Response letter for EGUSPHERE-2024-3180

Dear Editor and Reviewer:

We sincerely appreciate your help with our manuscript entitled "What makes the less urbanized city a deeper ozone trap: implications from a case study in the Sichuan Basin, southwest China" (EGUSPHERE-2024-3180) submitted to *Atmospheric Chemistry and Physics*. Your review efforts and valuable comments have been very helpful in improving the quality of the manuscript. We have carefully studied all the comments and made revisions according to the suggestions. We hope that our revisions could address your concerns on the manuscript adequately. The revised portions are marked in **red** or **blue** in the revised manuscript, and the responses to the reviewers in this document are in **blue**. In this response document, the line numbers marked to the comments (in black) indicate the lines in the previous clean version of revised manuscript, while the line numbers in **blue** text indicate the lines in the current revised manuscript (marked changes). The corrections and responses are summarized as follows:

Response to Reviewer #2

In this study, the authors analyzed the spatial pattern of ozone levels across the Sichuan Basin of China and further investigated the urban-rural intra-pattern of ozone in several cities using Chinese air quality reanalysis (CAQRA) and satellite data products. However, I do not believe this paper offers a substantial contribution to the existing body of knowledge, and its overall structure lacks clarity and organization. Although the manuscript is titled "What makes the less urbanized city a deeper ozone trap: implications of a case study in the Sichuan Basin, southwest China", the authors fail to convincingly address this central question with robust evidence or clear analysis. The spatial distribution of ozone and its precursors in the Sichuan Basin has already been well-documented in prior studies. What novel insights or significant differences does this work present in comparison? Moreover, the analysis relies solely on basic statistical methods to assess ozone and meteorological factors, which limits the scientific rigor of the study. As such, I don't

think that this paper merits publication in a high-standard journal as ACP.

Author's response: Thank you for your review efforts. We have to admit that our study has not fully quantify all the factors controlling the long-term spatial behavior of near surface ozone. However, our research purpose was not to fully resolve this issue. The novelty of our study lies in identifying the characterized long-term spatial behavior of the dipole-like asymmetric ozone trap over the two mega cities of the Sichuan Basin, with the causal analysis merely providing certain implications. As summarized in our introduction (Line 58~67 in the original manuscript), previous studies (Zhao et al., 2019; Liu et al., 2020; Ning et al., 2020; Deng et al., 2022; Wu et al., 2022) have neither reported this dipole-like asymmetric ozone titration phenomenon, nor conducted high spatial-resolution urban-rural ozone gradient analyzes, especially targeting this feature. As Reviewer #1 rightly noted: "The clarity of the analysis and the unique geographical context of the study make the manuscript a useful contribution to the field."

In this revision, we have adopted the newly released native 1-km spatial resolution data (February 2025), and the re-analyzed results based on the updated dataset remained consistent with our previous findings. For these reasons, we sincerely request you please to reconsider re-evaluating our manuscript.

Major comments:

1) The most critical flaw of this work is its neglect of the impact of complex urban anthropogenic emission changes on the unique deep-basin topography of the Sichuan Basin. Although the authors aim to address the question "What makes the less urbanized city a deeper ozone trap: implications from a case study in the Sichuan Basin, southwest China", they overlook the fact that ozone concentrations in highly urbanized areas are primarily influenced by precursor emissions. This is especially important considering the significant reduction in NO_x ***emissions in the Sichuan Basin since the implementation of China's Air Pollution Prevention and Control Action Plan (APPCAP) in 2013. It is widely acknowledged that meteorological conditions typically affect ozone concentrations only under extreme weather or climate events (such as the ozone surge in the Sichuan Basin during the summer of***

2022), while long-term ozone levels in urban areas tend to be more sensitive to changes in precursor emissions. It is undoubtedly insufficient to explain the phenomenon of a deeper ozone trap solely based on a few simple meteorological variables. However, the authors simply skip over the quantitative assessment of the impact of anthropogenic emission processes.

Author's response: Thank you for your review efforts. We fully acknowledge that ozone concentrations in urban areas are fundamentally determined by the chemical regime. As shown in our manuscript (both the revised and original version), we did isochronous spatial analyses on the urban-rural gradients of NO_2 level, and further added analyzes on formaldehyde and its ratio to NO_2 in the revision. The updated figures and table with new information brought about quantitative assessment of the chemical regime over the study region:

