

## General Comments:

The manuscript, titled "Extreme Weather's Impact on Ozone Pollution in China," investigates Ozone (O<sub>3</sub>) pollution in China, emphasizing the overlooked role of natural processes. It analyzes the effects of extreme weather events in the September 2022 Pearl River Delta (PRD) case, which led to prolonged high O<sub>3</sub> levels and extreme temperatures due to the Subtropical High and typhoon peripheries. Using field measurements, machine learning, and numerical modeling, the study examines the influence of weather-induced natural processes on O<sub>3</sub> pollution. Findings indicate that hot weather intensifies photochemical reactions, adding 10.8 ppb of O<sub>3</sub> compared to standard conditions, with temperature as the primary factor. The study also highlights the role of biogenic volatile organic compounds, particularly isoprene, in O<sub>3</sub> production. Additionally, it explores the chemical mechanisms behind isoprene's contribution. Notably, a nearing typhoon enhances cross-regional O<sub>3</sub> transport via stratosphere-to-troposphere exchange (STE). Model simulations suggest STE-induced O<sub>3</sub> reaching a maximum of approximately 8 ppb at the PRD surface. In summary, the study underscores the significant impact of natural processes exacerbated by extreme weather on O<sub>3</sub> pollution and offers insights for O<sub>3</sub> pollution control amidst global warming. Overall, the study is well-structured and provides valuable insights into the topic. However, there are some areas that require attention before publication. Therefore, I would recommend acceptance to this manuscript after the following minor revisions were made and the questions were answered.

**General Response to the reviewer:** Thank you very much for your valuable comments and suggestions. Your positive comments/suggestions have motivated us to improve the manuscript. Now, we have carefully revised the manuscript based on all your questions/suggestions, and hope the correction will meet with approval. We have marked the revised sentences in red color in the manuscript. Below is the point-to-point response.

## Specific Comments:

1. Page 3, Line 60: The phrase "sun-light driven" should be corrected to "sunlight-driven."

**Reply:** Thanks for the careful reminder. We have revised it as suggested.

2. Page 3, Line 66 & 86: The phrase " biogenic volatile carbon" should be corrected to " biogenic volatile organic compound". And please unify the abbreviation of "BVOC".

**Reply:** Thanks for pointing out our mistakes. We have changed them to "biogenic volatile organic compound". And we also unified the abbreviation of BVOC throughout the manuscript.

3. Page 3, Line 88: Abbreviation of “HOx” should be defined.

**Reply:** Thanks for the careful review. Abbreviation of HOx is now defined, hydrogen oxide (HOx) radicals.

4. Page 4, Line 101: The phrase “nature processes” should be corrected to “natural processes”.

**Reply:** Thanks. We revised the phrase “nature processes” to “natural processes” as.

5. Page 4, Line 105: Abbreviation of “CO” should be defined.

**Reply:** Revised as suggested. The phrase is corrected to “carbon monoxide (CO)”

6. Page 4, Line 107-109: After a semicolon (;), the following word should typically start with a lowercase letter, therefore, “Vegetation-released” and “An” should change to “vegetation-released” and “an”.

**Reply:** Thanks. We now used lowercase letter as suggested.

7. Page 4, Line 109: The phrase "largescale " should be corrected to "large-scale”.

**Reply:** Revised as suggested.

8. According to the ACP’s submission guide to author, the abbreviation "Fig." should be used when it appears in running text and should be followed by a number unless it comes at the beginning of a sentence. Labels of panels must be included with brackets around letters being lower case (e.g. (a), (b), etc.).

a. Page 5, Line 137: “Figure 1a” should be “Fig. 1(a)”.

b. Page 6, Line 161: “Figure 1a” should be “Fig. 1(a)”.

c. Page 7, Line 198: “Figure S1” should be “Fig. S1”.

d. Page 9, Line 272: “Figure S2” should be “Fig. S2”.

e. Page 9, Line 280: “Figure S3” should be “Fig. S3”.

