Surface ozone over the Tibetan Plateau controlled by stratospheric intrusion

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Abstract:

Tibetan Plateau is a global hotspot of stratospheric intrusion, and elevated surface ozone were observed at ground monitoring sites. Still, links between the variability of surface ozone and stratospheric intrusion at regional scale remain unclear. This study synthesized ground measurements of surface ozone over the Tibetan Plateau and analyzed their seasonal variations. The monthly mean surface ozone concentrations over the Tibetan Plateau peaked earlier in the south in April/May and later in the north in June/July. The migration of monthly surface ozone peaks was coupled with the synchronous movement of tropopause fold and westerly jet that created a conducive conditions for stratospheric ozone intrusion. Stratospheric ozone intrusion significantly contributed to surface ozone across the Tibetan Plateau, especially for the areas with high surface ozone concentrations during their peak-value month. We demonstrated that monthly variation of surface ozone over the Tibetan Plateau is mainly controlled by stratospheric intrusion, which warrants a proper consideration in understanding atmospheric chemistry and impacts of ozone over this highland region and beyond.

1. Introduction.

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). In contrast with other air pollutants, tropospheric ozone is significantly influenced by stratospheric intrusion, especially at high elevation (Bracci et al., 2012; Cristofanelli et al., 2010; Yin et al., 2017).

Tibetan Plateau is a vast plateau with an average elevation of ca. 4,500 m above sea level, making it the highest plateau in the world. It is thus often referred to as “the roof of the world” or “the third pole” (Yao et al., 2012). With limited human accessibility and low levels of industrialization, Tibetan Plateau is one of the most pristine regions in the world in terms of most of the air pollutants (Yin et al., 2017) and is regarded as an ideal area for studying the atmospheric environment in the free troposphere. It can provide deeper insights into changing background concentrations of atmospheric pollutants (Kang et al., 2019). The Tibetan Plateau is recognized as a global hotspot of deep stratospheric intrusion, and elevated surface ozone has been observed at some ground monitoring sites and very high concentrations of surface O_3 were reported (Cristofanelli et al., 2010; Yin et al., 2017; Zhu et al., 2004). At sites, it was reported that vertical ozone flux from vertical transport by convective and synoptic transport was prominent in the summer at Waliguan leading to high ozone levels (Ma et al., 2002; Ma et al., 2005). Studies at Shangri-La and Nam Co in Tibetan Plateau indicated that the input of ozone from the stratosphere had potential impacts on the seasonal variations of surface ozone at each site (Yin et al., 2017; Ma et al., 2014). Still, the link

between the variability of surface ozone and stratospheric intrusions at the regional scale remains unclear. In this study, 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) Distribution of 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 on surface ozone variation.

2. Materials and Methods.

2.1. Surface measurements.

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/). 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).

2.2. Reanalysis data.

The ECMWF (European Centre for Medium-Range Weather Forecasts) ERA-5 data (0.28125° latitude × 0.28125° longitude horizontal resolution, and 137 vertical levels) (Hoffmann et al., 2019) were used to analyze the monthly mean meridional cross-section structures of troposphere and stratosphere over the Tibetan Plateau. 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.

2.3. Model simulation.

The Community Atmosphere Model with Chemistry (CAM-chem), a global model, was used for simulations of global

tropospheric and stratospheric atmospheric composition (Lamarque et al., 2012) provided by University Corporation for Atmospheric Research (https://www.acom.ucar.edu/cam-chem/cam-chem.shtml). The horizontal resolution for CAM-chem is 0.5° × 0.5°. CAM-chem uses the MOZART (Model for OZone And Related chemical Tracers) chemical mechanisms (Lamarque et al., 2012), with various choices of complexity for tropospheric and stratospheric chemistry. The O_{3S} variable, a stratospheric ozone tracer that represents the amount of ozone in the troposphere originated in the stratosphere, in CAM-chem simulation was used in this study to indicate the intensity of the ozone transported from the stratosphere (Tilmes et al., 2016). O3S/O3 ratio was calculated to investigate the intensity of impacts from the stratospheric intrusion to ozone concentration in troposphere.

