

Response to Reviewer #3:

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 uses high-resolution WRF-CMAQ simulations to investigate the impacts of urban greening in Guangzhou, China through the use of different resolution input datasets on land cover and leaf area indices. The impact of these different model configurations is usefully explored through comparisons with observations. Representing urban green spaces in models is important for understanding ozone atmospheric chemistry in urban settings. This study is relevant to ACP. I recommend publication of this manuscript after the concerns specified below have been addressed.

Reply: We sincerely thank the reviewer for the thoughtful and encouraging feedback on our study. We are pleased that you recognize the relevance of our work in representing urban green spaces and investigating ozone atmospheric chemistry in urban settings. Your acknowledgment of the importance of high-resolution simulations and comparisons with observations is greatly appreciated. We will carefully address the concerns raised to improve the manuscript and ensure it meets the standards of ACP. Thank you for your constructive feedback and support.

Comment 2: The manuscript discusses the use of different resolution input datasets and uses a high resolution, nested model. It would be useful if the authors could include a section on the benefits and limitations of using high resolution. For example, you have 1 km versus 10 m land cover data, but when running the model, you have all of this information in a 1 km grid box. How do these high-resolution datasets impact the calculations of BVOCs in the model? What are the uncertainties here?

Reply: Thanks for the valuable suggestion. We have added a “Uncertainties and Limitations” section to explain it.

First, the 10-m resolution land use and land cover data still cannot fully capture the spatial pattern of UGS in Guangzhou. As shown in Figure S2, although UGS in Guangzhou is primarily composed of EBTs, most of these EBTs are distributed along urban edges. This may result from distortions in the definition of urban extent,

such as misclassifying mixed urban-vegetation grids as urban grids, caused by the coarse resolution of the 1-km land use and land cover data. The fuzzy definition of urban boundaries could lead to non-UGS areas being misclassified as UGS, potentially resulting in an overestimation of UGS-BVOC emissions.

Comment 3: Although the authors compare the simulations to observations of meteorological parameters, O₃, NO₂, and isoprene, there is very little discussion on how these observations were measured and the time resolution. It would be helpful to readers to elaborate more on this.

Reply: Thanks for the valuable suggestion. We have added some information about the measurement in Section 2.4.

The real-time hourly concentration of O₃ was measured by the ultraviolet absorption spectrometry method and differential optical absorption spectroscopy at each monitoring site. NO₂ concentrations are measured by the molybdenum converter method known to have positive interferences from NO₂ oxidation products (Dunlea et al., 2007). The instrumental operation, maintenance, data assurance, and quality control were properly conducted based on the most recent revisions of China Environmental Protection Standards (Zhang and Cao, 2015), and the locations of these air quality stations are depicted in Figure 1. Additionally, meteorological data also undergo thorough quality control. Subsequently, they are utilized to assess the model performance of WRF-CMAQ.

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 4: In this manuscript, the authors are addressing the sensitivity of O₃ to changes in BVOCs, but there is little discussion on whether O₃ is NO_x-limited or VOC-limited in Guangzhou. There is also no discussion on other potential sources affecting the area. The authors mention that there is rapid urbanization happening in Guangzhou. Does this mean that there is more industry, more vehicles, etc. that could be impacting O₃ production and loss processes? O₃ is nonlinear, and depending on the regime, and other factors like emissions, the composition and ratios of VOCs and NO_x in the area will affect O₃ differently.

Reply: Thanks for the very valuable suggestions. We have discussed the VOC-limited or NO_x-limited conditions of Guangzhou in Section 3.3 and 4.

Section 3.3:

N. Wang et al. (2019) reported that VOC levels can be highly sensitive in VOC-limited regions, where sufficient NO_x concentrations mean that even a small disturbance in VOCs can cause significant changes in O₃ concentrations. Similarly, metropolitan areas, such as Guangzhou, often experience VOC-limited conditions or NO_x-saturation (P. Wang et al., 2019). Consequently, the UGS-BVOC case results in an overall increase in MDA8 O₃.

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 5: There is an inconsistency with the units, which can be confusing to readers. At times, the authors use “ppb” and at other times they use “μg/m³” for gases. Please use a consistent unit throughout the paper.

Reply: Thanks for the valuable suggestion. We have changed all the unit “μg/m³” to “ppb” for gases.

Comment 6: For the tables and figures in the paper, please specify the timeframes that the concentrations are averaged over.

Reply: Thanks for the nice suggestions. We have added the timeframes in the titles of tables and figures.

Comment 7: Figure 1: This is a paper based on Guangzhou. For readers who might not be familiar with the area, could the authors provide more description on the urban, suburban, and rural regions? Is there much vegetation in the urban centers?

Reply: Thanks for the valuable suggestion. We have added some information about the city center, suburban, and rural regions.

The city center region has more UGS areas due to the higher urban land use and land cover fraction (Figure S1) compared to the suburban and rural regions.

Comment 8: Section 2.2: Did the authors use online or offline MEGAN? It seems to me that you are referring to MEGANv2.1, not v3.1. Please clarify, as the resolution of the model can have impacts on online emission calculations.

Reply: We used the online MEGAN in the CMAQ simulation. We have updated the Section 2.2 and the updated version is for the MEGANv3.1.