f. Page 9, Line 293: “Figure 1b” should be “Fig.1(b)”.

g. Page 10, Line 298: “Figure S4” should be “Fig. S4”.

h. Page 10, Line 300: “Figure 1b” should be “Fig. 1(b)”.

i. Page 10, Line 303: “Figure 1c” should be “Fig. 1(c)”.

j. Page 10, Line 305: “Figure 1c” should be “Fig. 1(c)”.

k. Page 10, Line 313: “Figure 1d” should be “Fig. 1(d)”.

l. Page 10, Line 323: “Figure 2” should be “Fig. 2”.

m. Page 12, Line 355: “Figure 3a” should be “Fig. 3(a)”.

n. Page 12, Line 359: “Figure 3b” should be “Fig. 3(b)”.

- o. Page 13, Line 381: "Figure 4" should be "Fig. 4".
- p. Page 14, Line 419: "Figure S5" should be "Fig. S5".
- q. Page 14, Line 425: "Figure S5" should be "Fig. S5".
- r. Page 14, Line 433: "Figure 5a" should be "Fig. 5(a)".
- s. Page 14, Line 436: "Figure S5" should be "Fig. S5".
- t. Page 14, Line 441: "Figure S6" should be "Fig. S6".
- u. Page 14, Line 445 "Figure 5b" should be "Fig. 5(b)".
- v. Page 14, Line 446 "Figure 5c" should be "Fig. 5(c)".
- w. Page 14, Line 446 "Figure 5d" should be "Fig. 5(d)".
- x. Page 15, Line 450 "Figure 5e" should be "Fig. 5(e)".
- y. Page 15, Line 451 "Figure 5f" should be "Fig. 5(f)".
- z. Page 15, Line 454 "Figure S7-S9" should be "Fig. S7-S9".
- aa. Page 15, Line 457 "Figure 6" should be "Fig. 6".
- bb. Page 15, Line 474 "Figure 7" should be "Fig. 7".

**Reply:** We appreciate so much for the careful review. We have consistently employed the abbreviation "Fig." throughout the text. Besides, labels of panels are also included with brackets around letters being lower case.

**9. Page 5, Line 144:**

a. In the sentence "meteorological parameters (surface winds, temperature and solar radiation)", some meteorological parameters including precipitation and relative humidity were not mentioned.

**Reply:** Thanks for the reminder. We added relative humidity, and precipitation here.

b. When and for how long were online observations conducted for O<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub>, CO, VOC components, and meteorological parameters at this observation base?

**Reply:** We added the following sentence to make it clear, "All the data are collected at the HZ Base from Sept. 1st to Sept. 30th, with a time resolution of 1 hour."

**10. Page 5, Line 151: What and how many VOC species were measured?**

**Reply:** The instrument could detect 56 volatile VOCs species (Table R1). We added the following description here "The target compounds of the instrument were the 56 VOCs designated as photochemical precursors by the US Environmental Protection Agency (EPA). The gas standards utilized were identical to those employed by the US EPA Photochemical Assessment Monitoring Stations (PAMS). More details were documented in our previous paper (Zou et al., 2015)."

