Response to Reviewer #2:

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 manuscript describes a numerical modeling study (WRF-CMAQ-MEGAN) to investigate the impact of urban greening on air pollution. This is an important topic that has been the subject of quite a few papers recently but there is certainly more that needs to be done to adequately address this topic. The authors use a relatively high-resolution (1 km) modeling system which is appropriate for advancing our understanding on urban BVOC impacts. They compare their results with observations which is helpful for seeing if there are improvements. They then present three scenarios (landcover data resolution, BVOC emission estimates, urban green space amount) to a base case to show the impacts of each one. This is a useful study but I have some concerns about the manuscript that should be addressed before accepting this paper for publication in ACP.

Reply: We sincerely thank the reviewer for their thoughtful and encouraging feedback on our study. We are pleased that you recognize the importance of investigating urban greening impacts on air pollution and appreciate your acknowledgment of our approach, including the use of high-resolution modeling and the comparison with observations. Your comments motivate us to continue refining our work and addressing the specific concerns raised to ensure the manuscript meets the high standards of ACP.

Comment 2 The introduction (Section 1) has statements that do not seem to be supported by the references given and these references do not appear to be very relevant for urban green spaces anyway. Examples include lines 63-64, 67-71, 71-73, 80-81, etc.

Reply: Thanks for this suggestion. We have added some relevant references.

Deposition involves the absorption of air pollutants onto vegetative surfaces, while dispersion refers to the reduction of air pollutant concentrations through aerodynamic effects caused by vegetation (Tiwari and Kumar, 2020; N. Wang et al., 2019a).

These conditions are influenced by several factors, such as the specific structure of the UGS vegetation properties (e.g., height, leaf density), the site context (e.g., street canyon geometry, proximity to emission sources), and prevailing meteorological conditions (e.g., wind speed and direction) (Jin et al., 2017; Tomson et al., 2021; Yang et al., 2020).

For example, dense tree canopies might impede ventilation in urban street canyons, while porous vegetation barriers in open-road settings could potentially intensify roadside air pollution concentrations (Chen et al., 2021; Jin et al., 2014).

UGS also have a complex role in air quality due to their production of biogenic volatile organic compounds (BVOCs). For instance, in cities like Los Angeles, the UGS-BVOC emissions contribute to a quarter of the secondary organic aerosol formation on hot days (Schlaerth et al., 2023).

Chen, X., Wang, X., Wu, X., Guo, J., Zhou, Z., 2021. Influence of roadside vegetation barriers on air quality inside urban street canyons. Urban For. Urban Green. 63, 127219. https://doi.org/10.1016/J.UFUG.2021.127219

Fu, X., Liu, J., Ban-Weiss, G., Zhang, J., Huang, X., Ouyang, B., Popoola, O., Tao, S., 2017. Effects of canyon geometry on the distribution of traffic-related air pollution in a large urban area: Implications of a multi-canyon air pollution dispersion model. Atmos. Environ. 165, 111–121. https://doi.org/10.1016/J.ATMOSENV.2017.06.031

Jin, S., Guo, J., Wheeler, S., Kan, L., Che, S., 2014. Evaluation of impacts of trees on PM2.5 dispersion in urban streets. Atmos. Environ. 99, 277–287. https://doi.org/10.1016/J.ATMOSENV.2014.10.002

Schlaerth, H.L., Silva, S.J., Li, Y., 2023. Characterizing Ozone Sensitivity to Urban Greening in Los Angeles Under Current Day and Future Anthropogenic Emissions Scenarios. J. Geophys. Res. Atmospheres 128, e2023JD039199. https://doi.org/10.1029/2023JD039199

Tiwari, A., Kumar, P., 2020. Integrated dispersion-deposition modelling for air pollutant reduction via green infrastructure at an urban scale. Sci. Total Environ. 723, 138078. https://doi.org/10.1016/j.scitotenv.2020.138078

Tomson, M., Kumar, P., Barwise, Y., Perez, P., Forehead, H., French, K., Morawska, L., Watts, J., 2021. Green infrastructure for air quality improvement in street canyons. Environ. Int. 146, 106288. https://doi.org/10.1016/j.envint.2020.106288

Xing, Y., Brimblecombe, P., 2019. Role of vegetation in deposition and dispersion of air pollution in urban parks. Atmos. Environ. https://doi.org/10.1016/J.ATMOSENV.2018.12.027

Yang, H., Chen, T., Lin, Y., Buccolieri, R., Mattsson, M., Zhang, M., Hang, J., Wang, Q., 2020. Integrated impacts of tree planting and street aspect ratios on CO dispersion and personal exposure in full-scale street canyons. Build. Environ. 169, 106529. https://doi.org/10.1016/j.buildenv.2019.106529

Comment 3: The comparison between the "default" low resolution run and the higher resolution data there are other differences in these runs and they do not really show what is the impact of 1) the resolution of the data used to derive landcover and 2) the resolution of the model simulations. It would be more useful to show the individual impacts of these two differences. Also, while it is reasonable to assume that 10-m landcover data is more appropriate than 1-km landcover data, there are still uncertainties associated with the 10-m data. There should be a detailed discussion of the uncertainties for each landcover input (LAI, growth form fractions, ecotypes and ecotype-specific emission factors) and how this compares with 1-km data.

