Response to Reviewer #1:

We gratefully thank the editor and all reviewers for their time spent making their constructive remarks and useful suggestions, which have significantly raised the quality of the manuscript and have enabled us to improve the manuscript. Each suggested revision and comment, brought forward by the reviewers was accurately incorporated and considered. Below are the comments of the reviewers and response point by point and the revisions are indicated. We use different colored fonts to distinguish between responses to reviewers and the revised sections of the manuscript.

1. Responses to reviewers are highlighted in blue.
2. *Revised sections of the manuscript are highlighted in red.*

Comment 1: This paper usefully demonstrates the impacts of updating biogenic VOCs in urban environments (called here UGS-BVOC) as well as updating land use cover data to high resolution maps (called here UGS-LUCC). The impact of each of these changes separately and together on ozone in Guangzhou, China is thoroughly explored. These emissions and model improvements are clearly important for accurately simulating ozone in this region. I recommend publication after major revisions specified below.

Reply: We sincerely thank the reviewer for their positive assessment of our study and for recognizing the importance of updating biogenic VOC emissions (UGS-BVOC) and high-resolution land use cover data (UGS-LUCC) in accurately simulating ozone in urban environments. We are pleased that you found our exploration of their separate and combined impacts on ozone in Guangzhou to be thorough and valuable. We will carefully address the specific concerns raised to further improve the manuscript and meet the standards required for publication. Thank you again for your constructive feedback and support.

Comment 2: While VOCs are important for ozone production, so are nitrogen oxides. This paper could be mis-leading to policy makers if NO_x is not also mentioned as an ozone precursor more clearly in the text.

Reply: Thanks for this valuable suggestion. We have demonstrated the O_3 formation from NO_x and VOCs in the "Introduction" Section and added some discussion about the impact of NO_x levels to the "Uncertainties and Limitations" (Section 4).

Introduction:

Surface O_3 is generally formed through chemical reactions of VOCs and NO_x in the presence of sunlight.

Section 4:

Finally, Guangzhou, the study area, is a highly urbanized Chinese metropolis with a VOC-limited region (Gong et al., 2018; Kai et al., 2011; Liu et al., 2021). As a result, even a relatively small amount of VOC

emissions, such as those from UGS-BVOC, can significantly impact ozone concentrations. Therefore, policymakers in Guangzhou should prioritize addressing the role of UGS-BVOC emissions in air pollution prevention and control. In other cities, particularly those with advanced urban development, high NO_x emissions—often resulting from factors like high motor vehicle ownership—can lead to VOC-limited conditions. In such areas, it is equally important to emphasize the role of UGS-BVOC emissions in ozone pollution. In contrast, cities with lower NO_x emissions identified as NO_x-limited regions may experience minimal impact from UGS-BVOC emissions on ozone concentrations.

Comment 3: Why does the abstract and throughout the text use ug/m^3 as the units for gases in the atmosphere. The convention is typically ppb in atmospheric chemistry. Please provide a valid reason or use the conventional unit. Additionally, throughout the paper the authors switch from ug/m^3 and ppb throughout the text and are not consistent with one unit. This is confusing for the reader. I would highly recommend switching all units to ppb for all gases.

Reply: Thank you for the nice suggestion, and we have converted all units to ppb for all gases.

Comment 4: The use of urban as both an "urban region" in Figure 1 and "urban landcover type" to mean two different things makes the paper hard to understand at first. This paper selects locations classified with the urban landcover type as urban and this is called UGS (urban green space), but then also classifies by region into 3 types called urban, suburban, and rural. Providing more detail on how the regions are determined in Figure 1 and lines 202 – 206 would be useful. And please consider calling this something other than "urban", so that readers can more easily differentiate when the authors are referring to urban as a region and or as a landcover type. Perhaps, terminology like "city center", "suburban", and "rural" could work.

Reply: Thank you for your helpful suggestion. We have revised the region definition by replacing "urban" with "city center" as recommended.

