Response to Referee #2

Thanks very much for your time reviewing our manuscript entitled “The critical role of oxygenated volatile organic compounds (OVOCs) in shaping photochemical O_3 chemistry and control strategy in a subtropical coastal environment”. We are very grateful to the reviewers for their valuable and helpful suggestions for our manuscript. We have made all the suggested changes and clarifications. We believe that the manuscript has been significantly improved based on those suggestions.

Our point-by-point responses to reviewers’ comments are as follows. We repeat the comments raised by the reviewers in *black italic* font and give our replies in the indent and normal font, and with the revised text in blue. The line numbers mentioned in responses correspond to the revised manuscript.

Evaluation: *“The critical role of oxygenated volatile organic compounds (OVOCs) in shaping photochemical O3 chemistry and control strategy in a subtropical coastal environment” presents a comprehensive and well written box modelling study of the drivers of photochemical O3 formation at a subtropical coastal region in South China, particularly highlighting the importance of including a detailed representation of oxygenated VOCs (OVOCs) in the modelling study. An OVOC sensitivity study is presented, including model results with and without detailed OVOC inclusion, relevant to the broader photochemical O3 production community. I would recommend this manuscript for publication, provided a few further questions are addressed.*

Response: We appreciate the positive comments from the reviewer. We have revised the manuscript and addressed the issues according to the reviewer’s comments.

1: *The authors present two case studies: Box modelling with and without the inclusion of OVOCs, and the impact this has on modelling photochemical O3 production. However, in reality, it is more typical for studies to include a subset of OVOCs (though not 63, as is the case in your study), since many of these species can be measured using GC techniques alone. It would be interesting to know how good a job your box modelling does if just a “common” subset of OVOCs is incorporated (e.g. acetone, methanol, acetaldehyde). Does this subset sufficiently describe the ozone production regime? Or is it necessary to include a much more comprehensive suite of OVOCs to fully describe the chemistry? It would be worth taking a look at some literature to see which OVOCs are typically included in modelling studies for this analysis. Some examples: Whalley et al., 2018, Whalley et al., 2021, Nelson et al., 2021. In the absence of re-running the model using a hybrid between no OVOCs and 63 OVOCs, perhaps the authors already have enough information from their current model study to comment on which OVOCs are particularly important to include?*

Response: We thank the reviewer for the insightful comment regarding the representativeness of OVOCs in photochemical modelling. Based on our review of previous modeling studies and comparison with OVOCs measured by the PTR in this work, we found that typical model inputs of OVOCs often include species such as acetaldehyde, acetone, MEK, and butanal (Whalley et al., 2021; Whalley et al., 2018; Yang et al., 2018; Feng et al., 2023; Shen et al., 2021). To assess whether this subset sufficiently captures the role of OVOCs in O_3 and RO_x radical production, we conducted an additional sensitivity case in which only these four OVOC species were included in the constrained inputs. We then compared the production rates of O_3, OH, HO_2, and RO_2 with the cases of full OVOCs constraints and without OVOCs constraints. The results show that with only these four OVOCs led to an increase of 12.5%-23.9% in daytime O_3 and

RO_x radical production rates compared to the case without OVOCs constraints, accounting for around 60% of the underestimation compared to considering all measured OVOCs. This indicates that the significant underestimation of O_3 and radical production rates without OVOCs constraints cannot be explained solely by the common subset of OVOC species. In other words, approximately 33.9%-36.5% of photochemical production potential arises from a broader range of OVOCs beyond the commonly OVOCs. This highlights the importance of incorporating a more comprehensive suite of OVOCs in atmospheric chemical modeling for accurately representing O_3 formation and radical chemistry in the atmosphere. We have added this new sensitivity case to the revised manuscript to strengthen the discussion on the importance of more comprehensive OVOCs in photochemical modeling.

The revised text reads:

L517-L522: A sensitivity test considering only four simple OVOCs including acetaldehyde, acetone, MEK, and butanal (Whalley et al., 2021; Whalley et al., 2018; Yang et al., 2018; Feng et al., 2023; Shen et al., 2021) resulted in an increase of 12.5%-23.9% in daytime O_3 and RO_x production rates than case without any OVOC constrains. This indicates that nearly 33.9%-36.5% reduction cannot be explained by these simple OVOCs alone, further highlighting the importance of comprehensive OVOCs in modeling.