Weather Research and Forecasting model coupled with Chemistry (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. The model domain has 25-km grid spacing and covers South Asia and the TP, with 155 and 135 grid cells in the west–east and north–south directions, respectively. The vertical configuration of the model includes 35 vertical sigma layers at the top pressure of 50 hPa. National Centers for Environmental Prediction Final (NCEP FNL) analysis data with a horizontal resolution of 1° × 1° at 6-h time intervals is used for the meteorological initial and boundary conditions. Both CAM-chem and WRF-Chem simulations were conducted for an entire year, from 1 November 2016 to 30 November 2017. The first month was considered the model spin-up time and was not analyzed. In WRF-Chem simulation, 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).

3. Results and Discussion.

3.1. Surface ozone variations over the Tibetan Plateau.

The mean surface ozone concentrations during 2016-2021 at 13 sites over Tibetan Plateau peaked in different months and moved gradually from April/May in the south to June/July in the north (Figure 1). A similar result was obtained in the month-to-month variation of mean ozone concentrations at 13 sites (Figure S1 in Supplementary). It is noteworthy that such regular migration of monthly ozone peaks occurred at large geographical scale, and surface ozone concentrations peaked synchronously at sites in the same zone despite the differences in size of cities and baseline levels of surface ozone. Previous studies also found that the sites in adjacent areas showed similar monthly variation of surface ozone (Ma et al., 2014; Lin et al., 2015; Ran et al., 2014; Cristofanelli et al., 2010; Zhu et al., 2004; Yin et al., 2017). The occurrence of the highest monthly concentrations of surface ozone is different in different areas over the Tibetan Plateau, ranging from April to July.

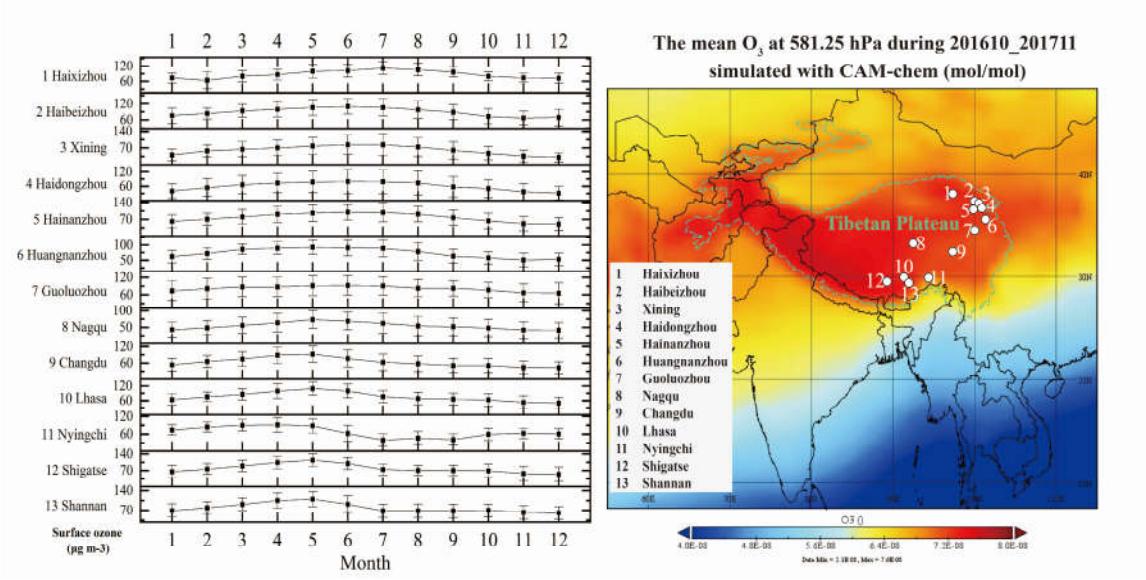

Figure 1. Study sites (right) and their monthly mean surface ozone variations during January 2016 to December 2021 across the Tibetan Plateau (left) used in this study. Map of yearly mean concentration of surface ozone during January 2016 to December 2021 at 581.25 hPa simulated with CAM-chem is shown with colored shadings.