Comment 5: There is very little comparison to isoprene observations even though this is the major development in this paper (Table 2 and line 235). It is an excellent opportunity that the model can be compared against these two monitoring sites for isoprene. For reader, clarity, can you please add these two monitoring sites to Figure 1? Looking at the average isoprene concentrations over the entire campaign (Sept – Nov) is not sufficient proof that the isoprene emissions have improved. Also I am confused on the time periods. The observations appear to be for Sept 20 – Nov 20, but in the methods section you state the model is only run for September 1 – Sept 30 (ignoring spin-up). Are you comparing the observations and model during the same time period? It is very important to compare the model and observations over the exact same time period since isoprene emissions are very seasonally dependent. Can you provide additional statistics like you do for the

meteorological variables in Table S2 and ozone and NO_2 in Table 3? Further evaluation of the diurnal cycle in the model compared to the observations and statistics for the different months would also be very useful in understanding whether the isoprene emissions have improved in the model. Since your main updates are isoprene emissions it is very important that this evaluation is clear on what has been done and that this is a good evaluation to ensure that the updates the author has made are robust.

Can you also provide more information on how the observations were collected at these monitoring sites? What instrument technique? Were any interferences considered if the instrument technique was a PTR (Coggon, et al., 2024 - https://doi.org/10.5194/amt-17-801-2024)? Was the diurnal cycle of the observations consistent with known chemistry of isoprene where isoprene concentrations rise during the day and fall rapidly at night in high NO_x urban locations with significant amounts of NO_3 radical? This can help you verify that any interferences for isoprene in your instrument technique are appropriately accounted for.

Reply: Thanks for this comment and we have added the ISOP monitoring sites to Figure 1.