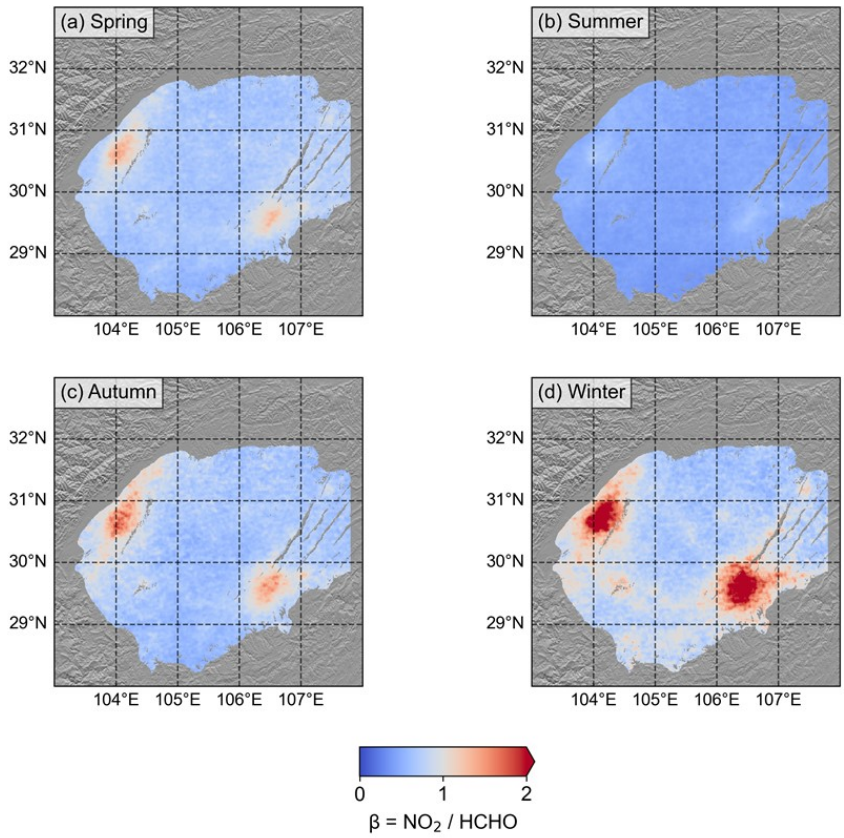

Figure R1 Spatial patterns for the number concentration ratio of NO_2 to formaldehyde over seasons during 2019-2023.

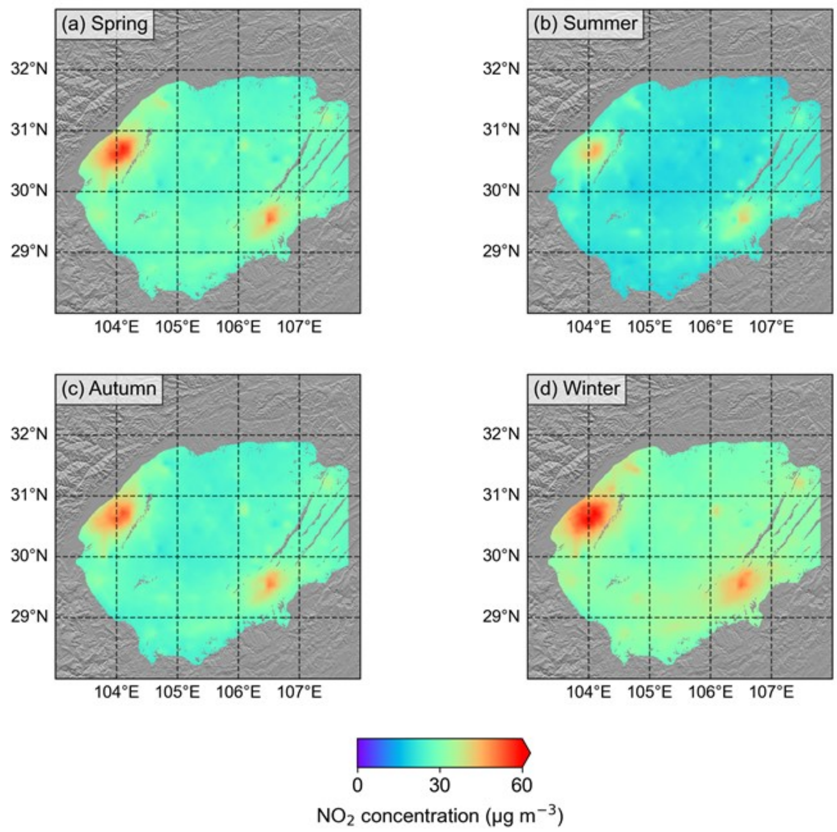

Figure R6 Spatial patterns for nitrogen dioxide (NO_2) concentrations over seasons during 2013-2019 from China High Air Pollutant (CHAP) dataset.