In this equation, E is the net emission flux ($\mu\text{g m}^{-2} \text{h}^{-1}$), and EF is the weighted average of the emission factor ($\mu\text{g m}^{-2} \text{h}^{-1}$) for each vegetation type calculated by Emission Factor Processor (EFP). The emission activity factor (γ) considers emission responses to changes in environmental and phenological conditions. Compare with earlier versions, γ in MEGANv3.1 adds quantifications for responses to high and low temperature, high wind speed, and air pollution (O_3).

$$\gamma = LAI \times \gamma_{TP} \times \gamma_{LA} \times \gamma_{SM} \times \gamma_{HT} \times \gamma_{LT} \times \gamma_{HW} \times \gamma_{CO_2} \times \gamma_{BD} \times \gamma_{O_3} \quad (\text{Eq. 2})$$

In this equation, the activity factor denotes the emission response to canopy temperature/light (γ_{TP}), leaf age (γ_{LA}), soil moisture (γ_{SM}), high temperature (γ_{HT}), low temperature (γ_{LT}), high wind speed (γ_{HW}), ambient CO_2 concentration (γ_{CO_2}), bidirectional exchange (γ_{BD}), O_3 exposure (γ_{O_3}), and Leaf Area Index (LAI). In this study, γ_{CO_2} was not considered in the BVOC emission estimation. The MEGANv3.1 approach can calculate the emissions at each canopy level as the product of the emission factor and emission activity at each level.

Comment 9: Section 2.3 and onwards: I would recommend that the authors refrain from using the term “scenario” to describe the different simulations. These are model configurations, whereas the term “scenario” tends to highlight a potential future or possible outcomes.

Reply: Thanks for the very nice suggestions. We have replaced the “scenario” to “case” in the whole manuscript.

Comment 10: Section 2.4: What kind of instrumentation was used to measure O_3 ?

Reply: Thanks for the very carefully checking. We have added the description of the instrumentation for measuring O_3 and NO_2 .

The real-time hourly concentration of O_3 was measured by the ultraviolet absorption spectrometry method and differential optical absorption spectroscopy at each monitoring site. NO_2 concentrations are measured by

the molybdenum converter method known to have positive interferences from NO₂ oxidation products (Dunlea et al., 2007). The instrumental operation, maintenance, data assurance, and quality control were properly conducted based on the most recent revisions of China Environmental Protection Standards (Zhang and Cao, 2015), and the locations of these air quality stations are depicted in Figure 1. Additionally, meteorological data also undergo thorough quality control. Subsequently, they are utilized to assess the model performance of WRF-CMAQ.

Comment 11: Table 2: Are these monthly concentrations? Please clarify in the text or in the caption.

Reply: Thanks for this suggestion. We have reorganized this table.

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.

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

Comment 12: Line 260: By “specified requirement”, do the authors mean the observed values? If so, I would refer to them as that because the authors are not referring to a standard of sorts.

Reply: Thanks for the good suggestion. We have rewritten this sentence.

Additionally, various statistical metrics were used to assess the performance of hourly O₃, MDA8 O₃, and NO₂ 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.

Comment 13: Line 265-267: The authors mention that integrating UGS-BVOC can improve the accuracy of NO₂ predictions. Can the authors elaborate on why this improvement in simulated NO₂ happens?

Reply: Thanks for the valuable suggestion. We have added the elaborate on why this improvement in simulated NO₂ happens.

The improvement in NO₂ predictions is attributed to the increased involvement of NO₂ in O₃ formation caused by the UGS-BVOC emissions, which reduces simulated NO₂ concentrations and narrows its bias against the observation.

Comment 14: Table 3: Again, what time frame are these concentrations averaged over? Monthly?

Reply: Thanks for the valuable suggestion. We have added the timeframe to the title of the table.

Table 2 Evaluation results of the simulated monthly mean hourly O₃, MDA8 O₃, and hourly NO₂ 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 15: Table 4: Same comment as tables 2 and 3.

Reply: Thanks for the valuable suggestion. We have added the timeframe to the title of the table.

Table 3 Evaluation results of simulated monthly mean hourly O₃ and MDA8 O₃ 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 16: Line 288: Do you mean “monoterpene” instead of “monoethylene”?

Reply: Thanks for the carefully checking. We have changed the “monoethylene” to “monoterpene”.

Comment 17: Line 473-475: This sentence is rather confusing. Can you clarify how an increase and simultaneous decrease leads to an overall increase in wind speed?

Reply: Thanks for this suggestion. We have rewritten this sentence.

Figure 10 illustrates that during Episode 1, the UGS-LUCC effects led to a notable increase in the frequency of higher wind speeds (1.2–1.4 m/s) and a simultaneous decrease in the frequency of lower wind speeds (0.9–1.1 m/s). This shift in the wind speed distribution suggests an overall increase in average wind speed due to the UGS-LUCC effects during Episode 1.

Comment 18: Technical Correction 1: At the beginning of the paper, the authors establish an abbreviation convention, such as TERP for monoterpenes, ISOP for isoprene, LUCC for land use cover change, etc. Throughout the paper, I found that the abbreviations were being described again on several occasions (i.e., line 288, line 277-278). Please make this consistent throughout the paper.

Reply: Thanks for this carefully checking. We have corrected these errors.

Comment 19: Technical Correction 2: When naming figures throughout the paper, the authors should use the naming convention of “Figure 1A and 1B” rather than “Figure 1 (A) and (B)”.

Reply: Thanks for this nice suggestion. We have changed the naming convention of the figures in the manuscript.

Comment 20: Line 74: “the complexity of these interactions, and they demonstrated that vegetation could exert nonlinear effects on” – Remove “they” from sentence.

Reply: Thanks for this suggestion. We have removed “they” here.

Furthermore, Seinfeld et al., (1998) underscores the complexity of these interactions, and demonstrated that vegetation could exert nonlinear effects on meteorological processes.

Comment 21: Line 97: Should be “investigated” instead of “investigating”.

Reply: Thanks for this suggestion. We have changed the “investigating” to “investigated”.

Comment 22: Line 210: Space between “Table 1” and “were”.

Reply: Thanks for this suggestion. We have added a space between “Table 1” and “were”.