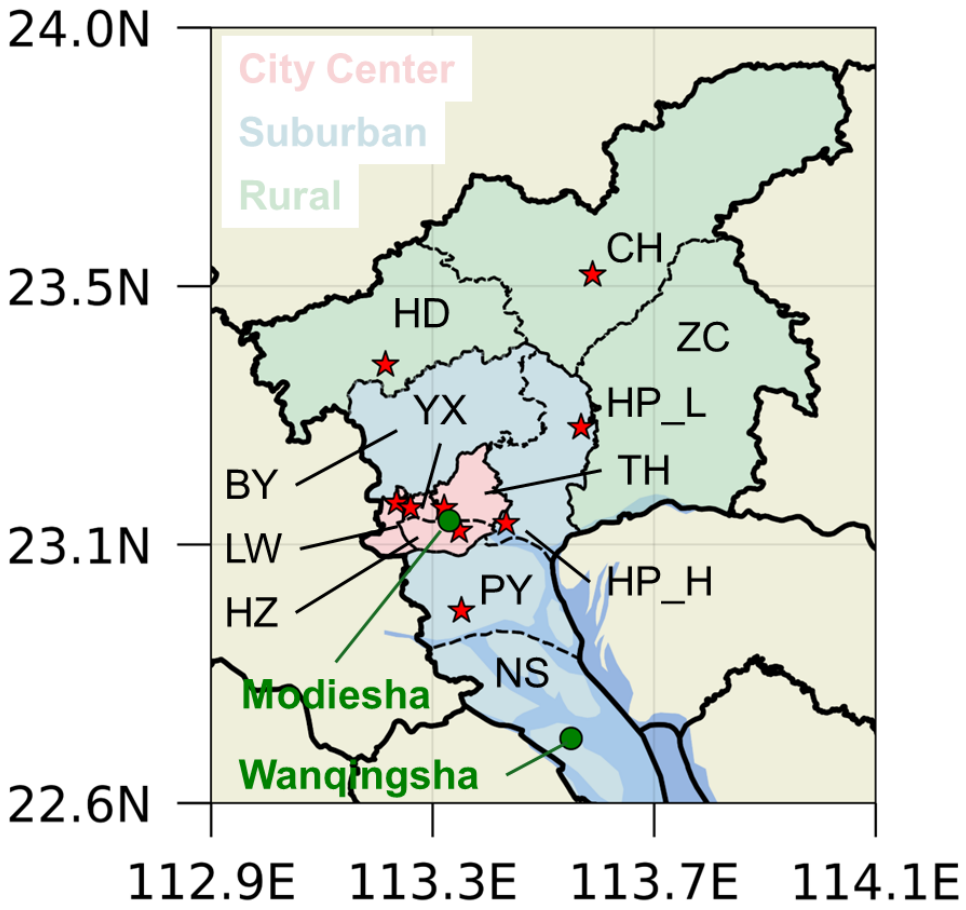

Figure 1 The innermost domain of WRF-CMAQ with various areas and the air quality station locations map. Modiesha and Wanqingsha are the observation sites for isoprene.

In this revised manuscript, thanks to the contribution of Prof. Senchao Lai's groups, we have got the hourly isoprene observations at Modiesha and Wanqingsha sites for a more detailed validation. We have listed his group members as co-authors of this revised study. The observed isoprene concentration shows a reasonably feature that is high during the day and low at night. We have compared the observation and model simulation

during the whole September, which have the same time period as the model simulation. Below please find the revised validation results for isoprene.

BVOCs are the major sources of ISOP and monoterpene (TERP), rendering the assessment of their concentrations a pivotal method for indirectly verifying the accuracy of BVOC emission estimates. Table 2 delineated within this study presents the mean concentrations of ISOP derived from various cases juxtaposed with the observed average concentrations. This comparative analysis in the Modiesha site reveals that after the incorporation of the UGS-BVOC emissions, there is an augmentation in the ISOP concentration from 0.29 to 0.35 ppb and from 0.23 to 0.29 ppb under distinct land use cover cases (Gdef and Ghr), relative to an observed concentration of 0.34 ppb. Meanwhile, the evaluation at the Wanqingsha site, where the observed mean ISOP concentration was 0.45 ppb from September 7 to September 30, 2017, shows that the modeled ISOP concentrations increased from 0.29 to 0.31 ppb and from 0.27 to 0.29 ppb under distinct land use cover cases (Gdef and Ghr) when UGS-BVOC emissions were included. This increment signifies a substantial diminution in the discrepancy between the modeled and observed concentrations attributable to the UGS-BVOC emissions. Analogously, the integration of the UGS-BVOC emissions yields a refinement in the estimation accuracy of ISOP concentrations at the Modiesha site, as evidenced by a reduced bias.

These findings reveal that ISOP concentrations are underestimated by 16.4% and 34.7% in the Modiesha and Wanqingsha sites when UGS-BVOCs are excluded, respectively, suggesting the important role of UGS-BVOCs emissions in modeling. Moreover, numerous studies highlight the significant role of ISOP in O_3 formation within the Pearl River Delta (PRD) region, including Guangzhou. For instance, Zheng et al., (2009) demonstrated that ISOP has the highest ozone formation potential among all VOCs. Therefore, incorporating UGS-BVOCs into ISOP concentration estimates is crucial for accurately modeling regional O_3 levels.

Table 1 The evaluation results for the monthly mean ISOP concentrations. The "Gdef_N", "Gdef_Y", "Ghr_N", and "Ghr_Y" columns show the various metrics from comparing the hourly observation and simulation values during September 2017 for the Modiesha site and 7 September 2017 to 30 September 2017 for the Wanqingsha site.

Site name	*Metrics*	*Gdef_N (ppb)*	*Gdef_Y (ppb)*	*Ghr_N (ppb)*	*Ghr_Y (ppb)*
Modiesha	*Sim.*	*0.29*	*0.35*	*0.23*	*0.29*
	Obs.	*0.34*	*0.34*	*0.34*	*0.34*
	MB	*-0.06*	*0.01*	*-0.11*	*-0.05*
	NME	*76.0%*	*68.7%*	*73.6%*	*66.2%*
	NMB	*-16.4%*	*3.5%*	*-31.3%*	*-13.1%*
	R	*0.44*	*0.46*	*0.37*	*0.39*
Wanqingsha	*Sim.*	*0.29*	*0.31*	*0.27*	*0.29*
	Obs.	*0.45*	*0.45*	*0.45*	*0.45*
	MB	*-0.15*	*-0.14*	*-0.17*	*-0.15*
	NME	*58.9%*	*56.8%*	*60.4%*	*58.1%*
	NMB	*-34.7%*	*-30.6%*	*-38.7%*	*-34.8%*
	R	*0.35*	*0.39*	*0.34*	*0.4*

In the revised version, we have added the description of the measurement of ISOP.