Table R1\* The calibration curves and detection limits of VOC species

Target compound	Calibration curve	Correlation coefficient	Detection limit (ppbv)
Ethene	$y = 1.0188x + 0.2659$	0.997	0.07
Acetylene	$y = 1.0409x + 0.1756$	0.998	0.08
Ethane	$y = 1.0162x + 0.2891$	0.997	0.08
Propene	$y = 0.9959x + 0.1506$	0.999	0.07
Propane	$y = 0.9824x + 0.2082$	0.998	0.09
i-Butane	$y = 0.9753x + 0.3785$	0.994	0.05
1-Butene	$y = 0.9587x + 0.3641$	0.994	0.06
n-Butane	$y = 0.9776x + 0.3718$	0.994	0.05
trans-2-Butene	$y = 0.9746x + 0.2747$	0.997	0.05
cis-2-Butene	$y = 0.9834x + 0.1606$	0.999	0.06
i-Pentane	$y = 0.9753x + 0.2135$	0.998	0.07
1-Pentene	$y = 0.919x + 0.1626$	0.998	0.05
n-Pentane	$y = 0.9557x + 0.2038$	0.984	0.07
Isoprene	$y = 1.0304x + 0.1653$	0.998	0.07
trans-2-pentene	$y = 0.9753x + 0.2135$	0.998	0.07
cis-2-pentene	$y = 0.9557x + 0.2038$	0.984	0.07
2,2-Dimethylbutane	$y = 0.9731x + 0.1971$	0.998	0.07
Cyclopentane	$y = 0.9993x + 0.1412$	0.997	0.06
2,3-Dimethylbutane	$y = 0.919x + 0.1626$	0.999	0.07
2-Methylpentane	$y = 0.9557x + 0.2038$	0.984	0.07
3-Methylpentane	$y = 0.9753x + 0.2135$	0.998	0.07
1-Hexene	$y = 0.9700x + 0.3300$	0.995	0.05
n-Hexane	$y = 0.9915x + 0.2626$	0.997	0.06
Methylcyclopentane	$y = 0.9749x + 0.1832$	0.999	0.07
2,4-Dimethylpentane	$y = 0.9993x + 0.1412$	0.999	0.05
Benzene	$y = 0.9753x + 0.2835$	0.997	0.06
Cyclohexane	$y = 0.9841x + 0.2744$	0.997	0.07
2-methylhexane	$y = 0.9744x + 0.2979$	0.996	0.05
2,3-dimethylpentane	$y = 0.9779x + 0.2953$	0.997	0.05
3-methylhexane	$y = 0.9735x + 0.3374$	0.995	0.05
2,2,4-trimethylpentane	$y = 0.9696x + 0.3947$	0.994	0.05
n-Heptane	$y = 0.9678x + 0.3635$	0.994	0.05
Methylcyclohexane	$y = 0.9819x + 0.3629$	0.995	0.05
2,3,4-trimethylpentane	$y = 0.9691x + 0.3994$	0.994	0.04
Toluene	$y = 0.9696x + 0.3397$	0.995	0.05
2-methylheptane	$y = 0.9603x + 0.4835$	0.990	0.04
3-methylheptane	$y = 0.9625x + 0.4550$	0.991	0.04
n-Octane	$y = 0.9524x + 0.5082$	0.989	0.04
Ethylbenzene	$y = 0.9629x + 0.4253$	0.992	0.04
m, p- Xylenes	$y = 0.9541x + 0.5844$	0.986	0.03
Styrene	$y = 0.9524x + 0.4132$	0.991	0.04
o-Xylene	$y = 0.9515x + 0.4926$	0.989	0.04
n-Nonane	$y = 0.9878x + 0.1635$	0.998	0.04
i-Propylbenzene	$y = 0.9418x + 0.5162$	0.986	0.04
n-Propylbenzene	$y = 0.9426x + 0.5468$	0.986	0.04
m-Ethyltoluene	$y = 0.9532x + 0.4838$	0.989	0.04
p-Ethyltoluene	$y = 0.9554x + 0.3953$	0.992	0.04
1,3,5-Trimethylbenzene	$y = 0.951x + 0.4724$	0.989	0.04
o-Ethyltoluene	$y = 0.9784x + 0.0956$	0.999	0.04
1,2,4-trimethylbenzene	$y = 0.9563x + 0.4509$	0.991	0.03
n-Decane	$y = 0.9651x + 0.3068$	0.995	0.04
1,2,3-trimethylbenzene	$y = 0.9537x + 0.3191$	0.993	0.04
m-Diethylbenzene	$y = 0.9541x + 0.4494$	0.991	0.04
p-Diethylbenzene	$y = 0.9607x + 0.3788$	0.993	0.04
n-Undecane	$y = 0.9519x + 0.3329$	0.992	0.04
n-Dodecane	$y = 0.9890x + 0.2711$	0.993	0.05

Reference: Zou, Y., et al. "Characteristics of 1 year of observational data of VOCs, NO<sub>x</sub> and O<sub>3</sub> at a suburban site in Guangzhou, China." Atmospheric Chemistry and Physics 15.12 (2015): 6625-6636.