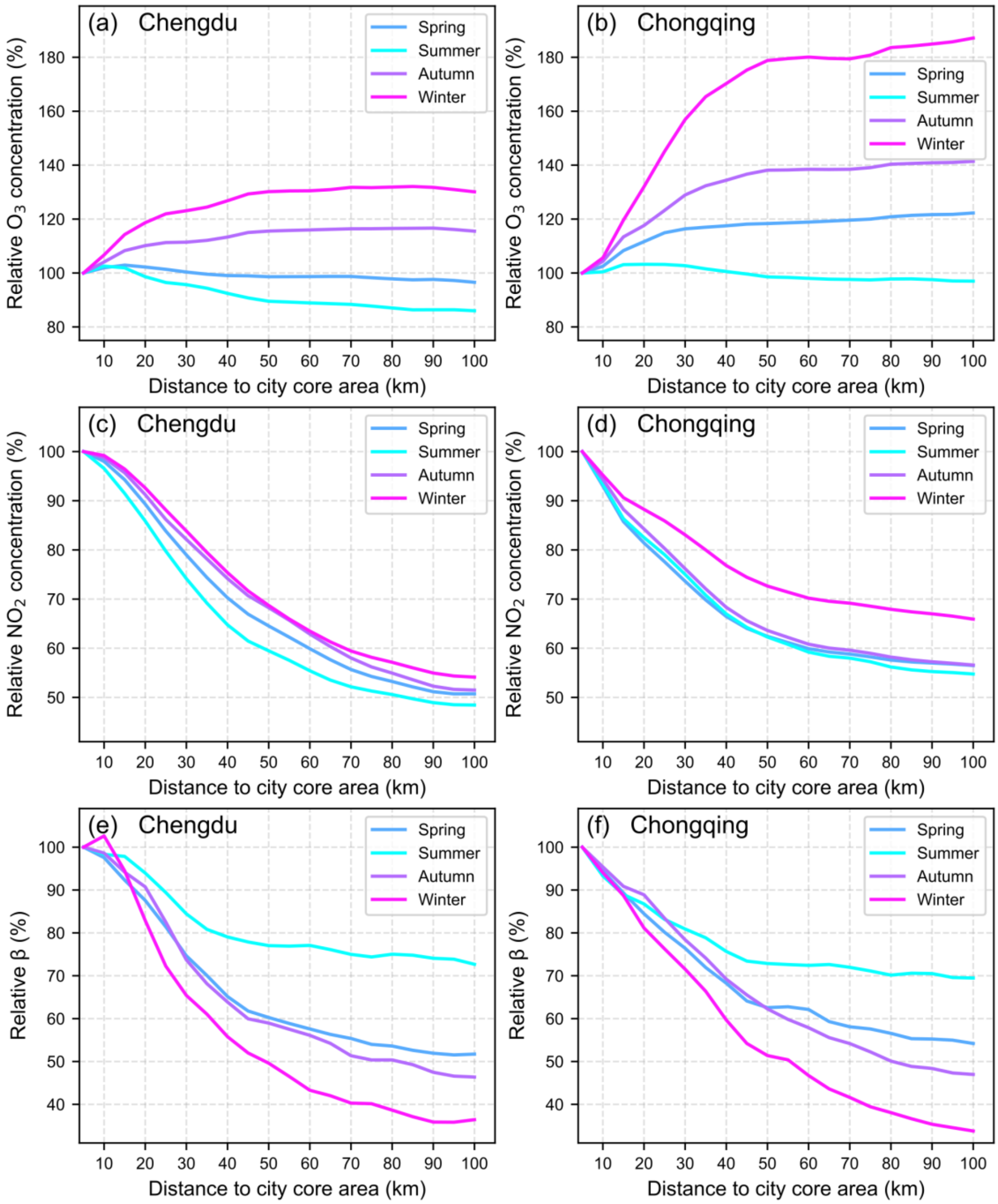

Figure R3 Relative O_3, NO_2, and β levels along with the core area distance in Chengdu (a, c, e) and Chongqing (b, d, f) over seasons. The average value within a 5-km radius centered to the city core area is used as the reference value. The gradients are referred within 5~100 km.

Table R1 Averaged concentrations within 5-km radius centered to the core area and the urban-rural gradients (to 100 km away) of relative O_3 and NO_2 levels, and NO_2/HCHO ratio.

City	Pollutant	Season							
		Spring		Summer		Autumn		Winter	
		core	slope	core	slope	core	slope	core	slope
Chengdu	O_3	110.1	-0.50	127.8	-1.67	62.3	1.02	47.9	1.88
	NO_2	55.7	-5.26	44.9	-5.07	51.0	-5.49	57.8	-5.34
	NO_2/HCHO	1.38	-4.78	0.67	-2.23	1.65	-5.34	2.43	-6.22
Chongqing	O_3	78.1	1.03	117.3	-0.56	48.0	2.80	27.5	6.49
	NO_2	49.3	-3.46	40.1	-3.91	47.8	-3.88	49.62	-3.17
	NO_2/HCHO	1.28	-4.15	0.65	-2.20	1.42	-5.66	2.53	-6.77

Note: Values on the left side are core area concentrations of O_3 and NO_2 (μg m^{-3}), while on the right side are concentration slopes (% $(10\ km)^{-1}$), representing the variability of O_3, NO_2 concentrations for every incremental 10 km radius distance centered to the city's core area. The NO_2/HCHO indicates a ratio of between NO_2 and HCHO during 2019-2023 from Sentinel-5P, their core area concentration levels and slopes are calculated in the same way as above.

2) The data processing approach is fundamentally flawed. In Line 120, the authors state that they "resampled the CAQRA data to a horizontal resolution of 1 km × 1 km via the LAF algorithm under CDO." However, such interpolation using CDO undoubtedly neglects the spatial heterogeneity of ozone as an air pollutant. This method lacks scientific rigor, especially considering that ozone distribution in urban centers typically exhibits strong spatial variability.

Author's response: Than you for your review efforts. we utilized new datasets to mitigate this omission. In 1 February 2025, a dataset named O_3 concentration dataset for mainland China (ChinaHighO_3) was released (Yang et al., 2025). This dataset used hybrid numerical dynamic modeling and machine learning works, produced 1 km horizontal resolution gridded data over China. Moreover, the ChinaHighO_3 belongs to a dataset series named CHAP (China High Air Pollutant) (Wei et al., 2023), all produced using similar methods as high-resolution air pollutant datasets. Most importantly, as we applied new datasets to our study, the results are highly consistent with previous findings (based on interpolated CAQRA) in terms of spatial patterns, still supporting the conclusion of a dipole-like ozone trapping pattern (**Figure R5, R6**):