For the isoprene (ISOP) evaluation, we use observation data from the Modiesha (23.11°N, 113.33°E) and Wanqingsha (22.71°N, 113.55°E) sites (Figure 1), where an online gas chromatography-mass spectrometry/flame ionization detector system (GC-FID/MSD, TH 300B, Wuhan) is used to measure VOCs in the ambient atmosphere. The system has a sampling rate of 60 mL/min for 5 minutes per sample, with a sampling frequency of once per hour (Meng et al., 2022). The ISOP observation data undergo rigorous quality control, which can be used for evaluating simulated ISOP concentrations. It is worth noting that the ISOP observational data for the Modiesha site covers September 2017, while the Wanqingsha site has data coverage from September 7 to September 30, 2017.

Comment 6: Title: "Unheralded" here in the title seems to suggest that no study before has attributed biogenic VOCs from urban greening to ozone production before, but there are many past studies that are cited in the introduction that have also concluded this. Could the authors choose a different word that better reflects the advancements and scope of their work?

Reply: Thanks for the good suggestion, and we have replaced the "Unheralded" with "Underappreciated" in the title.

Underappreciated contributions of biogenic volatile organic compounds from urban greening to ozone pollution: a high-resolution modeling study.

Comment 7: Line 21 – Can you expand more on what you mean by "advocated for mitigating urban atmospheric environment"? I'm not sure what this means.

Reply: Thanks for the good suggestion, and we have rewritten this sentence as follows.

Urban Green Spaces (UGS), such as parks, and gardens, are widely promoted as a strategy for improving the urban atmosphere and environmental health.

Comment 8: Table 3 – I believe that this is an hourly comparison between O_3 and NO_2. If so, can you add hourly to the title or table description for clarity to the reader.

Reply: Thanks for the good suggestion, and we have reorganized this table as follows.

Table 2: Evaluation results of the simulated monthly mean hourly O_3, MDA8 O_3, and hourly NO_2 mixing ratios for each case during September 2017.

Pollutant	*Case name*	*Sim (ppb)*	*Obs (ppb)*	*MB (ppb)*	*NMB*	*NME*	*R*
Hourly O_3	*Gdef_N*	*28.23*	*30.49*	*-2.26*	*-6.7%*	*23.6%*	*0.82*
	Gdef_Y	*28.67*	*30.49*	*-1.82*	*-5.3%*	*23.6%*	*0.82*
	Ghr_N	*28.89*	*30.49*	*-1.60*	*-4.8%*	*22.5%*	*0.83*
	Ghr_Y	*29.33*	*30.49*	*-1.15*	*-3.4%*	*22.4%*	*0.83*

MDA8 O_3	*Gdef_N*	*60.11*	*62.27*	*-2.16*	*-3.47%*	*21.71%*	*0.84*
	Gdef_Y	*61.04*	*62.27*	*-1.23*	*-1.97%*	*21.40%*	*0.84*
	Ghr_N	*61.07*	*62.27*	*-1.20*	*-1.92%*	*21.28%*	*0.84*
	Ghr_Y	*62.00*	*62.27*	*-0.26*	*-0.42%*	*21.23%*	*0.84*
Hourly NO_2	*Gdef_N*	*24.78*	*21.50*	*3.27*	*15.2%*	*45.7%*	*0.63*
	Gdef_Y	*24.74*	*21.50*	*3.24*	*15.0%*	*45.5%*	*0.63*
	Ghr_N	*24.35*	*21.50*	*2.84*	*13.2%*	*43.8%*	*0.63*
	Ghr_Y	*24.32*	*21.50*	*2.81*	*13.0%*	*43.6%*	*0.63*