3.2. Long-range transport and photochemical production are not likely the dominator of surface ozone.

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).

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.

Table 1. The influence ratios of long-range transport (WRF-Chem set 1) and in-situ photochemical production (WRF-Chem set 2) on surface ozone over the Tibetan Plateau.

	Annual	Spring	Summer	Autumn	Winter

WRF-Chem set 1	14.13%	13.18%	20.28%	14.52%	9.45%
WRF-Chem set 2	10.46%	8.43%	17.44%	11.95%	5.24%

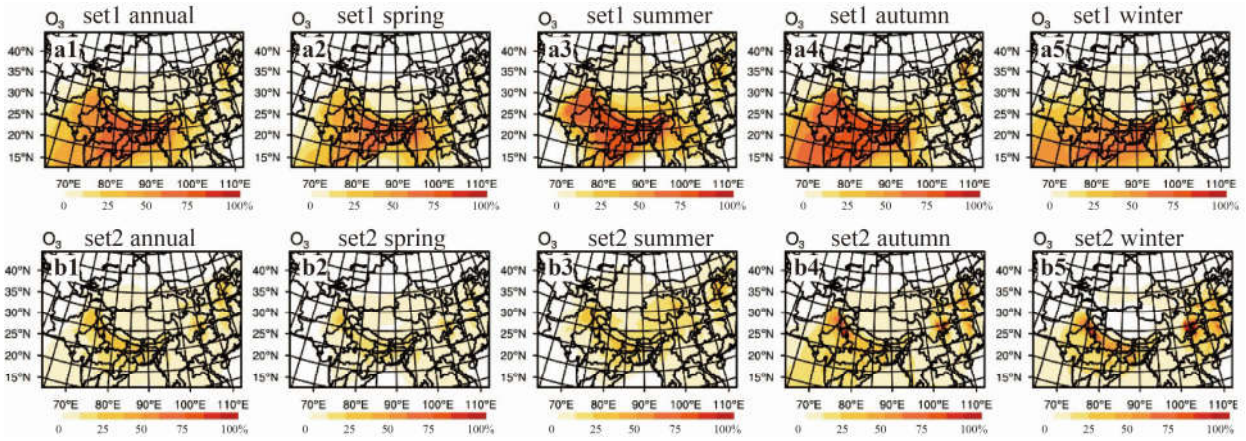

Figure 2. The influence ratios of long-range transport (WRF-Chem set 1) and in-situ photochemical production (WRF-Chem set 2) on surface ozone over the Tibetan Plateau.

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.

Even in the summer, when the contributions from these two factors are high, the total contribution from both long-range transport from South Asia and local photochemical generation of ozone over the Tibetan Plateau to the variation of surface ozone on the Tibetan Plateau was less than 38% (total contribution =37.72%). Based on the above results, we can infer that the variation of surface ozone over the Tibetan Plateau is dominated by other factors than long-range transport and in-situ photochemical production.

3.3. Evidences from reanalysis data and simulation for stratospheric intrusion.

Rather than long-range transport and photochemical production, the stratospheric influx of ozone is likely a main contributor to the surface ozone over the Tibetan Plateau. The impact of stratospheric intrusion on the surface ozone can be supported by the monthly averages of the total tropopause fold frequencies (tropopause fold was indicated by 2 potential

vorticity unit iso-surface, in Luo et al., (2019)) over the Tibetan Plateau (Figure 3). It was found that the displacement of the tropopause fold in the Tibetan Plateau was controlled by the movement of the westerly jet (Luo et al., 2019). The monthly mean meridional cross-section of westerly jet (indicated by the maximum speed of the U component of winds), tropopause fold frequencies, and the transport of ozone from the stratosphere to the troposphere was calculated and visualized from April to July 2017 (Figure 3).

