

Dear Editors and Reviewers,

Thank you very much for your careful review on our manuscript egosphere-2024-268. We appreciate very much your encouraging comments and constructive suggestions on improving our manuscript. We have accordingly made the careful and substantial revisions. The revised portions are marked up in the revised manuscript. Please find our point to point responses to the reviewers' comments as follows:

Responses to the reviewer #2

[1. Line 107: *“The portion of tropospheric O₃ concentrations originating from the stratosphere (O₃S)”, The author may need more description to substantiate this claim.*]

Response 1: Thanks to the reviewers for the valuable suggestion on our manuscript. According to the reviewer's comment, we added more description to substantiate the O₃S as follow (lines 111-117):

To evaluate the reproducibility of the WRF-Chem simulations for the stratospheric O₃ intrusion, the portion (O₃S) of tropospheric O₃ concentrations originating from the stratosphere was applied to compare with our simulation results. The stratospheric tracer tagging method in global chemistry models was used to track the transport of stratospheric O₃ to the troposphere by releasing stratospheric tracers (Barth et al., 2012; Chang et al., 2023). The tracer was set to 1 above the tropopause, and only physically transported and chemically decayed in the troposphere without chemical production (Chang et al., 2023; Ni et al., 2019). The O₃S in the troposphere was calculated by multiplying the concentrations of O₃ at the tropopause and the stratospheric tracers.

References

Barth, M. C., Lee, J., Hodzic, A., Pfister, G., Skamarock, W. C., Worden, J., Wong, J., and Noone, D.: Thunderstorms and upper troposphere chemistry during the early stages of the 2006 North American Monsoon. *Atmospheric Chemistry and Physics*, 12(22), 11003–11026, doi:<https://doi.org/10.5194/acp-12-11003-2012>, 2012.

Chang, F., Li, J., Li, N., and Liao, H.: Stratospheric intrusion may aggravate widespread ozone pollution through both vertical and horizontal advections in eastern China during summer. *Frontiers in Environmental Science*, 10, 2756, doi:<https://doi.org/10.3389/fenvs.2022.1115746>, 2023.

Ni, Z. Z., Luo, K., Gao, X., Gao, Y., Fan, J. R., Fu, J. S., and Chen, C. H.: Exploring the stratospheric source of ozone pollution over China during the 2016 Group of Twenty summit. *Atmospheric Pollution Research*, 10(4), 1267–1275, doi:<https://doi.org/10.1016/j.apr.2019.02.010>, 2019.

[2. Line 125 to 127: *“The differences in --- in the SI event”* , How sensitive is the "quantitative effect" to the ozone lateral boundary conditions used in the control experiments? It is best to provide information on how to set the ozone lateral boundary conditions at the top level of the model in WRF-CHEM.]

Response 2: Many thanks for the constructive suggestions on our manuscript. Since the stratospheric chemistry is not included in the WRF-Chem, an upper boundary condition (UBC) scheme derived from the Whole Atmosphere Community Climate Model was used to provide the initial and boundary chemical conditions for the stratosphere (including but not only the top level of the model) in the model (Barth et al., 2012). The UBC scheme could generate all key chemical species in the stratosphere, enabling the WRF-Chem to simulate the stratospheric intrusion processes more accurately (Barth et al., 2012; Lamarque et al., 2012; Zhao et al., 2021).

Following the reviewer's suggestion, we have added the above discussion to lines 130-134 in the revised manuscript.

More explanation about the UBC scheme here: According to the user guide of WRF-Chem, the UBC scheme will specify the O₃, NO, NO₂, HNO₃, CH₄, CO, N₂O, and N₂O₅ concentrations at the top of the model. These stratospheric concentrations override the original values as defined in the idealized chemical profile. From the top level of the model down to the tropopause, the concentrations are relaxed, using a 10-day time constant, to fixed values.

References

- Barth, M. C., Lee, J., Hodzic, A., Pfister, G., Skamarock, W. C., Worden, J., Wong, J., and Noone, D.: Thunderstorms and upper troposphere chemistry during the early stages of the 2006 North American Monsoon. *Atmospheric Chemistry and Physics*, 12(22), 11003–11026, doi:<https://doi.org/10.5194/acp-12-11003-2012>, 2012.
- Lamarque, J. F., Emmons, L. K., Hess, P. G., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C. L., Holland, E. A., Lauritzen, P. h., Neu, J., Orlando, J. J., Rasch, P. J., and Tyndall, G. K.: CAM-chem: Description and evaluation of interactive atmospheric chemistry in the Community Earth System Model. *Geoscientific Model Development*, 5(2), 369–411, doi:<https://doi.org/10.5194/gmd-5-369-2012>, 2012.
- Zhao, K., Hu, C., Yuan, Z., Xu, D., Zhang, S., Luo, H., Wang T., and Jiang, R. A modeling study of the impact of stratospheric intrusion on ozone enhancement in the lower troposphere over the Hong Kong regions, China. *Atmospheric Research*, 247, 105158, doi:<https://doi.org/10.1016/j.atmosres.2020.105158>, 2021.