Reply: Thanks for this comment, and we have added a section "Uncertainties and limitations" to address this comment.

In this study, we used land use and land cover data integrated at 1-km and 10-m resolutions to define the urban boundary and characterize the spatial distribution of UGS in Guangzhou. Additionally, we incorporate high-resolution LAI data, obtained through machine learning, as input for the MEGAN model. Using the WRF-CMAQ model, we quantify the effects of UGS-BVOC, UGS-LUCC, and their combined impacts on ozone concentrations in Guangzhou. However, some uncertainties and limitations remain.

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.

Second, due to resolution limitations, only larger patches of grassland, cropland, and woodland are recognized as UGS, while smaller UGS vegetation, such as street trees, often goes undetected at a 10-m resolution. This omission can lead to an underestimation of the UGS-BVOC emissions.

Third, the 10-m and 1-km resolution land use and land cover data, along with the growth forms and ecotype data, use simplified categorizations for grids, which cannot fully capture the diversity of vegetation species within UGS. Since different vegetation species have varying emission factors, this simplification introduces some errors. Similarly, the oversimplified classification of land grids limits this study's ability to provide specific planning strategies for UGS at the species level. Nevertheless, it can highlight the importance of considering UGS-BVOC and UGS-LUCC in air pollution prevention and control policies.

Comment 4: The comparison with ambient isoprene observations is a valuable addition to this study but it should be extended by discussing the issues with this limited comparison including how these concentrations are influenced by model emissions, dilution, chemical losses, etc which have major uncertainties. How do uncertainties compare with the differences in the results for the different scenarios? Also, include a description of the BVOC data used to evaluate the model and an assessment of the quality of the data.

Reply: Thanks for this comment, and we have added the description of the isoprene measurement. The validation results are constrained by the limited ISOP observations from only 2 sites. Additionally, the estimation of BVOC emissions by the MEGAN model is inherently uncertain, which may further contribute to validation bias.

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 5: A major missing component is a thorough discussion on what these findings can tell us about urban BVOC and air pollution- more than ozone will increase if there is sufficient NOx which is already well known. This needs to include a discussion of NOx as well as other complexities. Make it clear whether these findings are specific for Guangzhou or are also relevant for other cities. Discuss limitations such as not having representative BVOC emission factor data for these landscapes (that requires tree species composition data and accurate tree species-specific emission factors). Also, what are the uncertainties associated with growth form estimates. If the UGS is mostly grass then that will have a much lower emission than trees.

Reply: Thanks for this comment, and we have added related discussion to 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.

Gong, J., Hu, Z., Chen, W., Liu, Y., Wang, J., 2018. Urban expansion dynamics and modes in metropolitan Guangzhou, China. Land Use Policy 72, 100–109. https://doi.org/10.1016/J.LANDUSEPOL.2017.12.025

Kai, Z., Wen-jie, Z., Zhi-fang, W., Wei, C., Shao-lin, P., 2011. The influence of urbanization on atmospheric environmental quality in Guangzhou, China. 2011 Int. Conf. Electr. Technol. Civ. Eng. ICETCE 3667–3670. https://doi.org/10.1109/ICETCE.2011.5776281

Liu, Z., Doherty, R., Wild, O., Hollaway, M., O'Connor, F., 2021. Contrasting chemical environments in summertime for atmospheric ozone across major Chinese industrial regions: the effectiveness of emission control strategies. Atmospheric Chem. Phys. https://doi.org/10.5194/ACP-21-10689-2021

Comment 6: The authors discuss the impacts of LUCC on temperature and solar radiation- the temperature "heat island" is well known but the impact on solar radiation is less clear. What are the processes in the model and are they realistic? By "urban region receives less solar radiation than other regions likely due to the shading effect of urban canopies" do you mean that you are using the ground surface temperature and light to drive BVOC emissions? The ground surface values are not the light and temperature that you should be using to drive the BVOC emissions. It should be the canopy light and temperature. Reassess whether the impacts you are seeing are influenced by the model is using the wrong light and temperature.

Reply: Thanks for this nice question. We don't mean that we used the ground surface temperature and light to drive BVOC emissions. MEGANv3.1 use the 2-m temperature variable from the WRF model to calculate the BVOC emissions, and the 2-m temperature in WRF can be affected by the shading effect.