Comment 9: Line 272 and Table 4 – You mention MDA8 O_3 here in the text? Is Table 4 MDA8 O_3 or hourly O_3? If possible, calculating these statistics on hourly O_3 and MDA8 O_3 is the most useful for both Table 3 and 4. Hourly O_3 especially R helps understand if you have represented the diurnal cycle well. And MDA8 ozone is useful to just investigate daytime ozone when O_3 is highest for regulatory applications. I would recommend calculating both MDA8 O_3 and hourly O_3 for both tables here.

Reply: Yes, Table 4 is for MDA8 O_3, and we have added the evaluation of hourly O_3 in Table 3 and Table 4. Meanwhile, we rewritten the related text in the manuscript. Please also see our responses to the above comment.

Additionally, various statistical metrics were used to assess the performance of hourly O_3, MDA8 O_3, and NO_2 concentrations from the CMAQ simulation (Emery et al. 2017). These metrics comprise the correlation coefficient (R), normalized mean bias (NMB), and normalized mean error (NME). The formulas for these metrics are listed in Table S3. As shown in Table 3, the modeling performance for all cases are reasonably, albeit with some degree of underestimation. Despite these discrepancies, the model demonstrates sufficient reliability and can be effectively used in the subsequent study. Meanwhile, the MBs of MDA8 O_3 across various cases indicate a substantial improvement in the CMAQ simulation when UGS-BVOC, UGS-LUCC, and their combined effects are considered. Specifically, the MB values decrease from -2.16 ppb in the Gdef_N case to -0.26 ppb in the Ghr_Y case, demonstrating that incorporating UGS-BVOC, UGS-LUCC, and their combined effects can enhance the accuracy of predicted daytime O_3 concentrations. In addition, we also evaluate the simulation performance for NO_2 in each case and the results suggest that all models have R above 0.63, and while there is some overestimation, the NMB is 15.0%, 15.2%, 13.0%, and 13.2% for Gdef_N, Gdef_Y, Ghr_N, and Ghr_Y, respectively. It should be emphasized that integrating UGS-BVOC into the modeling process can slightly improve the accuracy of NO_2 predictions, reducing the MB from 3.27 to 3.24 ppb, and from 2.84 to 2.81 ppb for Gdef and Ghr cases, respectively. The improvement in NO_2 predictions is attributed to the increased involvement of NO_2 in O_3 formation caused by the UGS-BVOC emissions, which reduces simulated NO_2 concentrations and narrows its bias against the observation.

In terms of O_3, the UGS-BVOC, UGS-LUCC, and their combined effects have various performances in different regions (Table 4). These results indicate that the inclusion of UGS-BVOC emissions remarkable influences MDA8 O_3 and hourly O_3 concentrations in the city center region and this effect, primarily observed when comparing the Gdef_Y with Gdef_N and Ghr_Y with Ghr_N cases, is largely due to the VOC-limited

areas prevalent in Guangzhou (He et al., 2024). By integrating the UGS-BVOC emissions into the models (comparing Gdef_Y and Gdef_N cases), the MBs of MDA8 O_3 and hourly O_3 in all regions, including a notable improvement in the city center region from -3.62 to -0.75 ppb and -2.86 to -1.18 ppb, respectively, is reduced. Additionally, the UGS-BVOC emissions slightly enhance R values of MDA8 O_3 and hourly O_3 in the city center and suburban regions, indicating a more accurate the daytime trend and the diurnal cycle representation, respectively. The UGS-LUCC effects, as seen when comparing Ghr_N and Gdef_N cases, also greatly improve model biases and the combined effects of both UGS-BVOC and UGS-LUCC (comparing the Ghr_Y and Gdef_N cases) substantially ameliorate model biases in the city center and suburban regions.

