Response to referee comments

Response: We would like to thank the referees and editor for their interest in our work and the helpful comments and suggestions to improve our manuscript. We have carefully revised the manuscript according to all the reviewers' comments. The changes have been marked in the text using blue color. Page and line numbers in this document refers to the revised version of the main manuscript.

With these revisions, we believe the quality of the manuscript has been greatly improved. We hope the editor and the reviewers can find that all the concerns have been addressed adequately.

Reviewer(s)' Comments to Author:

Reviewer: 1

This paper by Yin et al. presents an interesting topic, i.e. the influence of stratosphere-to-troposphere transport to the surface ozone over high altitudes in the Tibetan-Himalayas region. The concept of using modelling analysis (i.e. the "Luo et al. 2019" approach) dealing with the identification of tropopause fold together with observations at multiple sites is intriguing. Also based on the available literature, I agree that there is a significant contribution from the stratosphere and that the meridional excursion of the jet stream can play a pivotal role in triggering this process. However, I think that the authors failed in support their hypothesis (i.e. the influx form the stratosphere is the main driver of the ozone variability in this region).The analysis suffers by several caveats, much of the discussion is qualitative and some tools are not well characterised or documented, indeed. Moreover, other minor issues are present along the paper. Thus, I cannot recommend publication on ACP in this current form. Below you can find a list of the major points that, in my opinion, prevent the publication of this work.

Response: Thank you so much for your valuable comments. We have carefully revised the manuscript according to your comments. In this revised manuscript, 1) ground observations of surface ozone at the sites over the Tibetan Plateau were used to reveal the difference of surface ozone variation over the Tibetan Plateau from south to north. 2) WRF-Chem and CAM-chem simulations were used to identify the impacts of long-range transport and in-situ photochemical production on surface ozone over the Tibetan Plateau. 3) Tropopause fold frequencies (calculated by ERA-5 data), position of westerly jet (U component of winds) and CAM-chem simulation was used to investigate the impact of stratospheric intrusion.

The authors used the outputs O3S from the CAM-chem model to infer an estimate of the quantity of stratospheric O3 which is contributing to the overall O3 variability. How was this specific product

validated? The authors must convince the reader that this model product is able to provide accurate and reliable quantification of the O3S/O3 ratio for the investigated region and sites. Unfortunately, I was not able to find any usable information on the cited paper by Tilmes et al. (2016). Before using it for this assessment, you should have shown how the CAM-cam model compares with observations at the considered sites (Mt. Waliguan, NCO-P, Nam-Co) and document how well the model is able to diagnose transport from the stratosphere and to quantify the contribution to the tropospheric ozone.

Response: Thank you so much for these valuable comments. In this revised manuscript, we compared CAM-cam, WRF-Chem and ground observations in revised manuscript as you suggested. We added this part in line 126-131 as: "We compared the simulated surface temperature (T2), relative humidity (RH2), wind at 10 meters (wind10), and wind at 500 hPa, with surface observations provided by China Meteorological Data Service Centre (http://data.cma.cn/) and ERA interim reanalysis datasets provided by ECMWF (https://apps.ecmwf.int/datasets/). It was found that simulation configuration captured the meteorological fields well, which is crucial to assure prediction accuracy of air pollutant concentrations (Figure S2-S5). Besides, surface ozone concentrations from ground observations, WRF-Chem, and CAM-chem were compared and showed a good fit (Figure S6)".

The period of investigation is not clear and not consistent. Section 3 discussed analyses based on 2012 and 2016 data. There is not discussion about how much this inconsistent data frame can affect the results. How much are these single years representative of a longer temporal period?

Response: In revised manuscript, we have expanded the dataset and used ground observation from 2016 to 2021 at 13 sites over the Tibetan Plateau to avoid showing accidental phenomena that only occur in a certain year. Mechanism of the surface ozone variation over the Tibetan Plateau was investigated with the ground observation data, WRF-chem results, CAM-chem results, and ERA-5 data during 2016-2017. Detail of observation and model simulation were described in revised manuscript in line 68-72: "This study synthesized ground measurements of surface ozone by analyzing the ground observation data collected at 13 sites (Shannan, Shigatse, Nyingchi, Lhasa, Changdu, Nagqu, Guoluozhou, Huangnanzhou, Hainanzhou, Haidongzhou, Xining, Haibeizhou, and Haixizhou) ranging from the southern to the central and northeastern Tibetan Plateau (Figure 1 and Table S1 in Supplementary). Surface ozone concentrations during January 2016 to December 2021 at 13 sites were provided by the China National Environmental Monitoring Center (http://106.37.208.233:20035/)" and line 103-104: "Both CAM-chem and WRF-Chem simulations were conducted for an entire year, from 1 November 2016 to 30 November 2017".