2: *Line 95 – Is it correct to say that summer is Sept 4 – Oct 12, autumn is Oct 13 – Dec 1, and winter is Dec 2 – Dec 20 in this region? This is quite a short timeframe to cover these seasons. How representative do you feel these windows are of the "summer", "autumn", and "winter" in South China?*

Response: We appreciate the reviewer's comment. Since Hong Kong is located in a subtropical region where seasonal weather patterns are influenced by the Asian monsoon system. As a result, the direction of upper-level winds is commonly used to characterize seasonal transitions in this region. In this study, the seasonal classification follows the approach adopted in previous studies (Feng et al., 2023), which is based on the occurrence of synoptic-scale events and abrupt changes in key meteorological parameters, including upper-level wind direction, sea-level pressure, and dew point temperature. The temporal variation of upper-level wind direction, sea level pressure and dew point in Hong Kong measured by Hong Kong Observatory Station from July 2021 to March 2022 was shown in Figure S1. A sudden increase in sea-level pressure accompanied by a notable decrease in dew point on 12 Oct 2021 indicated intrusion of relatively cold air masses, making the transition from summer to autumn. During the summer period, upper-level winds were predominantly easterly and/or southeasterly, reflecting typical monsoonal circulation patterns (Wong et al., 2022). Similarly, the transition from autumn to winter was characterized by abrupt changes in both dew point and upper-level wind direction, with the characteristics of cold high-pressure climate. Moreover, in winter, upper-level winds were primarily westerly and/or northwestly, consistent with the influence of the East Asian winter monsoon (Wong et al., 2022; Li et al., 2016). Therefore, the seasonal division and selected period in this study is considered representative of the characteristic features of summer, autumn, and early winter.

The revised text reads:

L96-L99: Seasonal classification in this study was based on the occurrence of synoptic events and abrupt changes in key meteorological parameters, including upper-level wind direction, sea-level pressure, and dew point temperature (Figure S1), as detailed in our previous studies (Feng et al., 2023).

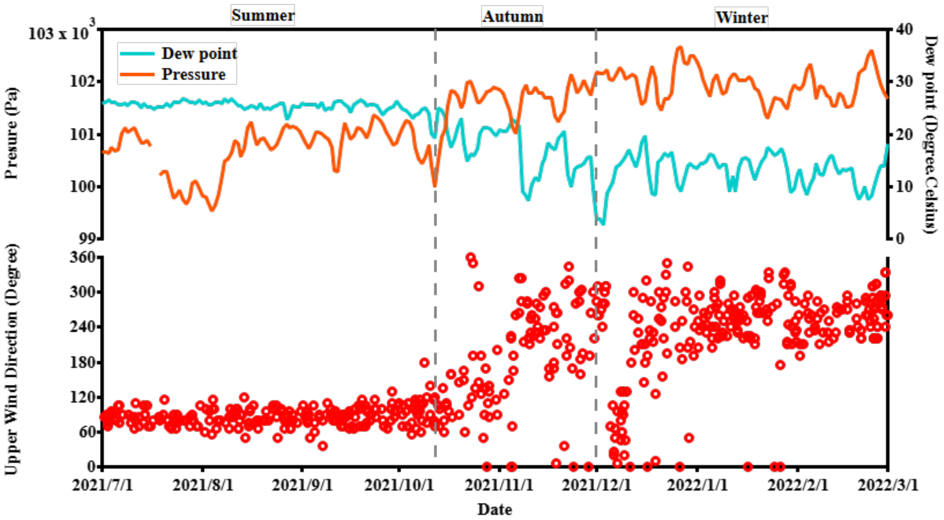

Figure S1. Temporal variation of upper-level wind direction, sea level pressure and dew point in Hong Kong measured by Hong Kong Observatory Station from July 2021 to March 2022. The seasonal transition from summer to early winter was characterized by a rapid shift from high dew point and low sea-level pressure to cold and high-pressure systems, accompanied by a change in upper-level wind direction from easterly/southeasterly to westerly/northwesterly (Li et al., 2016; Wong et al., 2022).

3: *Line 135 – We "attempted" to assign signals based on likely contributors, and that your quantification of OVOCs is semi-quantitative for uncalibrated species, is an honest and fair account of what you have done here. Whilst I completely understand that this must be the case due to instrument limitations, some readers may feel a little mis-sold up to this point, as the abstract implies that we are to expect 63 explicitly measured and quantified OVOCs. Perhaps you could be more upfront in the abstract on what you have done to describe the OVOCs, as this is a major component of your study. Even if you just said "X quantified and Y semi-quantified OVOC species", rather than 63 quantified (abstract – line 22).*

Response: Thanks for the valuable suggestion. We have revised the 63 quantified OVOC species to 8 quantified and 55 semi-quantified OVOC species in the abstract and the manuscript.