Table 4 Evaluation results of simulated monthly mean hourly O_3 and MDA8 O_3 mixing ratios in city center, suburban, and rural areas for each case during September 2017.

Variable	Regions	MB (ppb)				R			
		Gdef_N	Gdef_Y	Ghr_N	Ghr_Y	Gdef_N	Gdef_Y	Ghr_N	Ghr_Y
MDA8 O_3	City center	−3.627	−2.241	−2.110	−0.747	0.805	0.810	0.810	0.813
	Suburban	−4.076	−3.251	−3.210	−2.376	0.737	0.743	0.717	0.727
	Rural	−5.109	−4.757	−4.866	−4.528	0.665	0.655	0.695	0.690
Hourly O_3	City center	-2.862	-2.292	-2.086	-1.520	0.800	0.802	0.811	0.812
	Suburban	-3.148	-2.803	-2.647	-2.295	0.824	0.825	0.824	0.826
	Rural	-1.184	-1.630	-1.375	-1.164	0.742	0.741	0.751	0.750

Comment 10: Line 288 – By "monoethylene" here, do you mean "monoterpene". Is this a typo?

Reply: Thanks for your careful check, and we have fixed this typo.

A review of the data reveals that TERP and ISOP rank as the highest emitting species with proportions are 20.46% and 31.91% in this study, respectively, aligning with the findings of previous studies (Cao et al., 2022; Guenther et al., 2012b).

Comment 11: Line 297 – See major comment. I would suggest renaming urban in figure 1 and throughout the text including in this paragraph to something else so that readers can clearly differentiate between urban landcover type and what you are classifying as an urban region.

Reply: Thanks for the valuable suggestion, and we have renamed the "urban" as "city center" in Figure 1 and all related text in the manuscript.

Comment 12: Line 381 – Can you verify this statement "high-resolution land use cover data increase the estimation of the UGS-BVOC emissions in the urban and suburban regions." This is different from the text above and Figure 2A.

Reply: Thanks for the carefully check, we have rewritten this sentence.

The analysis also highlights high-resolution land use cover data increase the estimation of the UGS-BVOC emissions in the city center region.

Comment 13: Throughout 3.2, and all figures and tables therein, can you be clearer in the table and figure descriptions and throughout the text that these are all evaluated over September. I assume this is the case, but it would make it easier for the reader to state this clearly in the table and figure headings and descriptions.

Reply: Thanks for the carefully check, we have rewritten this sentence.

Comment 14: Lines 397 – 400 – These are very strong statements, but it's hard to confirm that these statements are really accurate without also plotting the absolute concentrations of MDA8 ozone and how much the UGS-BVOC and UGS-LUCC contribute to the total. Your conclusions here would be more impactful, if you plotted the total MDA8 ozone and then the contribution from UGS-BVOC and UGS-LUCC and where the regulatory metric is for MDA8 ozone as well. I would suggest showing a plot of your total MDA8 as well.

Reply: Thanks for the nice suggestion, we have softened the statements.

The observed increase suggests a potentially significant influence of UGS-BVOC emissions and UGS-LUCC on ozone levels, indicating that these factors may play an important role in ozone pollution research and should be carefully considered.

Comment 15: Figure 6 – If this is an average over September of the differences? Can you add this to the Figure description for clarity to the reader.

Reply: Thanks for the nice suggestion, we have changed the title for the figure.