11. Page 6, Line 161: The word "site" should be corrected to "sites".

**Reply:** Revised as suggested.

12. Page 7, Line 199:

a. Please clarify R (state clearly whether it is Pearson correlation coefficient or coefficients of determination) and the calculation of p-Value (from one-tail or two-tailed t-test).

**Reply:** Please see our modifications, "Pearson correlation coefficient (R) = 0.84 and p-Value (from two-tailed t-test) < 0.01"

b.  $R > 0.84$  should be " $R = 0.84$ " as stated in Fig. S1

**Reply:** Thanks again for pointing out our mistake. We corrected it to "Pearson correlation coefficient (R) = 0.84".

13. Page 7, Line 207: Please explain and elaborate how 3000 particulates were released at 100 m above sea level (a.s.l) over the site (HZ Base) and how their backward movement was tracked for 48 hours with a time resolution of 1 hour.

**Reply:** For the former question, these are the input configurations of the LPDM model, namely, we used 3000 particulates at 100m over the site. The reason we choose 100m a.s.l instead of ground level is to avoid the block effect of buildings/constructions, so that the simulated air masses could correctly represent the region's information. For the latter question, the calculation of the backward movement was using the HTSPLIT model. Generally, the calculation is a hybrid between the Lagrangian approach, using a moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location, and the Eulerian methodology, which uses a fixed three-dimensional grid as a frame of reference to compute pollutant air concentrations.

14. Page 7, Line 218: The format of the end-text citation "(Guenther et al. (2012))" was not correct. It should be changed to "(Guenther et al., 2012)".

**Reply:** Thanks. We changed it to "(Guenther et al., 2012)".

15. Page 7, Line 225: "Moderate-Resolution Imaging Spectroradiometer" should appear before its abbreviation "MODIS". Therefore, "MODIS (Moderate-Resolution Imaging Spectroradiometer)" should be corrected to "Moderate-Resolution Imaging Spectroradiometer (MODIS)".

**Reply:** Thanks. We corrected it as suggested.

16. Page 8, Line 229: “Weather Research Forecast-Community Multi Scale Air Quality” should appear before its abbreviation “WRF-CMAQ”. Therefore, “WRF-CMAQ (Weather Research Forecast-Community Multi Scale Air Quality)” should be corrected to “Weather Research Forecast-Community Multi Scale Air Quality (WRF-CMAQ)”.

**Reply:** Thanks. We corrected it as suggested.

17. Page 8, Line 233: Please explain why the meteorological fields of 2019-2021 were used as a parallel simulation of that in September 2022. Why only applied the data from previous three years but not four/five years?

**Reply:** Thanks for the interesting question. Our objective is to emphasize the extreme high temperatures of September 2022. To achieve this, we sought to compare the data for September 2022 with past averages. Yes, we did conduct a comparison using temperature data for different historical periods, namely, 2019-2021 (three years), 2018-2021 (four years), and 2017-2021 (five years). The resulting average temperatures for these periods were 31.5°C, 31.3°C, and 31.4°C, respectively. Given the relatively minor fluctuations in the temperature moving averages over the past few years, we believe that all these moving averages could highlight the exceptional high temperatures in September 2022. Considering that the use of computing resources would increase with more years considered, we opted for the meteorological data from 2019-2021. This decision does not compromise the primary goal of our study (to highlight the extreme high temperature in 2022 September).

18. Page 8, Line 241-242: In-text citation should be used in this sentence, it is suggested to be changed to “The application method was roughly in line with that in Lyu et al. (2022).”