Comment 7: Ozone responds to temperature for multiple reasons. Quantify the impact of BVOC emission response to temperature relative to these other reasons.

Reply: This is a valuable comment. However, we think this question beyond the scope of this study. Another study by our group (Li et al., 2024) demonstrated the responses of O_3 to temperature through multiple mechanisms, including changes in chemical reaction rates, BVOCs emissions, soil NO_x emissions, dry deposition, PAN decomposition, and etc. The major one among all mechanisms varies in regions. Please refer to more information in this paper.

Li, S., Lu, X., and Wang, H.: Anthropogenic emission controls reduce summertime ozone-temperature sensitivity in the United States, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2024-1889, 2024.

Comment 8: Summarize the data shown in section 3.3 in a table that provides an overview of these results.

Reply: Thanks for this suggestion, and we have added a table to summarize these results.

Table 6 presents the overall results for the impacts of UGS-LUCC and UGS-BVOC on MDA8 O_3 concentrations. The effects show slight variations across different regions during September, while the effects during the two episodes exhibit more significant changes. In the city center region, which shows the largest changes, the UGS-BVOC effect shows increases by +1.6 ppb in Episode 1 and +5.9 ppb in Episode 2, indicating that the UGS-BVOC effects influence MDA8 O_3 concentrations in the city center during ozone *episodes, while their impact is minimal in suburban and rural regions. These results highlight the important effects of UGS-LUCC and UGS-BVOC in urban areas, especially during O_3 pollution periods.*

Table 1 Summary of Average MDA8 O_3 Concentrations (ppb) for Various Effects during September 2017.

Regions	Periods	UGS-BVOC effect	UGS-LUCC effect	Combined effect
City center	Monthly	+0.4	0.0	0.4
	Episode 1	+1.6	+1.6	+3.2
	Episode 2	+2.9	+5.9	+8.9
Suburban	Monthly	+0.4	0.0	0.4
	Episode 1	+1.5	+0.9	+2.0
	Episode 2	+1.6	+1.7	+3.4
Rural	Monthly	+0.4	0.0	0.4
	Episode 1	+0.5	+0.6	+1.1
	Episode 2	+0.5	0.0	+0.5

Comment 9: The manuscript would benefit from editing for English language usage.

Reply: Thank you for your suggestion. We have carefully reviewed and improved the English language throughout the manuscript to enhance its clarity and readability.

Comment 10: Line 1: "Unheralded" is not justified for the title and should be deleted.

Reply: We have reworded this title.

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

Comment 11: 27: In addition to the 666 Gg BVOC value for Guangzhou, include the emission per area and also how BVOC compare to AVOC.

Reply: Thanks for this suggestion. We have added some information about this in the abstract and main text.

***Abstract**:*

Our findings indicate that the UGS-BVOC emissions in Guangzhou amounted to 666 Gg (~90 Mg/km^2), with isoprene (ISOP) and monoterpene (TERP) contributing remarkably to the total UGS-BVOC emissions. In comparison to anthropogenic VOC (AVOC) and BVOC emissions, UGS-BVOC emissions account for approximately 33.45% in the city center region.

Section 3.2:

Furthermore, Table 5 reveals that in September, the UGS-BVOC emissions in Guangzhou amounted to 666 Gg (~90 Mg/km^2), with ISOP and TERP contributing remarkably to the total UGS-BVOC emissions. In comparison to anthropogenic VOC (AVOC) and BVOC emissions, UGS-BVOC emissions account for approximately 33.45% in the city center region.

Comment 12: 28: What does it mean 30% of urban ISOP? Is the other 70% anthropogenic isoprene or is it other biogenic isoprene. Along these lines, is UGS BVOC all BVOC? Or are there BVOC from other vegetation such as street trees which are generally not thought of as green space vegetation.

Reply: Thanks for this question and this description is misleading. We have removed this sentence.

Comment 13: 36: how does this study highlight the need for selecting low-emission vegetation and refining vegetation classification? I see little here that provides specific guidance to air quality managers.

Reply: Thanks for this question. We have removed this description.

Comment 14: 63-64: Clarify the point being made regarding the importance of dispersion over deposition. What do the references say that actually support this?

Reply: We have added this reference, and we also briefly describe the method adopted in this paper.

Notably, Ramanathan et al. (2001) reported that dispersion effects are significantly more impactful than deposition, exceeding it by an order of magnitude via a radiative forcing modeling method.

Comment 15: Line 126 to 129: This is not the default LAI for MEGAN3.1. The LAI data referred to here is probably the MEGAN2.1 LAI data. It can be used with MEGAN3.1 but there is no default MEGAN3.1 LAI data. It is expected that users generate their own LAI data for any MEGAN 3.1 simulation.