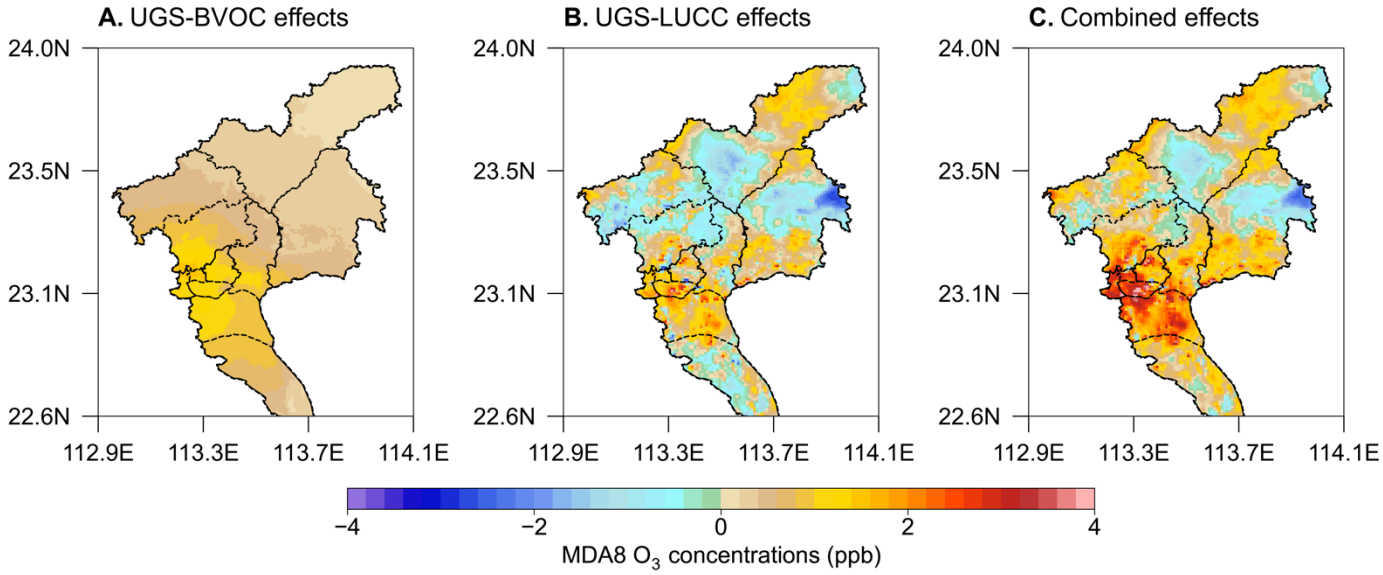

Figure 1 The map of UGS-BVOC effects (a), LUCC effects (b), and combined effects (c) in MDA8 O_3. Each map shows the difference in average MDA8 O_3 concentrations for each case (Gdef_Y, Ghr_N, and Ghr_Y) relative to the Gdef_N case during September 2017.

Comment 16: Figure 7A – Why plot your base case scenario (Gdef_N) here for MDA8 ozone. Why not your improved scenario (Ghr_Y) or at least both?

Reply: Thanks for the nice suggestion, we have replotted the figure using the simulation from the base case.