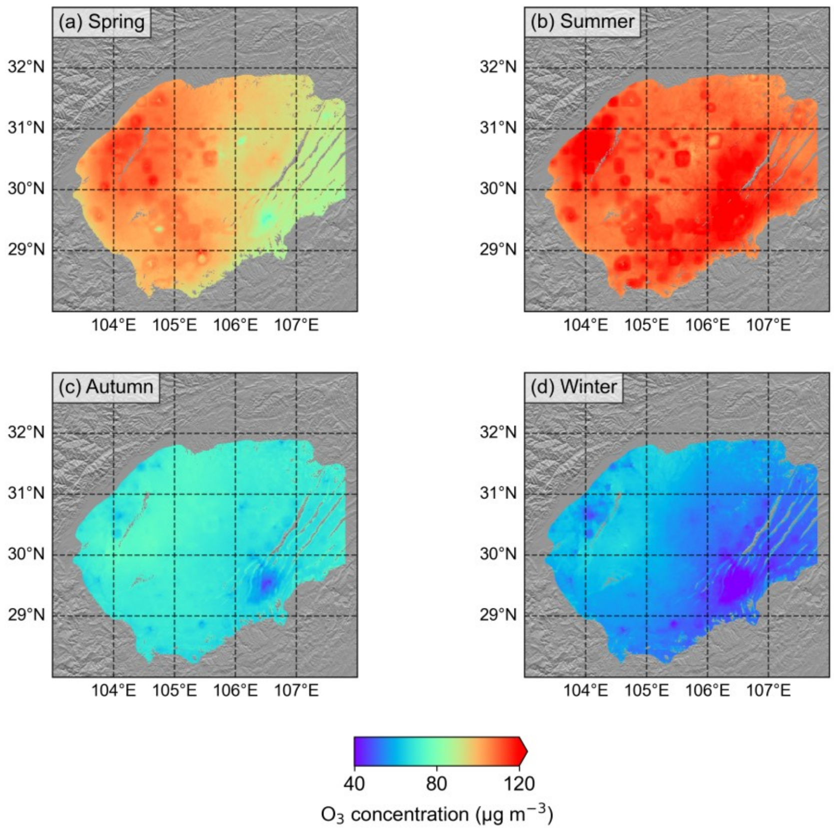

Figure R5 Spatial patterns for ozone (O_3) concentrations over seasons during 2013-2019 from ChinaHighO_3.

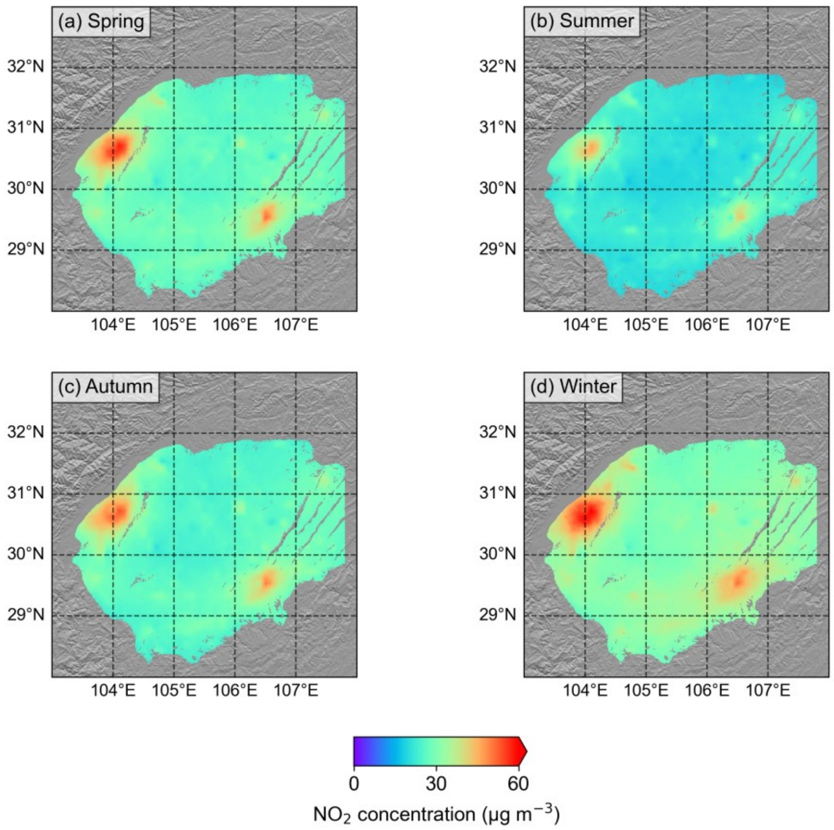

Figure R6 Spatial patterns for nitrogen dioxide (NO_2) concentrations over seasons during 2013-2019 from China High Air Pollutant (CHAP) dataset.

References:

Wei Jing, Li Zhanqing, Wang Jun, Li Can, Gupta Pawan, Cribb Maureen. Ground-level gaseous pollutants (NO_2, SO_2, and CO) in China: daily seamless mapping and spatiotemporal variations. *Atmospheric Chemistry and Physics*, **2023**, 23(2): 1511-1532. https://doi.org/10.5194/acp-23-1511-2023.

Yang Zeyu, Li Zhanqing, Cheng Fan, Lv Qiancheng, Li Ke, Zhang Tao, Zhou Yuyu, Zhao Bin, Xue Wenhao, Wei Jing. Two-decade surface ozone (O_3) pollution in China: Enhanced fine-scale estimations and environmental health implications. *Remote Sensing of Environment*, **2025**, 317: 114459. https://doi.org/https://doi.org/10.1016/j.rse.2024.114459.

3) Meteorological data: The authors used a limited number of ground-based meteorological stations for visibility and sunshine duration. However, for two key factors influencing ozone concentration—surface wind speed and surface temperature—they selected ERA5 reanalysis data and MODIS satellite data, respectively. This inconsistency is very confusing to the readers. The Sichuan Basin already has an extensive network of ground-based observation stations that provide comprehensive and direct measurements of 10-meter wind speed and 2-meter air temperature, which are significantly more accurate than satellite or reanalysis data. Without thorough validation of the reliability of surface wind and temperature data, especially the MODIS-derived temperature, I doubt the results derived from such a mixture of data sources.