Reply: Thanks for the valuable suggestion, and we have rewritten this sentence.

The default LAI dataset to drive the MEGANv2.1 model which can used for MEGANv3.1 is derived from the enhanced Moderate Resolution Imaging Spectroradiometer (MODIS) /MOD15A2H in 2003 with 1 km spatial resolution (Myneni et al., 2015).

Comment 16: Line 155-164: These equations and text describe MEGAN2.1, not MEGAN3.1.

Reply: Thanks for the valuable suggestion, and we have changed this part.

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 17: Line 310-311: How do you define "natural area" vs "UGS".

Reply: Thanks for this suggestion, and we have changed the "natural area" to "non-UGS area".

Figure 2B offers a clear depiction of the proportion of UGS-BVOC emissions relative to non-UGS area BVOC emissions in each region of Guangzhou City, which presents that the UGS-BVOC emissions in the city center region constitute 57.34% of the total BVOC emissions in this region because of the larger urban proportions in the city center region (Figure 5), while the UGS-BVOC emission proportion in suburban and rural are 19.44% and 1.86% respectively.

Comment 18: Line 398- this is not clear. Please reword this sentence

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

This finding underscores the essential role that integrated urban planning and environmental management play in controlling ozone pollution within metropolitan regions. By considering UGS-BVOC emissions in air quality models and management plans, managers can make more informed decisions to mitigate ozone levels and improve regional air quality.

Comment 19: Line 399-400: If there is a "revelation" here then you should make it clear how managers can use this and what they should be doing.

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

This finding underscores the essential role that integrated urban planning and environmental management play in controlling ozone pollution within metropolitan regions. By considering UGS-BVOC emissions in air quality models and management plans, managers can make more informed decisions to mitigate ozone levels and improve regional air quality.

Comment 20: 486: replace "was" with "is" unless you are referring to a specific episode (in which case make that clear)

Reply: Thanks for this nice suggestion.

The rapid urbanization process is accompanied by a higher frequency of ozone episodes.

Comment 21: 488-490: This meaning of this sentence is not clear.

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

Guangzhou, located in southern China and known as a pioneer city in reform and opening-up policies, has experienced rapid urbanization over the past thirty years, leading to increased challenges with ozone pollution.

Comment 22: 27 and 497: should note that isoprene and terpenes are only half of the total. "primary" could lead readers to think they are almost all of it.

Reply: Thanks for this nice suggestion.

In September 2017, the UGS-BVOC emissions in Guangzhou totaled 666 Gg, with ISOP and TERP as the major species, emitting 213 and 136 Gg, respectively.

Comment 23: 498: are there UGS outside of urban areas? I would expect that they should all be urban.

Reply: Yes, there are UGS outside of the city center area (i.e., suburban, and rural region). See Fig. S1.

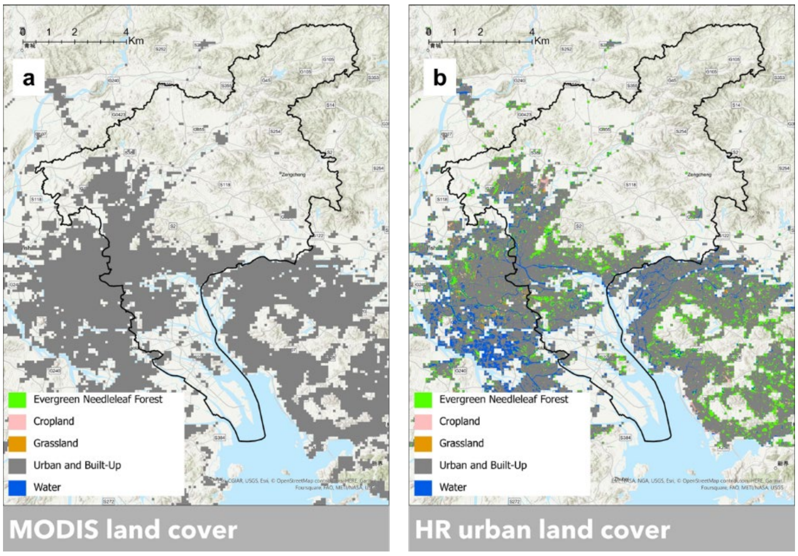

Figure S1 The processed land cover dataset. (a) is the MODIS land cover without UGS and (b) has characterized the UGS base on MODIS land cover.

Comment 24: 505-506: Are you saying that managers should not plant EBTs? But some EBTs have low emissions- lower than other vegetation. They should not be all grouped together.

Comment 25: 507-508- should also point out that there are other possibilities for ozone underestimation.

Reply: Thanks for this nice suggestion. 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.