L175-L178: Given these limitations, our quantification of the 63 OVOCs measured by PTR should be considered as semi-quantitative for 55 uncalibrated species, and as high-confidence for the 8 calibrated species.

4: *Line 160 – I was struggling to follow exactly what you've done here. "VOC species from daytime canister samples were linearly interpolated to hourly resolution for the model input". Is this just the VOCs measured using the GC-MS/FID/ECD? I assume the PTR measurements are online? Please be clear on the time resolution of the measurement of both things. How often were canister samples taken? Is interpolating the data to hourly resolution appropriate for this measurement resolution?*

Response: Thanks for the valuable comment. We apologize for the confusion and have revised the manuscript to clarify the data sources and their temporal resolutions. Please find our responses below:

The VOC species used for linear interpolation were measured using offline canister sampling and analyzed by GC-MS/FID/ECD. These measurements primarily included 38 species including alkanes, alkenes, alkynes, and alkyl nitrates, which were used as input for model simulations. These canister samples were collected every 3 hours from 09:00 to 18:00 LT during the selected polluted days. Given the limited temporal resolution of the offline sampling method, a linear interpolation approach was applied to estimate hourly concentrations during the daytime, following methodologies adopted in previous studies (Yang et al., 2018). While the nighttime concentrations were estimated using linear regression relationships with continuously measured tracers obtained from PTR-ToF-MS measurements. In contrast, continuous online measurements of ambient VOCs were conducted in this study using PTR-ToF-MS, with a time resolution of 10 seconds. However, we opted not to interpolate the offline data to a resolution finer than one hour, as doing so would likely introduce greater uncertainty and potentially compromise the accuracy and reliability of the model simulations.

The revised text reads:

L103-L105: A PTR-ToF-MS (Ionicon Analytik GmbH, Innsbruck, Austria) with H_3O^+ as the primary reaction ion was used to measure the gaseous VOC and OVOC species with high time resolution of 10 seconds during the whole field campaign.

L179-L180: In addition, canister samples of VOCs were collected every 3 hours from 09:00 to 18:00 LT in three seasons.

L202-L210: For the offline canister VOC samples measured by GC-MS/FID/ECD, daytime data from 9:00 to 18:00 were linearly interpolated to an hourly resolution for the model input (Yang et al., 2018), while nighttime concentrations of unmeasured C_2-C_{10} hydrocarbons (excluding isoprene and monoterpenes) and alkyl nitrates were estimated using linear regression relationships with continuously measured hydrocarbons (e.g., C_3H_6, C_5H_{10}, C_6H_{10}) and nitrophenols obtained from PTR-ToF-MS measurements. The PTR measured species used in the linear regression calculation were selected based on their strong correlations with corresponding compounds in the canister data to ensure more reliable estimates.

5: *225 – A lot of effort has been put into speciating the OVOCs, but what about the biogenic species? The authors state there were measurements made of 2 biogenic species – isoprene, and monoterpenes. However, there are only 3 monoterpenes in the MCM (the pinenes and limonene), and they later describe their monoterpene measurement as the pinenes only. As these are highly reactive species, it is important to discuss how your assignment of the total monoterpene measurement to (what I assume is 50:50?) a- and b- pinene. There are also many monoterpene species not included in the MCM, some with faster reaction rates than these species. I understand the need for your assumption to be made, but please acknowledge the potential implications this has in the text.*