The analysis of the variability of monthly mean O3 values with monthly mean CO and NO2 values at the urban sites is too basic. Honestly, I cannot understand how this could support a dominant role of

stratospheric transport to the appearance of the yearly O3 peak. Moreover, no discussion about the robustness of the correlation analysis was provided.

Response: Thank you for your suggestion. In this revised manuscript, we used WRF-Chem results to consolidate the analysis in the impact of local photochemical generation of ozone. Results were added in line 145-154: "The local photochemical production of ozone is generated through a chain of photochemical oxidation of CO and VOCs (volatile organic compounds). The monthly variation in surface ozone at three different types of sites in the central Tibetan Plateau (Lhasa: urban site; Dangxiong: rural site; Nam Co: remote site) with varying conditions of ozone precursors were similar (Yin et al., 2017), implying the limited impact from local photochemical production of ozone on the monthly variation of surface ozone at these sites. Indicated by results in WRF-Chem simulation setup 2 (Figure 2b), local photochemical generation of ozone over the Tibetan Plateau contributed to the surface ozone in the Tibetan Plateau from 5.24% to 17.44% in each season with annual average in 10.46% (Table 1). Furthermore, the net ozone photochemical production was found to be negative in the summer when surface ozone reached a maximum in the northeastern Tibetan Plateau (Zhu et al., 2004; Ma et al., 2002). Therefore, the monthly mean surface ozone peaks in Tibetan Plateau were not mainly attributed to the local photochemical production of ozone".

The authors implicitly assumed that the variability of ozone in the region is only due to local photochemistry and input from the stratosphere. They completely neglected the role of the transport (from the regional to the large scale). As an instance, despite what the authors concluded, a notable fraction of the highest O3 values observed at NCO-P during spring (pre-monsoon) were mostly related to transport of pollution often related with wildfires. They cited the paper by Putero et al. (2014) who nicely assessed this contribution.

Response: Thank you for your suggestion. In this revised manuscript, we used WRF-Chem results to investigate the impact of transport. Results were added in line 132-137: "Generally, elevated tropospheric ozone budgets at ground sites result from three processes: in-situ photochemical production, long-range transport, and influx of ozone from the stratosphere. South Asia is one of the biggest sources of air mass that arrived the Tibetan Plateau by long-range transport (Yin et al., 2021) and the concentrations of air pollutions, including the precursors of surface ozone (e.g. CO, VOCs), in South Asia were high (Kumar et al., 2018). Indicated by results in WRF-Chem simulation setup 1 (Figure 2a), long-range transport from South Asia contributed to the surface ozone in the Tibetan Plateau from 9.45% to 20.28% in each season with annual average in 14.13% (Table 1), showing a limited impact".

The quality of the figures is really poor. In some cases (e.g. Figure 1), it was not possible to understand labels and numbers: this prevents a serious evaluation of their scientific content.

Response: Thank you for your suggestion. We updated all figures to high quality.

I don't think that the use of data from external data originator was well recognised/acknowledged. As an instance the paper by Putero et al. (2014) did not report any usable dataset for NCO-P. How the data were obtained? Why the authors did not take the data from traceable data repository like https://ebas-data.nilu.no/ ? I suppose that the same can apply for Nam-Co and Mt. Waliguan.

Response: Thank you for your suggestion. In this revised manuscript, we used surface ozone ground observation data during January 2016 to December 2021 at 13 sites provided by the China National Environmental Monitoring Center (http://106.37.208.233:20035/). Data from NCO-P, Nam Co, and Mt. Waliguan were removed.

Minor points:

Line 43-46: In the Introduction, more recent bibliographic references should be provided besides than Crutzen (1988). You can consider the valuable papers produced within the TOAR initiative. Bracci et al. (2012) is specifically devoted to the analysis of synoptic-scale processes leading to stratospheric intrusions over the southern Himalayas: it should be better contextualised. In the global troposphere, ozone burden is not only affected by photochemical production and STE but also chemical loss and dry deposition. When discussing ozone variability over specific regions or sites other processes must be considered (among the others PBL dynamic, air-mass transport occurring at different scales).