In April, the westerly jet was mainly above the region within 25°N-40°N (Figure 3). The highest frequencies of total tropopause fold were mainly distributed in ca. 25-35°N latitude band covering parts of South Asia, the southern and the central Tibetan Plateau (Figure 3). The most hotspot of stratospheric ozone intrusion to troposphere was in ~29°N (Figure 3), similar with the area with high ozone concentration in April (Figure 4). In May, the westerly jet was northward moved to be center with ~ 35°N. The stratospheric ozone intrusion to troposphere was strong in ~37.5°N and ~30°N similar with the areas with high frequencies of total tropopause fold (Figure 3), contributing to high ozone concentrations in these areas (Figure 3). In June, the westerly jet was moved northward and hotspot of stratospheric ozone intrusion to troposphere was in ~38°N and ~32°N (Figure 3) and these two areas were in high ozone concentrations (Figure 4). In July, the westerly jet was even more northward moved and only the northeastern Tibetan Plateau was under significant impact of stratospheric ozone intrusion within 31.7°N-40°N (Figure 3) and induced high ozone concentration in the northeastern Tibetan Plateau (Figure 4). Consistent with the monthly spatial distribution of the westerly jet and tropopause fold occurrence, stratospheric ozone intrusion showed a northward movement from April to July.

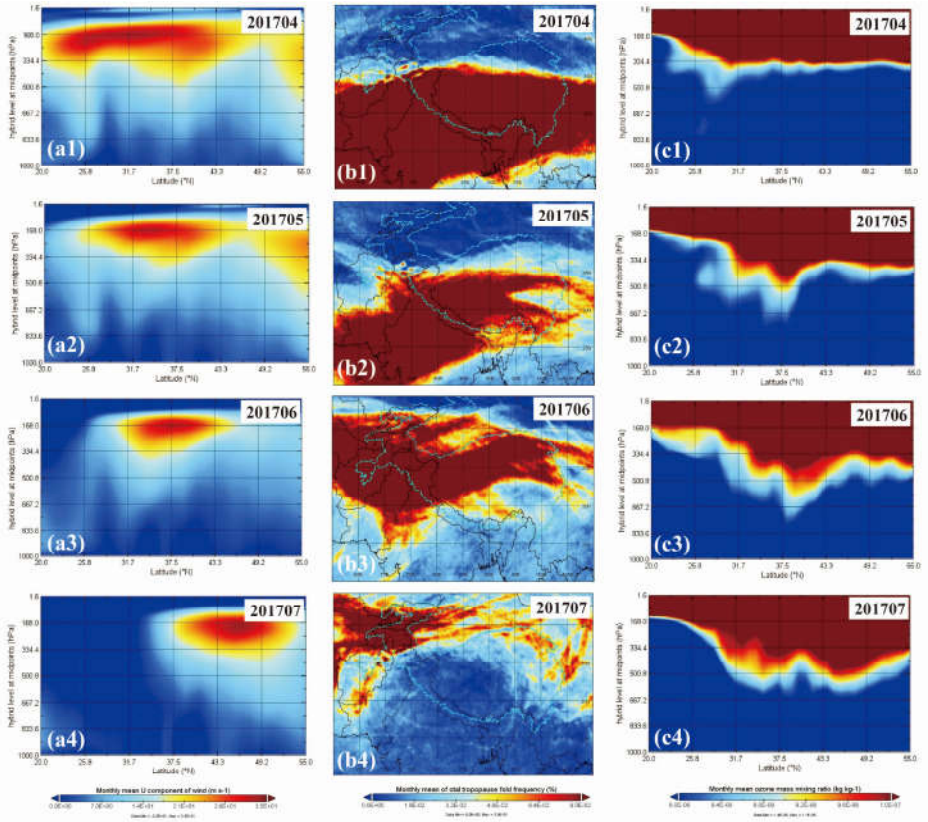

Figure 3. Monthly mean meridional cross-section of ERA-5 data at 91°E (~ the central Tibetan Plateau) showing the zonal winds c) and ozone (d) in April, May, June, and July in 2017. Monthly average of total tropopause fold frequencies (b) in April, May, June, and July in 2017.