[3. Line 130: *“Fig.S2”* , *Is it the average of all observations in domain 03? Please explain in detail.*]

Response 3: Our meteorological and environmental observation data were collected from the China Meteorological Observation Network and the National Air Quality Monitoring Network, and almost all observation stations in the networks are located in cities and towns. Therefore, we calculated the averages of meteorological elements and O₃ concentrations observed at all stations in the innermost domain and the averages of simulated meteorological elements and O₃ concentrations in the model grids corresponding to the station locations to conduct the modeling validation.

Following the reviewer’s suggestions and comments, we have added the above discussion to lines 144-146 in the revised manuscript.

[4. Line 138 to 139: *“Therefore, our simulation results are available and convincing.”* , *Please see the first question above*]

Response 4: Thanks to the reviewer for pointing out our inappropriate expression. We have corrected this sentence in lines 154-155 as follows:

All these evaluations indicate that our model simulations performed well in reproducing the variations of O₃ and meteorological parameters during the SI process.

[5. Line 148: *“Fig. S4b”* , *Is the value of air temperature in the picture negative or positive?*]

Response 5: The values of air temperature in Fig. S4b are all negative, indicating the colder stratospheric air mass invade to the troposphere. We have slightly modified Figure S4 to enhance its readability:

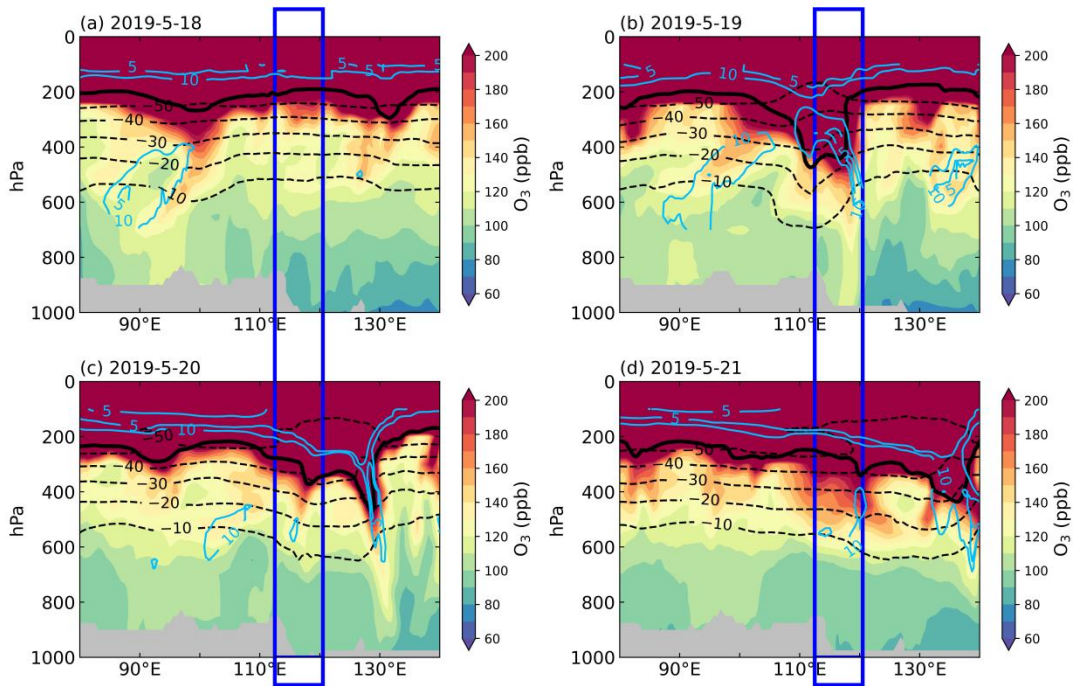


Figure S4: Latitudinal vertical sections of O₃ concentrations (color contours) averaged over 32°N–40°N from the MERRA2 data during May 18–21, 2019. Black solid lines indicate the dynamical tropopause labeled by PV=2. The dashed black lines represent air temperature (°C), the solid blue lines represent relative humidity (%), and the blue rectangles mark the NCP region.