Response: Thank you for your suggestion. References were updated to recent publications in revised manuscript in line 43-46: "Tropospheric surface ozone (O_3) has attracted great attention during the last decades due to its impact on the oxidative capacity of the troposphere, the radiative forcing of the atmosphere, the detrimental effects on the vegetation and ecosystem, and human health (Lu et al., 2018). The tropospheric (surface) ozone is controlled by in situ photochemical production in the troposphere and by the intrusion of ozone-laden stratospheric air (Kumar et al., 2016; Yang et al., 2022)".

Line 61: here you should also mention the long-range transport as well as the transport of polluted air-masses from the neighbour regions.

Response: Thank you for your comment. In this revised manuscript, we added the WRF-Chem simulation results to investigate the impact of long-range transport. It was found that transport of ozone from the major air mass source region (South Asia) as well as the local photochemical production of ozone could contribute to limited variation of surface ozone over the Tibetan Plateau.

Line 67: please introduce the background sites. You should give to the readers, at least, some basic descriptions of the sites in terms of geographical locations and already documented processes that can affect ozone.

Response: Thank you for your comment. In revised manuscript, we removed observations from background sites and used 13 sites surface ozone results provided by the China National Environmental Monitoring Center. Information of these sites were added in line 68-72: "This study synthesized ground measurements of surface ozone by analyzing the ground observation data collected at 13 sites (Shannan, Shigatse, Nyingchi, Lhasa, Changdu, Nagqu, Guoluozhou, Huangnanzhou, Hainanzhou, Haidongzhou, Xining, Haibeizhou, and Haixizhou) ranging from the southern to the central and northeastern Tibetan Plateau (Figure 1 and Table S1 in Supplementary). Surface ozone concentrations during January 2016 to December 2021 at 13 sites were provided by the China National Environmental Monitoring Center (http://106.37.208.233:20035/)".

Line 71: the description of the measurement methodology was unsatisfactory. No information are provided about uncertainties related to the measurements, data coverage, reference to metrological standards. Why two different year are considered 2012 and 2016?

Response: In revised manuscript, ground observation of surface ozone data during 2016 to 2021 were used. Both CAM-chem and WRF-Chem simulations were conducted for an entire year, from 1 November 2016 to 30 November 2017. Measurement methodology of surface ozone was revised in line 71-80: "Surface ozone concentrations during January 2016 to December 2021 at 13 sites were provided by the China National Environmental Monitoring Center (http://106.37.208.233:20035/). Surface ozone was measured with the UV-spectrophotometry method. The specifications and test procedures of ozone followed the Specifications and Test Procedures for Ambient Air Quality Continuous Automated Monitoring System for ozone (National Environmental Protection Standards of the People's Republic of China, HJ 654-2013). The inlet of the instrument was 3-20 m above the ground surface, 1 m higher than the roof of the building or the wall. The observation stations were located at least 50 m from any obvious stationary pollution sources. The data quality assurance and controls followed technical guidelines on environmental monitoring quality management (National Environmental Protection Standards of the People's Republic of China, HJ 630-2011), and the data were checked for validity based on ambient air quality standards (National Standards of the People's Republic of China, GB 3095-2012)".

Line 83: 2106 should be 2016. Data for NCO-P cannot be obtained by the web site that you indicated.

Response: In revised manuscript, this part was totally revised and data from NCO-P were removed.

Line 92: the identification methodology should be described.

Response: Thank you for your suggestion. Identification methodology was described in line 84-87: "Monthly mean total tropopause fold frequency was investigated in this study and tropopause fold was defined by 2 pvu (potential vorticity unit) iso-surface and identified by the method described in Luo et al. (2019). Tropopause folds were defined as multiple crossings of the dynamical tropopause in a vertical profile".

Line 101: According with Tilmes et al. (2016) the O3S product is not affected by dry deposition in CAM-chem. How much this can create bias on your estimate if compared to the real world? The ability of the model to reproduce the O3 variability at the considered sites must be demonstrated before using it (e.g. no comparison with the real observations are provided and discussed).

Response: Thank you for your suggestion. Surface ozone concentrations from ground observations, WRF-Chem, and CAM-chem were compared in revised manuscript and showed a good fit. For O3S product, we have limited evidence to estimate the bias in CAM-chem and it is indeed a question worthy of further investigation. We hope to have reliable means to evaluate it in the future.