3.4. Impact of stratospheric intrusion evidenced by stratospheric ozone tracer.

In the southern Tibetan Plateau, it was stated that the surface ozone mixing ratios were significantly increased by the stratospheric intrusions (+13 ppb during stratospheric intrusions) (Cristofanelli et al., 2010). The previous studies stated that the peak of surface ozone in June at Waliguan in the northeastern Tibetan Plateau was due to the downward transport of ozone from the upper troposphere and lower stratosphere (Ma et al., 2005; Ding and Wang, 2006). This is evidenced by analysis of a stratospheric ozone tracer of O_{3S} (a stratospheric ozone tracer that represents the amount of ozone in the troposphere originated in the stratosphere in CAM-chem) across the south-north transection of the Tibetan Plateau. Figure 4 (a1-a4) showed the surface ozone variation from April 2017 to July 2017 over the Tibetan Plateau in CAM-chem simulation. In April 2017, the hotspot area of high ozone concentration mainly distributed in the southern Tibetan Plateau which was consistent with the distribution of high O_{3S} concentration (Figure 4a). The average of O_{3S}/O_3 representing indicator of the stratospheric ozone contribution, demonstrated that 36.5% of surface ozone in the hotspot of the southern Tibetan Plateau (85°E-92.5°E and 27.8°N-30.6°N) was contributed by stratospheric ozone. In May and June 2017, the southern Tibetan Plateau was still in high ozone concentration and part of the northeastern Tibetan Plateau became a new hotspot with high ozone concentration and O_{3S} in the northeastern Tibetan Plateau was in high concentration (Figure 4b). The average of O_{3S}/O_3 was 35.9% in May and 29.5% in June in the northeastern Tibetan Plateau (the region within 90°E-100°E and 34.4°N-39.1°N). The average of O_{3S}/O_3 was higher than 25% in the southern Tibetan Plateau (85°E-90°E and 27.8°N-30.6°N) in May and in the southwestern Tibetan Plateau (80°E-85°E and 27.8°N-32.5°N). In July 2017, only the northeastern Tibetan Plateau was in high ozone concentration and O_{3S}/O_3 was higher than 25% in this region. It can be stated that stratospheric ozone intrusion could elevate at least 25% of surface ozone over the Tibetan Plateau at hotspot regions during April to July.