Author's response: Thank you for your review efforts. The adoption of multi-source observations—including ground stations, satellite retrievals, and reanalysis data—was based on three key considerations. First, for sunshine duration and visibility, gridded data products are neither necessary nor appropriate for our objectives. Since the analysis specifically focuses on urban core areas, we compiled all available site observations as publishable as possible, to adopt these two variables within study regions to fulfill the required assessment. Then, for land surface temperature (LST), satellite remote sensing provides spatial coverage unattainable by ground sites. Given the research requirement for spatial continuity and comprehensive coverage, satellite-derived LST was selected. Finally, ERA5 reanalysis dataset was employed for windspeed observations due to its superior spatial continuity. Wind speed analysis necessitates statistically robust coverage over extended and uninterrupted domains—a criterion for which ERA5's reliability is well-established in the research community.

Given the above, we did not conduct integrated analysis of the three heterogeneous sourced datasets, but rather elucidated their respective potential influences from distinct analytical perspectives.

4) Another major concern is the lack of innovation. The mechanism proposed by the authors to explain the urban-rural ozone gradient has already been well-documented in previous studies. On the one hand, differences in anthropogenic activities determine the emission levels of ozone precursors—vehicular and industrial emissions in urban areas are generally much higher than in rural areas—resulting in distinct ozone formation regimes and consequently affecting ozone concentrations. On the other hand, urbanization-induced heat island effects may alter local atmospheric circulation, thereby influencing the transport of air pollutants. However, most of the conclusions in this study are drawn from basic statistical analysis and lack robust evidence to support the underlying mechanisms discussed.

Author's response: Thank you for your review efforts. We acknowledge that the urban-rural ozone gradient has already been well-documented in previous studies both via chemical regimes and meteorological conditions. However, what we tried to convey to research community is the asymmetric dipole-like ozone trap phenomenon under a detailed-targeted analysis. As shown in the following **Figure R3** based on in-situ climatology and seasonality, although there are high NO_2 centers both in the core area of Chengdu and Chongqing, the pronounced negative O_3 gradients only appeared during winter. In summer, the urban-rural gradients of O_3 are even slightly positive. Since our study adopted the newly released high resolution O_3 datasets in 1 February 2025, this analysis with such a high-level spatial-temporal continuity could substantially convey certain new information to the research community.