Figure 2. Please express PV in pvu and Ozone in ppb. The scale of the plate (a) should be increased to values higher than 10%. It looks that the 2 pvu surface is well detached from the ground. I agree that there are signals of stratospheric transport occurring (the relatively high PV – but well lower than 2 pvu -stretching down from the stratospheric "reservoir"), but it's difficult to use this plot to support a dominant role of stratospheric transport for surface ozone variability. Do you have a similar cross section for the tropopause fold frequency?

Response: Thank you for your suggestion. In revised manuscript, we have almost re-conducted the all the simulation for both WRF-Chem and CAM-chem and Figure 2 was revised into Figure 3 and Figure 4 in revised manuscript as below:

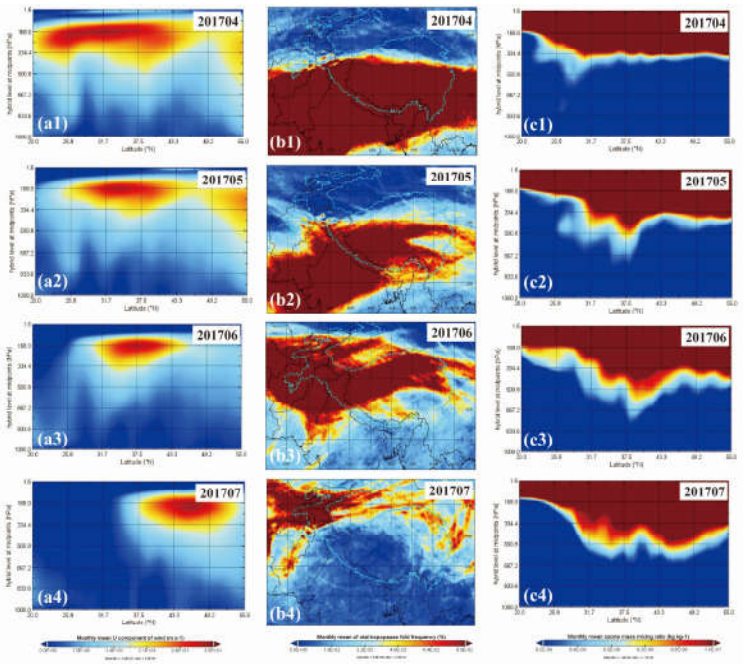

Figure 3. Monthly mean meridional cross-section of ERA-5 data at 91°E (~ the central Tibetan Plateau) showing the zonal winds c) and ozone (d) in April, May, June, and July in 2017. Monthly average of total tropopause fold frequencies (b) in April, May, June, and July in 2017.