Response: Thanks for the valuable suggestion. As noted by the reviewer, biogenic species are highly reactive and play a critical role in atmospheric photochemical processes. In this study, biogenic species measured by PTR-ToF-MS mainly included C_5H_8 and $C_{10}H_{16}$. $C_5H_8H^+$ signal mainly corresponds to isoprene and could also be interfered with fragments from higher-carbon aldehydes and cycloalkanes, while $C_{10}H_{16}H^+$ signal primarily corresponds to various kinds of monoterpenes, such as pinenes, limonene, camphene, and so on (Coggon et al., 2024; Claflin et al., 2021; Yuan et al., 2017; Zhang et al., 2025). Moreover, the interferences of fragmentation and the distributions of isomers may vary with altitudes and environmental conditions influenced by both biogenic and anthropogenic emission sources, which makes the isomer distribution more complex and introduces large uncertainties (Coggon et al., 2024). However, due to the limitations of PTR-ToF-MS, accurate quantification of isomers cannot be achieved at present. Therefore, assumptions regarding the distribution of isomers and potential fragment interferences were made based on previous studies using the gas chromatography pre-separation. For the attribution of C_5H_8 signals, isoprene was allocated the fraction of 63% reported in previous studies employing PTR-MS measurements coupled with GC, which effectively minimizes interference from fragments of higher molecular compounds (Koss et al., 2018). While for the distribution of $C_{10}H_{16}H^+$ signals, based on previous studies, α- and β-pinene generally accounted for the most to $C_{10}H_{16}H^+$ signals in ambient air, with α-pinene as the most contributor (Kim et al., 2009; Byron et al., 2022; Kammer et al., 2020). Therefore, in this study, as the reviewer pointed out, we allocated equally between α-pinene and β-pinene at a 50%:50% ratio as the assumption for model simulation. α- and β-pinenes are highly reactive species which can rapidly react with atmospheric oxidants such as OH radicals, however, their reactivity remains lower than that of certain other $C_{10}H_{16}$ species, such as limonene (Bouvier-Brown et al., 2009). Therefore, simplifying the assumption of representing all $C_{10}H_{16}$ solely by α- and β-pinenes may introduce some uncertainties in the simulation of atmospheric photochemical processes, particularly affecting the simulation of radical budgets and O_3 production. Moreover, the absence of other highly reactive monoterpenes and their associated oxidation mechanisms in the MCM may further compromise the accuracy of photochemical simulations. Incorporating a more comprehensive representation of monoterpene chemistry is thus essential for improving the reliability of future atmospheric modeling efforts.

The revised text reads:

L169-L174: C_5H_8 may be affected by fragment interferences from higher-carbon aldehydes and cycloalkanes (Coggon et al., 2024; Claflin et al., 2021; Yuan et al., 2017; Zhang et al., 2025), therefore, the attribution of C_5H_8 to isoprene follows the proportion of 63% reported in previous studies employing PTR-MS coupled with GC pre-separation, which effectively minimizes interference from fragments of higher molecular compounds (Koss et al., 2018).

L152-L155: For example, for $C_{10}H_{16}$, given that α-pinene and β-pinene are typically the predominant contributors (Kim et al., 2009; Byron et al., 2022; Kammer et al., 2020), an equal 50:50 allocation between the two species was adopted as a modeling assumption for the apportionment.

L280-L285: BVOCs refer specifically to C_5H_8 and $C_{10}H_{16}$, which primarily correspond to isoprene and various monoterpenes (e.g., pinenes, limonene, camphene, etc.), respectively. It is important to note that C_5H_8 may be affected by fragment interferences from higher-carbon aldehydes and cycloalkanes (Coggon et al., 2024; Claflin et al., 2021; Yuan et al., 2017; Zhang et al., 2025), which may potentially lead to an overestimation of the contribution of BVOCs, particularly isoprene.

L369-L372: Nevertheless, due to the inherent limitations of PTR-ToF-MS, accurate quantification of isomers with distinct chemical reactivities remains challenging, introducing some uncertainties in atmospheric photochemical modeling.

6: *300 – Just to reiterate my early point, the authors say your measurements are high-resolution here, but the resolution needs to be stated more explicitly earlier in the text.*

Response: Thanks for the reviewer's comment. We have added a description of the 10-second time resolution used in the PTR-ToF-MS measurement to the Method Section (**Lines 104-105**).

7: *344 - More discussion of the implications of BVOCs in your model here – again, worth pointing out that this result is based on the monoterpenes being split between the pinenes only.*

Response: Thanks for the valuable suggestion. We have added more discussion of the impacts of BVOCs on model simulations.

The revised text reads:

L350-L355: It should be noted that current photochemical models typically represent monoterpenes using only α-and β-pinenes, neglecting some highly reactive species such as limonene. Moreover, gaps in the MCM, such as the absence of certain highly reactive monoterpenes and associated oxidation pathways, may further introduce uncertainties in assessing the role of BVOCs in atmospheric photochemistry.