**Reply:** Thanks. We corrected it to “The application method was roughly in line with that in Lyu et al. (2022).”

19. Page 8, Line 243: Abbreviation of “SO<sub>2</sub>” should be defined.

**Reply:** Revised as suggested.

20. Page 8, Line 248: Abbreviations of “NO” and “NO<sub>2</sub>” should be defined.

**Reply:** We defined as the following “A ‘family conservation’ that set the total NO<sub>x</sub> to the observed value every hour and allowed nitrogen monoxide (NO) and nitrogen dioxide (NO<sub>2</sub>) to evolve over time was applied.”

21. Page 8, Line 256: The word “exacted” should be replaced by “extracted”.

**Reply:** Thanks for the careful review, we corrected it to “extracted”.

22. Page 9, Line 262: Abbreviation of “NCAR” should be defined.

**Reply:** We corrected it to “National Center for Atmospheric Research (NCAR)”

23. Page 9, Line 263-264: Abbreviation of “STE” has already mentioned once in the previous content, so “STE” should be used instead of “stratosphere-to-troposphere exchange”.

**Reply:** Yes. We corrected it to “STE” here.

24. Page 9, Line 265-267: Abbreviations of “MERRA2” and “MOZART-T1” should be defined.

**Reply:** “National Center for Atmospheric Research (NCAR)” and “Model for Ozone and Related Chemical Tracers (MOZART)” were updated here.

25. Page 9, Line 273-276: Please provide supporting references or evidence for these mentioned uncertainties if any.

**Reply:** Supporting references are added here. “it was worth noting that the satellite retrievals themselves contain uncertainties, mainly from the impact of clouds, aerosols, surface albedo and the inversion algorithms (Briegleb et al., 1986; De Smedt et al., 2010; Povey and Grainger, 2015)”

Briegleb, B., Minnis, P., Ramanathan, V., and Harrison, E.: Comparison of regional clear-sky albedos inferred from satellite observations and model computations, *Journal of Applied Meteorology and Climatology*, 25, 214-226, 1986.

De Smedt, I., Stavrou, T., Müller, J. F., Van Der A, R., and Van Roozendaal, M.: Trend detection in satellite observations of formaldehyde tropospheric columns, *Geophysical Research Letters*, 37, 2010.

Povey, A. and Grainger, R.: Known and unknown unknowns: uncertainty estimation in satellite remote sensing, *Atmospheric Measurement Techniques*, 8, 4699-4718, 2015.

26. Page 9, Line 277: It was mentioned that the simulated O3 showed good agreement with the AIRS data. To enhance clarity, could you provide specific details on the extent of this agreement, such as relevant figures or quantitative measures?

**Reply:** With regard to the suggestion, we added an additional figure in the supplementary file by comparing AIRS data with CAM-Chem simulations in Eastern China (Fig. S3). The following discussion was added in the text, “It was found that the correlation coefficient in Eastern China was 0.79, passed a 95%”

significance test, indicating the CAM-Chem model relatively well produced O<sub>3</sub> at higher levels.”

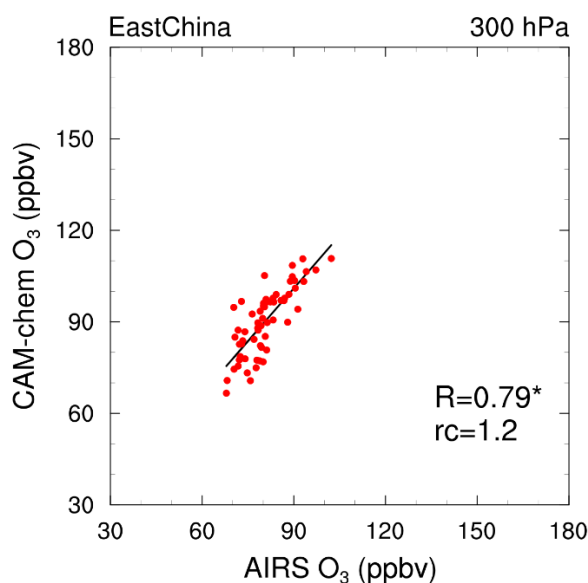


Fig. S3 Validation of AIRS O<sub>3</sub> and CAM-Chem simulated O<sub>3</sub> at 300hPa in Eastern China (R indicates correlation coefficient; rc indicates regression coefficient)