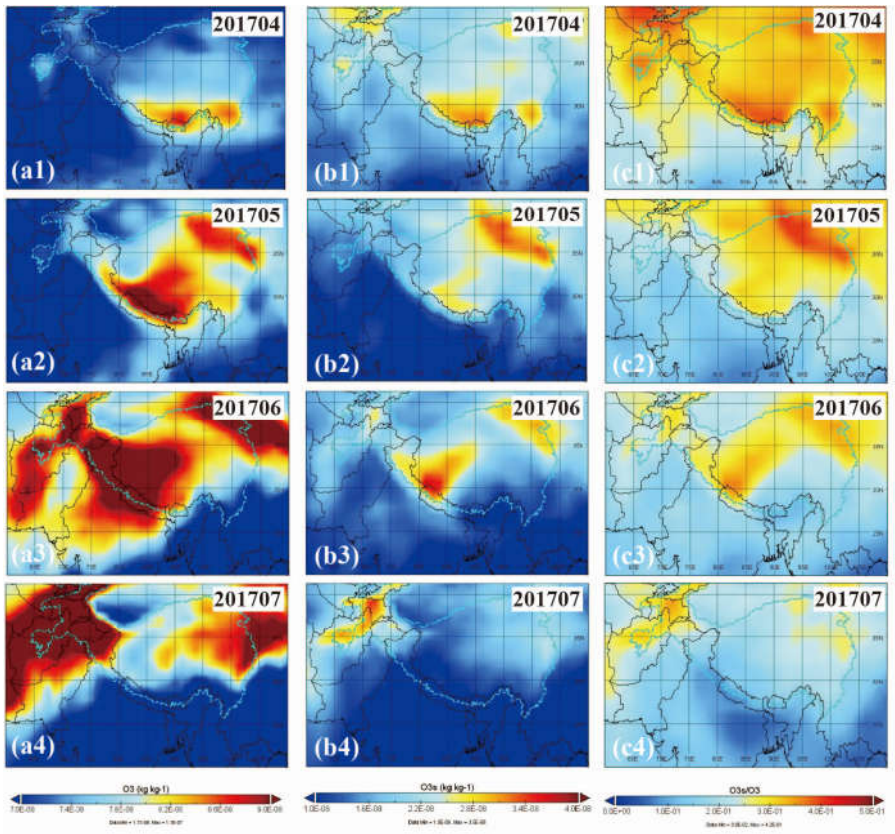

Figure 4. Monthly average of (a) ozone, (b) O_3S, and (c) O_3S/O_3 at 581.25 hPa in April, May, June, and July in 2017. The location of the Tibetan Plateau is shown with cyan contour in the maps.

Figure S4 did not report the measurement unit for O3S.

Response: The measurement for O3S is kg kg^{-1} (10^9 ppb). Figure S4 was removed in revised manuscript.

Reviewer: 2

The manuscript by Yin et al. presented an interesting topic related to the influence of stratospheric ozone input on tropospheric surface ozone variability. Through analyses of monthly surface ozone variability, modeled stratospheric O3 tracer (unjustified), and weather structure of the upper troposphere/lower stratosphere, the authors concluded that stratospheric ozone contributed ~ 65 % of surface ozone across the high mountain Asia. Although in general I agree that stratospheric ozone inputs may be important for surface ozone budget at midlatitudes & high mountain regions, ~ 65% contribution is too overwhelmed, and which is inadequately supported by evidence and assessments presented in the manuscript. I think there are sufficiently more analyses that the authors need to conduct to support the conclusion, and to promote the manuscript to be potentially publishable.

Response: Thank you so much for your valuable comments. We have carefully revised the manuscript and include more analyses and evidences to reinforce the conclusion. In revised manuscript, 1) ground observations of surface ozone at the sites over the Tibetan Plateau were used to reveal the difference of surface ozone variation over the Tibetan Plateau from south to north. 2) WRF-Chem and CAM-chem simulations were used to identify the impacts of long-range transport and in-situ photochemical production on surface ozone over the Tibetan Plateau. 3) Tropopause fold frequencies (calculated by ERA-5 data), position of westerly jet (U component of winds) and CAM-chem simulation was used to investigate the impact of stratospheric intrusion. We have updated the dataset and almost re-conducted the all the simulation for both WRF-Chem and CAM-chem and the contribution from stratospheric ozone intrusion was re-calculated.

Major comments:

1. sections 3.2, the rejection of the role of in-situ chemistry on surface ozone variations is a rush. Simply analyzing the seasonal patterns of O3 with NO2 and CO cannot rule out the role of in-situ chemistry, especially the author didn't explain why such correlated or uncorrelated data can reveal their causal

relationships. In general, NO2 and VOCs, as well as CO are precursors of O3, but sunlight (photochemistry) is also necessary. In the plateau, soil emissions would be important sources of NOx and VOCs, which in general depends on temperature. How this precursors and local actinic flux varied with O3? And how was transport affects surface O3? All these need to be assessed. Meanwhile, don't forget CO is also an indicator of stratospheric air incursion as which is characterized as low CO and high O3.

Response: Thank you for your suggestion. In revised manuscript, we used Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) results to investigate the impacts of local photochemical generation of ozone and long-range transport. WRF-Chem was conducted by the setup adopted in Yang et al. (2022) to investigate the long-range transport from South Asia and local photochemical generation of ozone over the Tibetan Plateau. Experiments with 2 setups were conducted as setup 1 (CO and NO_2 in South Asia were set to 0 to investigate the influence of long-range transport from South Asia) and setup 2 (CO and NO_2 in the Tibetan Plateau were set to 0 to investigate the influence of local photochemical generation of ozone over the Tibetan Plateau).

For in-situ chemistry, we revised the manuscript in line 145-154 as: "The local photochemical production of ozone is generated through a chain of photochemical oxidation of CO and VOCs (volatile organic compounds). The monthly variation in surface ozone at three different types of sites in the central Tibetan Plateau (Lhasa: urban site; Dangxiong: rural site; Nam Co: remote site) with varying conditions of ozone precursors were similar (Yin et al., 2017), implying the limited impact from local photochemical production of ozone on the monthly variation of surface ozone at these sites. Indicated by results in WRF-Chem simulation setup 2 (Figure 2b), local photochemical generation of ozone over the Tibetan Plateau contributed to the surface ozone in the Tibetan Plateau from 5.24% to 17.44% in each season with annual average in 10.46% (Table 1). Furthermore, the net ozone photochemical production was found to be negative in the summer when surface ozone reached a maximum in the northeastern Tibetan Plateau (Zhu et al., 2004; Ma et al., 2002). Therefore, the monthly mean surface ozone peaks in Tibetan Plateau were not mainly attributed to the local photochemical production of ozone".