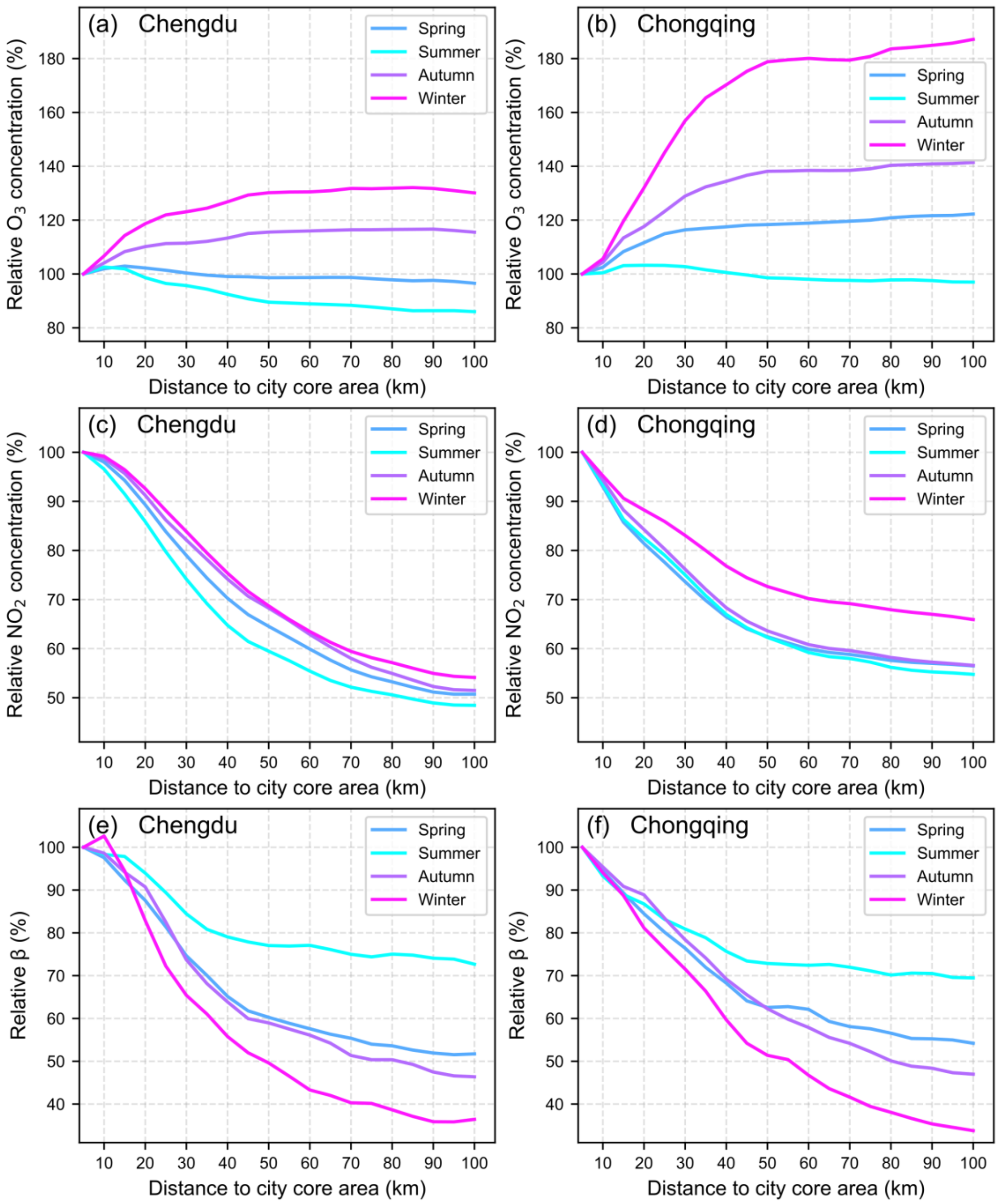

Figure R3 Relative O_3, NO_2, and β levels along with the core area distance in Chengdu (a, c, e) and Chongqing (b, d, f) over seasons. The average value within a 5-km radius centered to the city core area is used as the reference value. The gradients are referred within 5~100 km.

5) The study also fails to fundamentally address the core question posed in the title: "What makes the less urbanized city a deeper ozone trap: implications from a case study in the Sichuan Basin, southwest China." The mechanism proposed in Figure 9 is merely a simplistic textbook-level extension. The authors neither tested the feasibility of this hypothesis using box modeling approaches such as PBM-MCM nor validated the proposed mechanism through three-dimensional chemical transport models (WRF-Chem or WRF-CMAQ). Such an unvalidated and systematically unassessed hypothesis falls well below the standards expected by Atmospheric Chemistry and Physics (ACP).

Author's response: From a technical simulation perspective, there remain significant challenges to adopt a congruent modelling work to the study in case the spatial analysis is under a 1-km horizontal resolution. Theoretically, the 1-km resolution modeling is feasible, but the currently available emission inventories only provide data at 0.25-degree (≈27 km) resolution. Not only that, key technical also constraints include: 9~12 km simulations demonstrate marginal usability in practical applications based on current reported studies and our experience. Spatial interpolation below 9 km resolution introduces substantial deviation amplification. Then, result matching becomes increasingly unreliable with finer-scale downscaling. The core challenges stem from resolution disparity between modeling capacity (1 km) and input data quality (27 km). Also, error propagation through multi-stage downscaling processes, as well as unquantifiable uncertainty in cross-resolution validation.

Moreover, our research purpose was not to fully resolve the issue about this featured ozone spatial behavior. The novelty of our study lies in identifying the characterized long-term spatial behavior of the dipole-like asymmetric ozone trap over the two mega cities of the Sichuan Basin, with the causal analysis merely providing certain implications.

Given the above, we sincerely request you please to reconsider re-evaluating our manuscript.

6) Line 245: The authors mention TROPOMI NO_2 and HCHO column data and claim that the HCHO data do not exhibit featured spatial patterns. This statement is completely incorrect. Formaldehyde (HCHO), due to its short atmospheric lifetime, high yield from the oxidation of non-methane volatile organic compounds (NMVOCs), and detectability by satellite instruments, has been widely used to characterize the intensity of anthropogenic activities. Moreover, meteorology-corrected TROPOMI HCHO column data can effectively reflect long-term trends in anthropogenic VOC emissions, which is particularly important for explaining ozone variations in urban areas such as Chengdu and Chongqing.

Author's response: Thank you for you review efforts. In the revision, we have analyzed the ratio changes of formaldehyde to NO_2 from VOC-limited to NO_x-limited conditions across the gradient for the two cities seasonally, some new findings to explain the ozone trap discrepancy were obtained. Shown as β (ratio of NO_2 to formaldehyde) over the basin (**Figure R1**), the concentration of NO_2 is lower than formaldehyde in all areas during summer. But in winter, the two megacities exhibited concentration centers where NO_2 was significantly higher than formaldehyde.

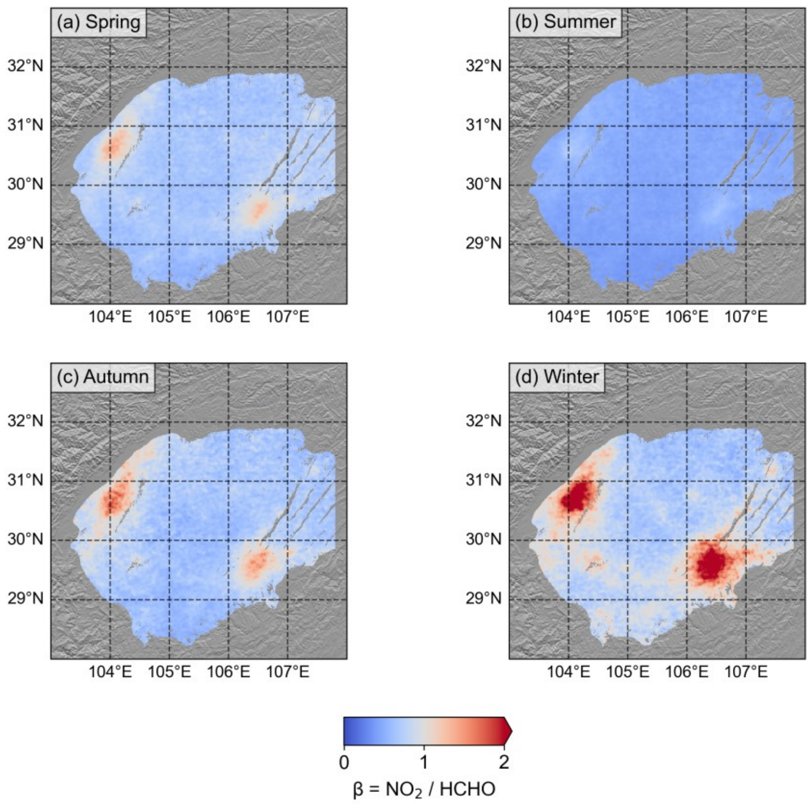

Figure R1 Spatial patterns for the number concentration ratio of NO_2 to formaldehyde over seasons during 2019-2023.