[6. Line 148: Line 172 to 176: “the intense --- to the near-surface layer” , Since both subsidence and ascending motion occur in extratropical cyclone systems, stratospheric intrusion ozone reaching the surface will rapidly diffuse and be carried back to the upper troposphere. Therefore, such events are difficult to observe.]

Response 6: Thanks for the reviewer’s helpful suggestions on our manuscript.

The peripheric subsidence and central ascending motion meteorologically occur in extratropical cyclones with a typical horizontal scale of 1000km. Therefore, parts of the stratospheric intrusion ozone reaching the surface can rapidly diffuse and are rarely carried back to the upper troposphere. While the center of the Northeast Cold Vortex and extratropical cyclone that prevailed the ascending motion was over Northeast China. Our studied region, the North China Plain (NCP), was located at the southwest periphery of the Northeast Cold

Vortex and the stimulated extratropical cyclone system and in the sinking zone of the vertical circulation of this system. Meanwhile, under the influence of the horizontal circulation of this system, the NCP experienced both the control of subsidence motions and the imposition of strong northwest winds. The intensity of the horizontal wind was much higher than the vertical velocity. Therefore, the invading stratospheric O₃ tongue tilted to the southwest and reached the surface under the comprehensive effect of vertical and horizontal winds (Figure 3), and then the stratospheric O₃ was gradually transported downstream in the southwest direction (Figure 8).

[7. Line 215: “week” , weak??]

Response 7: Thanks to the reviewer for pointing out our oversight. We have corrected this word to line 231 in the revised manuscript.

[8. Line 225: “Figure 5” , How many weather observatories are there and how many ozone and carbon monoxide observatories are there? Why do the authors use regionally averaged observations? Have the authors looked at single-site observations of ozone in particular?]

Response 8: The observed meteorological elements from 639 sites and O₃ and CO concentrations from 440 environmental observatories in the innermost domain (domain 03) were applied in Figure 5. Following the reviewers' suggestion, the changes in observed meteorological and environmental elements from the representative sites SJZ and JN (The red dots in Fig. S3) were examined in Fig. S6. The results showed that the diurnal cycles of O₃ concentration presented noteworthy characteristics compared with the regional averages. The SJZ in the northwest received stratospheric O₃ earlier and reached the spike at 10:00 LST on May 19. Then the O₃ concentrations gradually decreased under the influence of strong winds but still maintained a high level in the early morning of May 20. The JN city in the southeast was affected by the stratospheric intrusion later. While under meteorological conditions conducive to the dissipation of pollutants (wind speed up to 8 m·s⁻¹), higher O₃ concentrations than the previous day were still observed, reflecting the additional contribution of stratosphere intrusion to near-surface O₃.

We have added the above discussion to lines 256-263 in Section 3.2 and Fig. S6 was added in the supplement.

Furthermore, we added the number of meteorological and environmental observation sites to line 109 in the revised manuscript.