8: *397 – The authors say that including the OVOCs means that observed O3 was successfully reproduced. Are the observed and model concentrations identical? I would expect some impact from transportation to play a role here, since not all the observed O3 can be expected to be photochemically produced in situ. Please discuss.*

Response: Thanks for the reviewer's comment. We apologize for the unclear clarity in our previous explanation. Our main point is that incorporating a broader range of OVOCs into the model will lead to a relatively improved representation of O_3 formation compared to simulations including only a limited set of OVOCs or without OVOC species. However, this improvement does not imply a complete agreement between modeled and observed O_3 concentrations. As the reviewer pointed out, the model captures only in situ photochemical processes, whereas ambient O_3 levels are also influenced by other factors, such as regional transport. Therefore, exact agreement between modeled and observed O_3 concentrations is not expected. Indeed, as shown in the time series of comparison (Figure S13), there are many days on which the simulated O_3 levels are lower than the observations, even with the inclusion of broader OVOCs in the modeling. This discrepancy is particularly evident during the autumn and early winter, when the influence of Asian monsoon is strong, and during the nighttime when photochemical reactions are minimal.

The revised text reads:

L470-L472: Incorporating a broader range of OVOCs improved the simulation of O_3, particularly in autumn and early winter, where daytime concentrations were underestimated by 26.5% and 35.7%, respectively, without OVOCs constraints.

L474-L478: It should be noted that the model considers only in situ photochemical processes and does not include influences such as regional transport. As a result, discrepancies between observed and simulated O_3 remain, especially in autumn and early winter, when periods typically influenced by the Asian monsoon, and during nighttime when photochemical activity is minimal.

9: *Line 444 – "our results highlight that many other OVOCs,…,remain overlooked", and line 451 "key OVOC species such as methanol, acetaldehyde, and acetone,…, were underestimated". These key OVOCs are more typically incorporated into box modelling studies in the literature, as they can be measured using GC techniques (see my main suggestion earlier in this review).*

Response: We thank the reviewer for the valuable comment and apologize for the lack of clarity in our original description. What we intended to emphasize is that, under the scenario without OVOC constraints, the primary reason for the underestimation of O_3 and RO_x production is the substantial underestimation of OVOC concentrations in the model simulation. The reference to "methanol, acetaldehyde, and acetone being underestimated by 73%-99%"

was intended merely as an example to illustrate this issue. Our intention was not to imply that these compounds are absent from other models reported in the literature.

The revised text reads:

L525-L528: These discrepancies were largely due to the underestimation of multiple OVOC species, for example, methanol, acetaldehyde, and acetone were underestimated by 73%-99% in early winter simulations without OVOC constraints (Table S5).

10: *Line 494 – When you vary the VOCs and NOx for the isopleth analysis, do you also vary OVOCs? The difficulty here is that some of your OVOCs will be formed photochemically, and some from primary sources, meaning the two will not necessarily ever decrease or increase uniformly. Do you also reduce biogenic species, or just anthropogenic? How do you navigate this issue to make the isopleth findings relevant from a policy perspective?*

Response:

We thank the reviewer for raising this important point regarding the treatment of OVOCs in O_3 isopleth analysis and its implications for policy relevance. We acknowledge that the diverse sources of OVOCs (including both primary emissions and secondary formation) present a significant challenge in accurately representing their variability in sensitivity analysis. Due to the current lack of a more robust framework to account for the dynamic behaviors of OVOCs under different emission scenarios, we adopted a simplified approach in this study. Specifically, with OVOCs constraints, changes in OVOC concentrations were scaled proportionally to the variations in their precursor VOCs. This assumption reflects a linear relationship between precursor VOC levels and secondary OVOC formation, providing an approximation of their response to emission changes. While we recognize that this approach does not fully capture the nonlinear chemistry involved in secondary OVOC production, it offers a pragmatic representation of their influence on ozone formation under varying emission conditions. We consider this a reasonable interim approach, pending further advancement in the mechanistic understanding and modeling of OVOC chemistry. And we added relevant clarification of OVOCs in this section.

Regarding biogenic VOCs, we applied uniform scaling to both anthropogenic and biogenic VOCs in the isopleth analysis to explore the full spectrum of chemical regimes and assess O_3 formation sensitivity under varying precursor conditions. However, emissions of biogenic VOCs are predominantly driven by natural processes and are not readily amenable to direct control. Therefore, scenarios involving reductions in biogenic VOCs are not intended to reflect realistic policy interventions, but serve as sensitivity tests to better understand the chemical regime transitions and the role of biogenic VOCs in O_3 production. For policy-relevant interpretation, we place emphasis on anthropogenic VOC reductions, which are more feasible and actionable from an emissions control perspective.

The revised text reads:

L571-L575: It should be noted that due to the absence of a robust mechanism to represent the nonlinear formation and diverse sources of OVOCs, we employed a simplified scaling approach based on precursor VOCs in O_3 isopleth analysis. Despite inherent uncertainties, this provides a practical approximation for assessing OVOC impacts on O_3 formation under varying emission scenarios.

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