Then, we calculated β across different ISA (impervious surface area) levels (**Figure R2**). With increasing ISA, the summer β steadily rises within the range of 0.5~0.6, while the winter values reach a level of 1.2~1.8. Combined with the spatial pattern maps mentioned above, it can be clearly observed that the spatial distribution characteristics of β are primarily determined by NO_2.

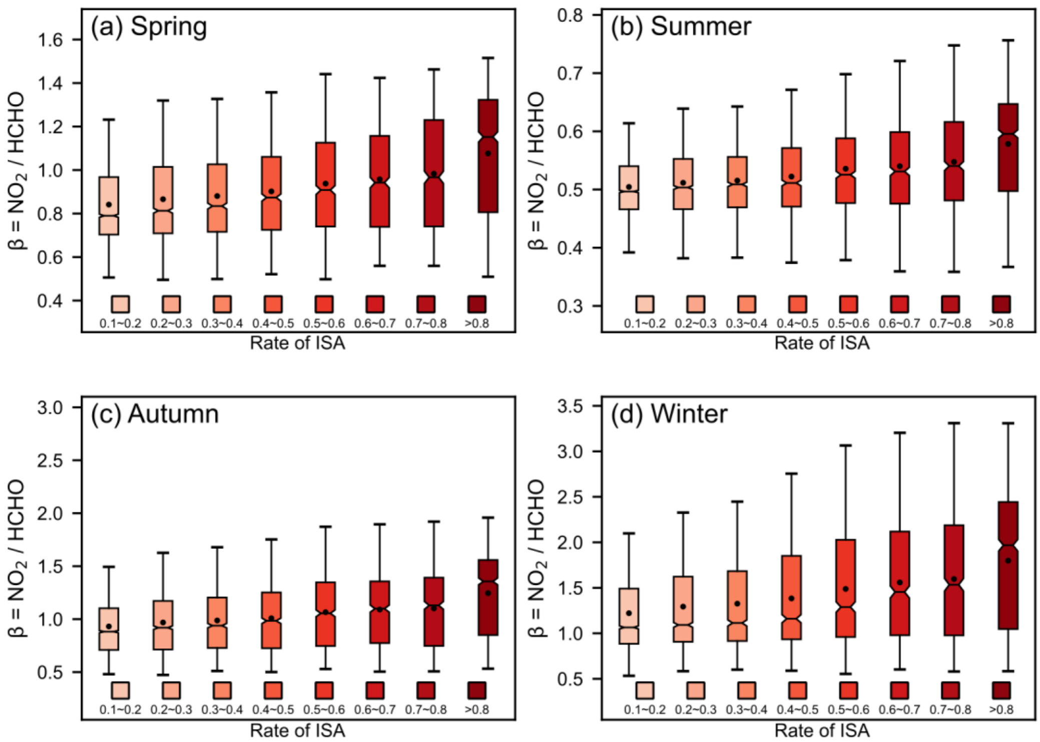

Figure R2 Response of β to rate of ISA over seasons during 2019-2023. The solid lines in the mid of the boxes are medians, the notches are the 95% confidence interval of the medians, the black dots are mean values, and the whiskers are interquartile ranges.

Further, we examined the urban-rural gradients of β across the two cities. It is found that although the gradient of NO_2's urban-rural decay curve in Chongqing is smaller than that in Chengdu, the decay curves of β are nearly identical. Notably in winter, the decay level at 100 km from the urban center even exceeds that of Chengdu (**Figure R3**). Quantitatively, during winter, the core area level and urban-rural slope of NO_2 are 57.8 μg m^{-3} and -5.34% in Chengdu, while 49.6 μg m^{-3} and -3.17% in Chongqing, respectively (**Table R1**). When we examine the β instead of solely examine the pattern of NO_2, it is found that while Chengdu demonstrates a steeper urban-rural decay rate in NO_2 concentration compared to Chongqing, the β reveals an inverse pattern—both core area levels and decay rates are higher in Chongqing. Therefore, under the condition of ozone gradient 6.49% (Chongqing) versus 1.88% (Chengdu), although β does not play a decisive role, the spatial behavior of β likely contributes a certain portion to Chongqing's more pronounced ozone sink effect relative to Chengdu.