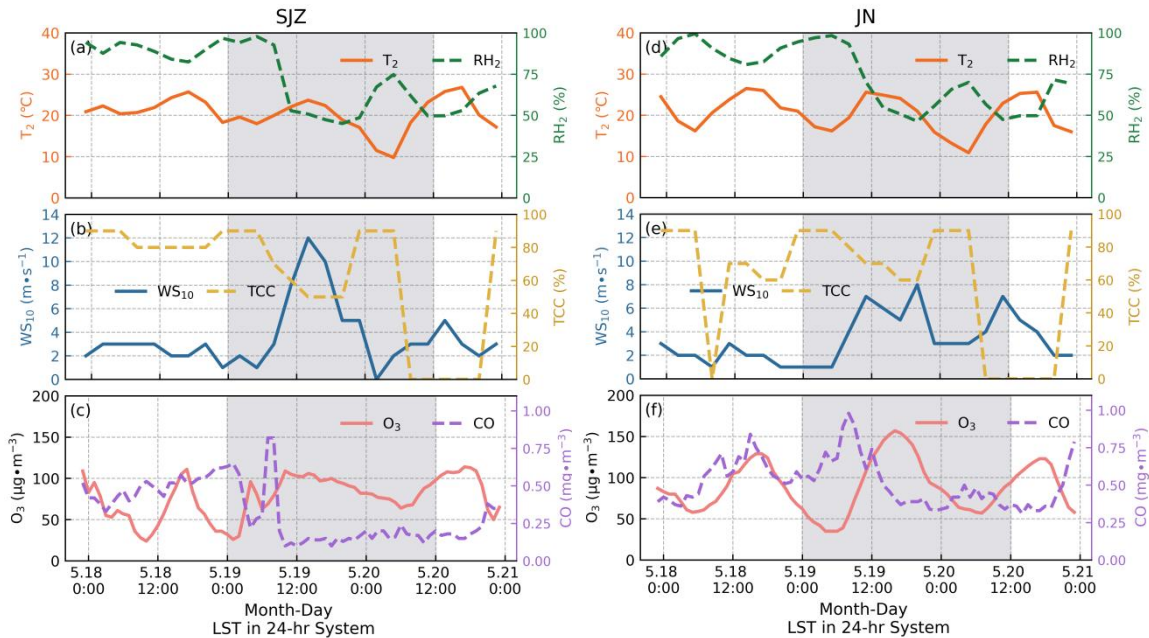


Figure S6: Temporal variations of (a, d) T_2 , RH_2 , (b, e) WS_{10} , and total cloud cover (TCC), (c, f) near-surface O_3 and CO concentrations in representative cities SJZ and JN from the observations in the NCP region. The shaded areas mark the periods of the SI to the near-surface layer.

[9. Line 258: “vertical mixing (VMIX) --- vertical advection (ADVZ)”, What is the difference between VMIX and ADVZ?]

Response 9: In the integrated process analysis of the WRF-Chem model, the VMIX term represents the impact of vertical entrainment mixing caused by turbulent motion on pollutants, which mainly occurs within the atmospheric boundary layer. The ADVZ term reflects the vertical advection transport of pollutants driven by vertical wind.

[10. Line 266: “Fig. 6”, How did the authors choose the IPR times shown in Figure 6 for SJZ and JN?]

Response 10: The horizontal northwest wind drives the stratospheric O₃ that invades the surface to present notable characteristics of downstream transport (Figure S3), causing the temporal variation of the contributions of ADVZ to O₃ in the boundary layer to be unsynchronized in SJZ and JN. Therefore, we chose different times to discuss the IPR in SJZ and JN. Furthermore, we considered that discussing the IPR at all times during the stratospheric intrusion would be a little redundant in plotting and writing. Therefore, based on the temporal variations of the simulated contribution of stratospheric O₃ to near-surface O₃ in SJZ and JN (Figure 8b and d), we selected several discontinuous but representative times that can reflect the temporal variation characteristics of the IPR during the intrusion process to conduct the discussions.

[11. Line 280: “without o3 pollution” , confused]

Response 11: Thanks for the helpful comment on our manuscript. We have deleted “without O₃ pollution” in the revised manuscript. What we want to express here is:

Although the intrusion of stratospheric O₃ has the potential to augment surface O₃ levels, rapid dispersion and removal facilitated by the peripheral horizontal winds of the cyclone mitigated this impact. Therefore, steep rises in surface O₃ concentrations were conspicuously absent during the SI process, and no sustained regional O₃ pollution emerged over the NCP. This conclusion could be reflected in Figure 5c, which shows that the regional averaged O₃ concentration over the NCP was around 100 μg·m⁻³ during the SI period. Only the hourly O₃ concentrations in part of the sites exceeded 160 μg·m⁻³ temporarily, and none of them reached 200 μg·m⁻³ exceeding the standard of O₃ pollution.

According to the reviewer’s comment, we have added the following discussion to lines 302-305 of the revised manuscript:

Namely, although the intrusion of stratospheric O₃ has the potential to augment surface O₃ levels, rapid dispersion and removal facilitated by the peripheral horizontal winds of the cyclone mitigated this impact. Therefore, the strong intrusion of stratospheric O₃ into the near-surface layer during the SI process without surface O₃ concentrations exceeding the hourly standard of O₃ pollution of 200 μg·m⁻³ over the NCP (Fig. 5c).

[12. Line 290: “Figure 7” , Is it the model simulation result on domain 03?]

Response 12: Yes, Figure 7 presents the simulated differences in ADVZ contribution to O₃ in different vertical layers between the innermost domain of CASE_{STRO3} (the control experiment) and CASE_{noSTRO3} (the simulation experiment), indicating the temporal variations of stratospheric O₃ transport to various atmospheric layers in the troposphere.