Besides, we also compared the surface O<sub>3</sub> levels between the in-situ observation and the model simulation as demonstrated in Fig. S4. The daily magnitude and variation trend were successfully captured in Guangzhou, with a mean bias error (MBE) of -7.9 ppb and a root mean square error (RMSE) of 16.3 ppb. Given the relatively well performance of CAM-Chem in both high levels and the surface level, we believe that the results could be accepted for further analyses

27. Page 9, Line 284: The word “have” should be changed to “has” if only one paper is included.

**Reply:** Revised as suggested.

28. Page 9, Line 293: The word “concentration” should be changed to “concentrations”.

**Reply:** Thanks. We changed it to “concentrations”

29. Page 12, Line 355: It was mentioned that there is a significant concentration difference in in-situ observed isoprene between day and night. Could you please provide the specific data and, if available, relevant figures to illustrate the significance of this difference?"

**Reply:** Thanks for the suggestion. During the daytime (6:00 – 17:00), the in-situ observed isoprene concentration ranged between 0.52 – 1.25 ppb, while at other



times, the average concentration was 0.1 ppbv. The description was documented in the manuscript “Besides, the in-situ observed isoprene exhibited a significant concentration difference between day and night, i.e., 0.52 – 1.25 ppbv during 6:00 – 17:00 and an average of 0.10 ppbv at other times (Fig. S6)”.

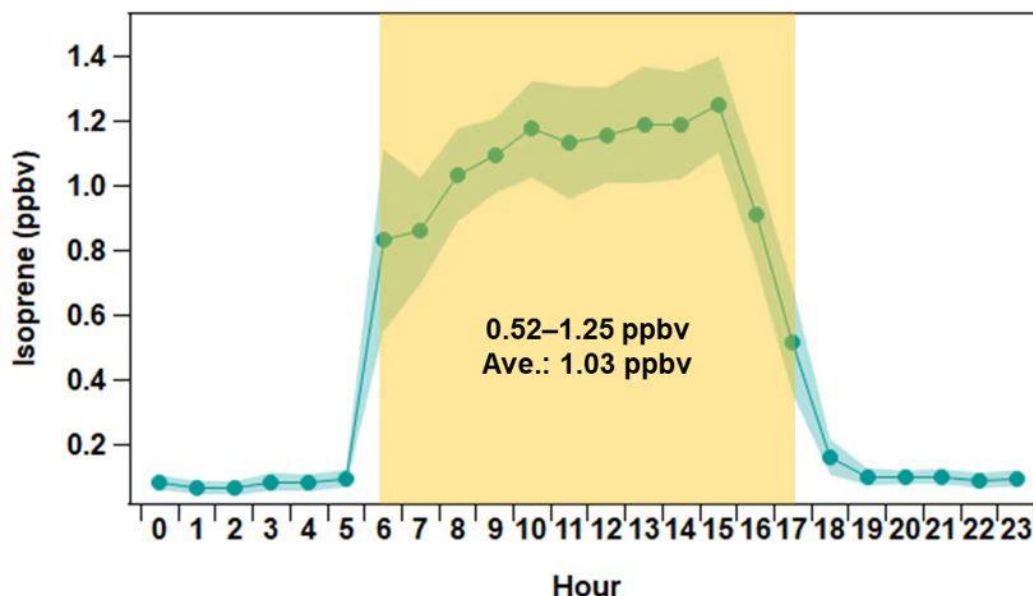


Figure S6 Diurnal variation of isoprene concentrations at HZ Base. (The yellow shaded highlights the daytime averaged concentrations, 0.51-1.25 ppbv. The daily averaged concentration was 1.03 ppbv.)