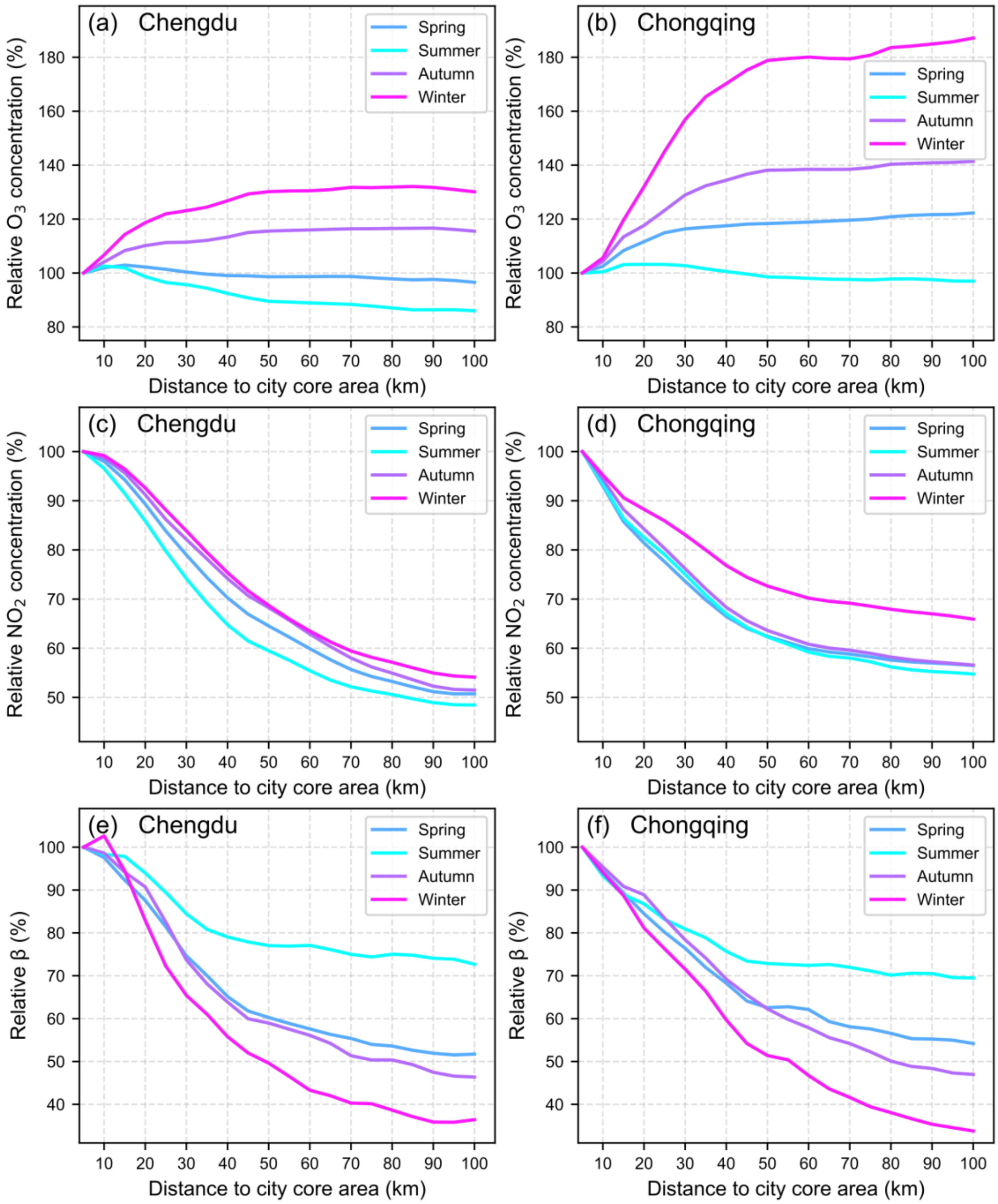

Figure R3 Relative O_3, NO_2, and β levels along with the core area distance in Chengdu (a, c, e) and Chongqing (b, d, f) over seasons. The average value within a 5-km radius centered to the city core area is used as the reference value. The gradients are referred within 5~100 km.

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Chongqing	O_3	78.1	1.03	117.3	-0.56	48.0	2.80	27.5	6.49
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7) The authors utilized the Chinese Air Quality Reanalysis (CAQRA) dataset for their analysis, yet they did not validate the accuracy or reliability of this dataset using surface ozone concentration observations from the China National Environmental Monitoring Center (CNEMC). How do the authors ensure the quality of the CAQRA data and its capability to accurately capture ozone concentrations in the Sichuan Basin?

Author's response: Thank you for your review efforts. we utilized new datasets to mitigate this omission. In 1 February 2025, a dataset named O_3 concentration dataset for mainland China (ChinaHighO_3) was released (Yang et al., 2025). This dataset used hybrid numerical dynamic modeling and machine learning works, produced 1 km horizontal resolution gridded data over China. Moreover, the ChinaHighO_3 belongs to a dataset series named CHAP (China High Air Pollutant) (Wei et al., 2023), all produced using similar methods as high-resolution air pollutant datasets. The validation of the dataset including observations from ground sites over China is well-documented in the dataset's reference paper.

References:

Wei Jing, Li Zhanqing, Wang Jun, Li Can, Gupta Pawan, Cribb Maureen. Ground-level gaseous pollutants (NO_2, SO_2, and CO) in China: daily seamless mapping and spatiotemporal variations. *Atmospheric Chemistry and Physics*, **2023**, 23(2): 1511-1532. https://doi.org/10.5194/acp-23-1511-2023.

Yang Zeyu, Li Zhanqing, Cheng Fan, Lv Qiancheng, Li Ke, Zhang Tao, Zhou Yuyu, Zhao Bin, Xue Wenhao, Wei Jing. Two-decade surface ozone (O_3) pollution in China: Enhanced fine-scale estimations and environmental health implications. *Remote Sensing of Environment*, **2025**, 317: 114459. https://doi.org/https://doi.org/10.1016/j.rse.2024.114459

8) The TROPOMI NO_2 and HCHO products and quality control process are not mentioned in Methods section.

Author's response: Revision done. The related information is supplemented appropriately.

9) The manuscript contains numerous grammatical and phrasing errors, making it difficult to follow.

Author's response: Revision done. The grammatical and phrasing errors of the manuscript's language are revised thoroughly.

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