence and to quantify any interactions affecting the observed SOA mass and yield."

Changed

Lines 928-9. Here is "straightforward" used in two sentences in a row. Suggest rewording.

Reworded

Line 931. This reviewer is not sure the term "reference point" is the best. Suggest rewording.

Changed to "baseline comparator"

Line 939. See earlier comment about "unique-to-mixture".

"Uniquely found in the mixture" is now used in the revised caption of the figure, now found in the SI.

Line 940. Suggest "...of the particles depends on the rates...".

Changed

Line 942. Suggest "...for our understanding of atmospheric...".

Changed

Line 943. Suggest "...the interpretation is complex, and both the experimental design and evaluation need to be...".

Changed

Figure S4 caption. Suggest "...in the single precursor α-pinene and...". Also, why is the x-axis scale of the plot logarithmic? The symbols are very similar and hard to distinguish. Suggest changing to make clearer.

We have changed as well as enlarged the size of the symbols in the updated Figure S4 (see below) to improve it's clarity. The x- axis is logarithmic to enable the low mass points to be distinguished which is not possible on a linear scale.

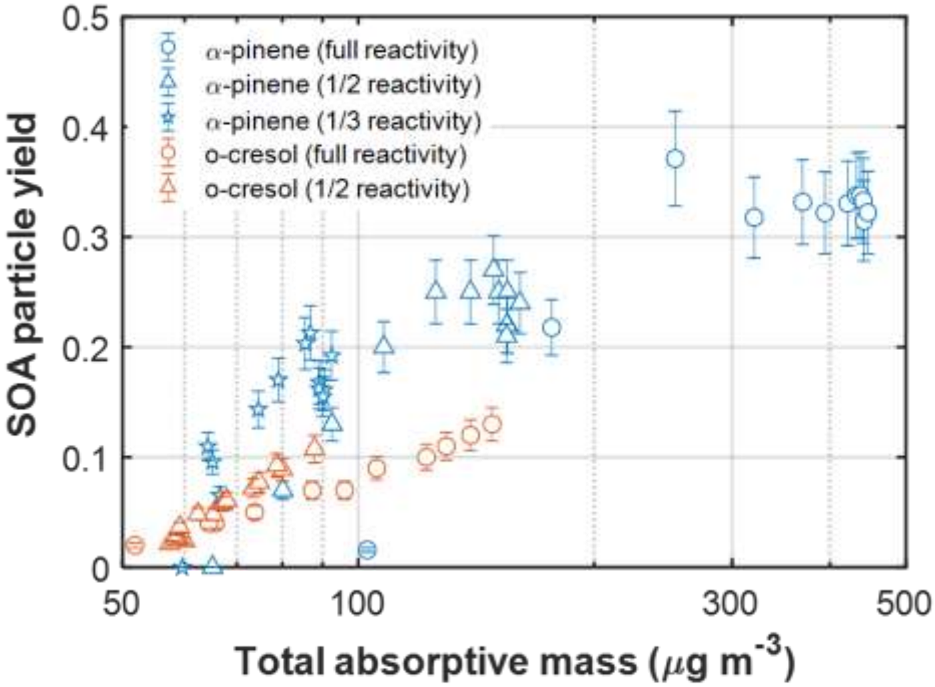

Figure S4: SOA particle mass yield as a function of total absorptive mass, including the remaining inorganic seed mass, in the single precursor α-pinene and o-cresol experiments at all initial concentrations. Error bars represent the propagated uncertainties in all measurements and in the particle wall loss corrections applied.

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