2. Section 3.3, it is entirely unclear how the contribution of stratospheric O3 to surface O3 was estimated, by the ratio of stratospheric O3 tracer (O3s) to the **modeled** or the **observed** surface O3? How was the modeled surface O3 vs. the observed surface O3 in the model?

Response: O3s is a stratospheric ozone tracer that represents the amount of ozone in the troposphere originated in the stratosphere in CAM-chem simulation. We used the ratio of O3s and O3 at site to indicate the influence from stratospheric ozone on surface ozone.

In revised manuscript, modeled surface O3 and the observed surface O3 as well as meteorological fields were compared as described in line 126-131: "We compared the simulated surface temperature (T2),

relative humidity (RH2), wind at 10 meters (wind10), and wind at 500 hPa, with surface observations provided by China Meteorological Data Service Centre (http://data.cma.cn/) and ERA interim reanalysis datasets provided by ECMWF (https://apps.ecmwf.int/datasets/). It was found that simulation configuration captured the meteorological fields well, which is crucial to assure prediction accuracy of air pollutant concentrations (Figure S2-S5). Besides, surface ozone concentrations from ground observations, WRF-Chem, and CAM-chem were compared and showed a good fit (Figure S6)".

3. section 3.4, the CAM model is partially driven meteorological parameters, where the interactions between the troposphere and the stratosphere are determined by jet stream and/or tropopause folding. Thus it is kind of a loop to compare the modeled O3s with such synoptic events as it is that these events transport air out and in the stratosphere (and vice versa) at the boundary of stratosphere and troposphere. This should put before or right after analyses on the effects of in-situ chemistry, but before the model analysis. The formers can be qualitative, but the latter should be quantitatively conducted. In fact, why not used the CAM model to also assess contributions of tropospheric processes to the observed surface O3? In general, most atmospheric chemistry model (online or offline coupled) has a better performance on tropospheric chemistry simulations than stratospheric chemistry simulations. Why don't show the modeled surface O3 concentrations as well as the fractions of tropospheric contributions by simply deducting O3s from total model surface O3.Without such comparisons, both section 3.2 and 3.3 are incomplete and are insufficient to support the conclusion.

Response: Thank you for your suggestion. As you suggested, section 3.4 (the qualitative evidences from reanalysis data) were placed to section 3.3 before quantitative WRF-chem simulation part (as in revised section 3.4).

Tropospheric contribution was investigated based on WRF-Chem simulation results in revised manuscript in line 145-154 as: "The local photochemical production of ozone is generated through a chain of photochemical oxidation of CO and VOCs (volatile organic compounds). The monthly variation in surface ozone at three different types of sites in the central Tibetan Plateau (Lhasa: urban site; Dangxiong: rural site; Nam Co: remote site) with varying conditions of ozone precursors were similar (Yin et al., 2017), implying the limited impact from local photochemical production of ozone on the monthly variation of surface ozone at these sites. Indicated by results in WRF-Chem simulation setup 2 (Figure 2b), local photochemical generation of ozone over the Tibetan Plateau contributed to the surface ozone in the Tibetan Plateau from 5.24% to 17.44% in each season with annual average in 10.46% (Table 1). Furthermore, the net ozone photochemical production was found to be negative in the summer when surface ozone reached a maximum in the northeastern Tibetan Plateau (Zhu et al., 2004; Ma et al., 2002). Therefore, the monthly mean surface ozone peaks in Tibetan Plateau were not mainly attributed to the local photochemical production of ozone".

In addition, I suggest the authors to move SI materials to the main text, overall, the length of the main text is short and lacks of sufficient details in results and discussions, and putting related materials in SI makes further difficulties in understanding the arguments/assessments the authors stated.

Response: Thank you for your suggestion. Manuscript was revised and we add WRF-Chem and CAM-chem simulations to identify the impacts of long-range transport and in-situ photochemical production on surface ozone over the Tibetan Plateau to deliver more results and discussions.