30. Page 12, Line 369: Please specify the figure you are referencing in this section.

**Reply:** Revised as suggested.

31. Page 13, Line 408: Abbreviation of “STE” has already mentioned once in the previous content, so “STE” should be used instead of “stratosphere-to-troposphere exchange”.

**Reply:** Thanks. We have made this modification as suggested.

32. Page 14, Line 427: Abbreviation of “PRD” has already mentioned once in the previous content, so “PRD” should be used instead of “Pearl River Delta”.

**Reply:** Yes, we changed it to “PRD”.

33. Page 14, Line 433: Abbreviation of “PV” has already mentioned once in the previous content, so “PV” should be used instead of “potential vorticity”.

**Reply:** Thanks. We have made this modification as suggested.

34. Page 14, Line 445: For consistency, the term “dry air” should be replaced with “specific humidity”, which was mentioned in fig. 5(c).

**Reply:** We replaced “dry air” with “low specific humidity”.

35. Page 15, Line 453: Similar patterns were consistently observed on other days between September 13-16, 2022 (Figure S7-S9), however, figures corresponding to September 14, 2022, appears to be missing in the appendix.

**Reply:** Sorry for the misunderstanding. The figure corresponding to September 14, 2022 was provided in Fig. 5 in the manuscript. We now have specified the date in the manuscript. “According to Fig. 5(a), a notable high value of PV was observed in eastern China, specifically spanning from the North China Plain (NCP) area to southern China on September 14, 2022.”. Also, the date information is also included in the caption of Fig. 5.

36. Page 15, Conclusion and implication: It appears that meteorological factors, natural emissions, chemistry pathways, and atmospheric transport each contribute to O<sub>3</sub> pollution in this study. Could you please provide numerical quantifications for the contribution of each factor?

**Reply:** Thanks for the good suggestion. Since the methods we use to study the impact of these factors on O<sub>3</sub> concentrations are founded on different principles, for example, the contribution from meteorology are based on machine learning methods, the contribution from natural emissions are based on in-situ observation constrained box model calculations, and the contributions of atmospheric transport are from a chemical transport model simulations, it is inappropriate to perform a closed calculation of the contributions quantified based on different principles. However, we tried to improve the writing by providing numerical quantifications for the contribution of each factor in the Conclusion and Implication Part. For the meteorological contribution, “we identified that meteorological factors contribute an additional 10.8 ppb to O<sub>3</sub> levels compared to the same period in previous years” (Page 15); for the impact of BVOCs, “hot weather stimulated BVOC emissions (increased by 10%)” (Page 15) and “BVOC emissions aggravated photochemical reaction and contributed nearly half of in-situ O<sub>3</sub> production” (Page 16); for the contribution by atmospheric transportation, “This process resulted in a non-negligible contributor to the surface levels in downwind area (such as the PRD, reached a maximum of ~ 8 ppb)” (Page 16).

37. Page 17, Line 517: In Figure 1(c), Please specify the time frame or date range for the distribution of 500 hPa pressure and winds.

**Reply:** The illustration has been changed to “Distribution of 500 hPa pressure and winds of September 2022.”

38. Page 17, Line 518: In Figure 1(d), Please specify the abbreviations for TEMP, UVB, PRECIP, WS, RH, and BLH for clarity.

**Reply:** Thanks for the suggestion. We have revised them to “(d) Comparisons of meteorological parameters (temperature (TEMP), precipitation (PRECIP), relative humidity (RH), ultraviolet radiation b (UVB), wind speed (WS) and boundary layer height (BLH)) between 2022 and 2019-2021”

39. Supplementary Materials, Page 5, Figure S3: Please specify the legend keys for “Obs” and “CAM-chem” for clarity.

**Reply:** We have added the illustration in the caption. “Obs refers to data from observations and CAM-Chem refers to the model simulated result.”

40. Supplementary Materials, Page 11, Table S1: Please capitalize all column names, including 'model' and 'resolution'.

**Reply:** Thanks for the suggestion, we have capitalized the column names. Please see the revised Table S1.