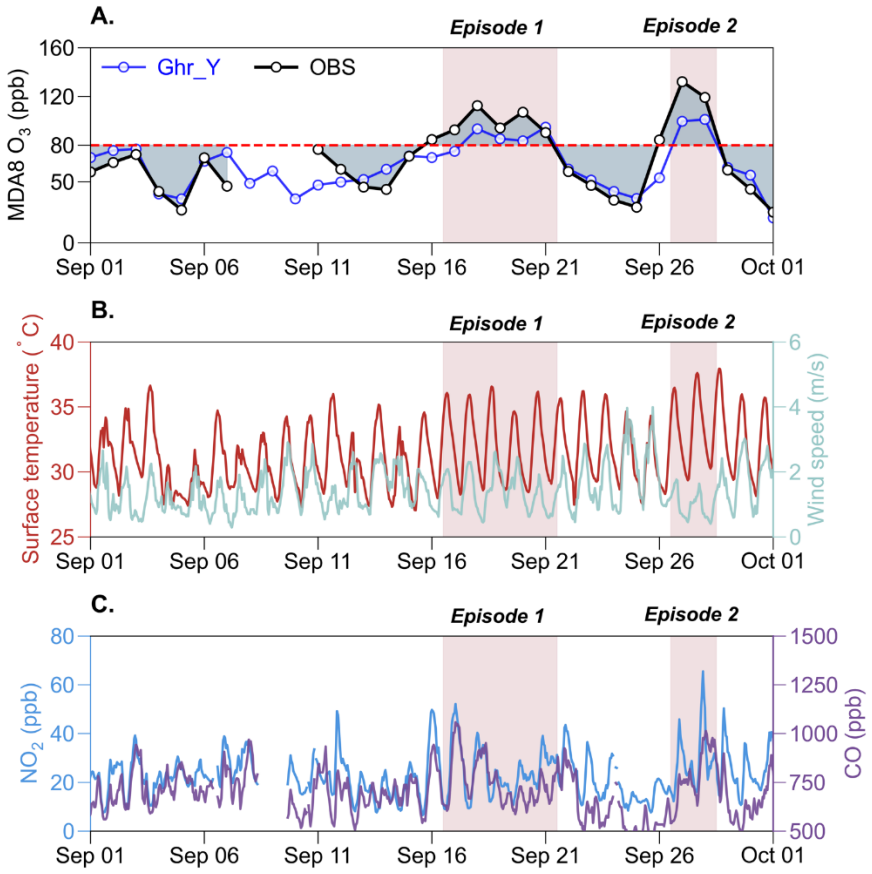

Figure 2 The comparison during September 2017 between the average values from simulation results grids which have air quality stations produced by the Gdef_N case and the average observation values for MDA8 O_3 (A). (B) is the meteorological fields from the average values from the simulation result grids, which have the same locations as the air quality stations. (C) is the observed average values for NO_2 and CO concentrations from all air quality stations.

Comment 17: Line 459 – What do you mean by "cumulative effects" here?

Reply: It means the combined effects of UGS-LUCC and UGS-BVOC. We have rewritten this sentence.

Furthermore, the UGS-LUCC effect's maximal contribution to the urban MDA8 O_3 levels could escalate to 2.2 ppb in Episode 1 and 23.7 ppb in Episode 2 while the combined effects of UGS-LUCC and UGS-BVOC emissions are projected to enhance MDA8 O_3 concentrations to 4.8 ppb and 25.2 ppb for the respective episodes.

Comment 18: Line 519 – I'm not sure these are what everyone would consider significant changes in ozone. Please add the percent changes in ozone in parenthesis here, so that readers can confirm your definition of significant is similar to their definition.

Reply: Thanks for this commend, and we have rewritten this sentence.

The UGS-BVOC emissions have a remarkable impact on ozone concentrations, with increases ranging from 1.0-1.4 ppb (+2.3-3.2%) in the city center regions.

Comment 19: Data availability – ACP journals require at least model code be made public (https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html). Please upload your WRF-Chem model code to be available online.

Reply: Thanks for the reminder, and we have rewritten this part.

The WRF (Weather Research and Forecasting Model) code can be obtained from the official repository at https://github.com/wrf-model/WRF. The CMAQ (Community Multiscale Air Quality Model) code is accessible at https://github.com/USEPA/CMAQ. Model output data used for analysis and plotting, and the code used for simulations can be made available upon request (Haofan Wang, wanghf58@mail2.sysu.edu.cn).

Comment 20: Line 210 need a space between Table 1 and were.

Reply: Thanks for the carefully check, and we have added a space between "Table 1" and "were".

In this study, four distinct cases, as listed in Table 1 were established to investigate the impacts of UGS-LUCC, UGS-BVOC, and their combined effects on the ozone simulation.

Comment 21: Line 507 – not a complete sentence.

Reply: Thanks for the carefully check, and we have rewritten this sentence.

Considering the UGS-BVOC and UGS-LUCC effects can effectively mitigate the underestimation of surface ozone concentrations by regional air quality models, though other factors such as inaccuracies in emissions inventories, chemical mechanisms, and meteorological inputs may also contribute to these underestimations.

Comment 22: Line 520 – typo with a comma instead of a period.

Reply: Thanks for the carefully check, and we have rewritten this sentence.

The UGS-BVOC emissions have a remarkable impact on ozone concentrations, with increases ranging from 1.0-1.4 ppb (+2.3-3.2%) in the city center regions.