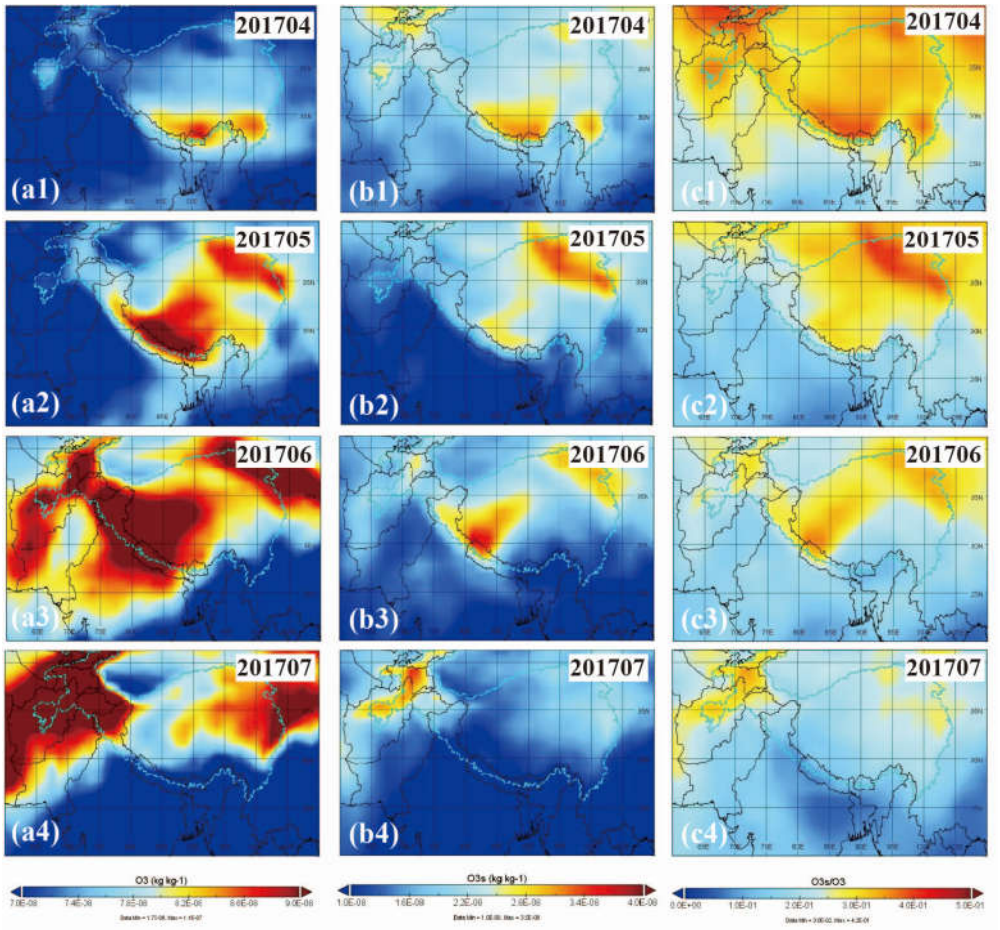

Figure 4. Monthly average of (a) ozone, (b) O_3s, and (c) O_3s/O_3 at 581.25 hPa in April, May, June, and July in 2017. The location of the Tibetan Plateau is shown with cyan contour in the maps.

4. Conclusions.

The ground measurements of surface ozone at 13 sites spread across the Tibetan Plateau were used for the first time in this study, as opposed to previous studies which focused mostly on individual sites. Simulation results from WRF-Chem, CAM-chem and analysis of ERA-5 reanalysis data were used to investigate the influences of long-range transport and in-situ photochemical production, and downward influx of ozone from the stratosphere on the variation in surface ozone. Results showed that the peak of monthly mean surface ozone concentration occurs first over the southern and the central Tibetan Plateau in April/May, and then moves progressively northwards in June/July. Such peak shift is concurrent with northward movement of the tropopause folds and westerly jet, which act as the two main control mechanisms. The position of tropopause folds was controlled by the position of the westerly jet and created the conditions conducive for the influx of ozone rich air from the deep stratosphere to the troposphere. The migration of the folds led to a similar migration from south to north of stratospheric intrusions. This lead to a significant contribution of stratospheric intrusions to the surface ozone, elevating the surface ozone concentrations. Long-range transport and in-situ photochemical production of ozone can lead to limited variation

of surface ozone over the Tibetan Plateau. We suggest that the transport of stratospheric ozone to the surface is a regionally coherent important contributor to surface ozone across the Tibetan Plateau, which needs a proper consideration in understanding the atmospheric chemistry and the impacts of ozone over this highland region and beyond.

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Data Availability Statement: Surface ozone data at 13 sites are available through the China National Environmental Monitoring Center (http://106.37.208.233:20035/). ERA-5 data are available through the European Centre for Medium-Range Weather Forecasts (https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels?tab=form, after login). CAM-chem data are available through University Corporation for Atmospheric Research (https://www.acom.ucar.edu/cam-chem/cam-chem.shtml).

Author contributions: Xiufeng Yin: Conceptualization, visualization, writing - original draft. **Benjamin de Foy, Dipesh Rupakheti, Zhenming Ji, Zhiyuan Cong, Guoshuai Zhang, and Maheswar Rupakheti:** Writing - review & editing. **Junhua Yang and Ping Li:** Visualization. **Yuling Hu and Jiali Luo:** Visualization, writing – review. **Shichang Kang and Qianggong Zhang:** Conceptualization, writing - review & editing.

Competing interests

The authors declare no competing interests.

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