

Influence of stratosphere-to-troposphere exchange on the seasonal cycle of surface ozone at Mount Waliguan in western China

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[1] Surface ozone over the Qinghai-Tibetan Plateau exhibits a summertime maximum contrasting to a spring peak observed at many remote locations in the Northern Hemisphere. In this study we conducted meteorological simulations for April–August 2003, when intensive measurement of trace gases was carried out at remote Mount Waliguan, to elucidate the influence of atmospheric dynamics on the observed ozone events and seasonal cycle. Examinations of potential vorticity and trace gases suggest that most of the synoptic-scale ozone enhancements were due to stratospheric intrusions, as opposed to transport of anthropogenic pollutions. Case studies show that the existence of jet stream is the main dynamical cause for these intrusions. Further analysis of upper-level mean zonal winds suggests that stronger subtropical jet streams lead to more frequent stratospheric intrusions in summer compared to spring, which may have contributed to the higher summertime surface ozone there and possibly also in other regions of Central Asia. **Citation:** Ding, A., and T. Wang (2006), Influence of stratosphere-to-troposphere exchange on the seasonal cycle of surface ozone at Mount Waliguan in western China, *Geophys. Res. Lett.*, *33*, L03803, doi:10.1029/2005GL024760.

1. Introduction

[2] Tropospheric ozone (O_3) has two sources, i.e., photochemical production within the troposphere and downward transport from the stratosphere [Danielsen, 1968; Logan, 1985; Oltmans and Levy II, 1992]. The relative importance of these two sources can vary regionally and seasonally [Mauzerall et al., 1996], and their conjunct effects control the seasonal cycle of tropospheric O_3 . In the remote locations of the Northern Hemisphere (NH), a spring maximum of surface O_3 has been widely observed [Logan, 1985; Monks, 2000, and references therein]. Despite extensive research on the relative role of photochemistry versus stratospheric transport, there appears no consensus as to the dominant cause of this springtime peak [Monks, 2000]. In comparison, many rural areas in the industrialized regions have observed maximum in early summer [e.g., Logan, 1989; Wang et al., 2001], which is generally attributed to strong photochemical production involving anthropogenic and biogenic emissions.

[3] In contrast to most of the NH remote areas, a distinctly different seasonal pattern of surface O_3 has been observed at Mount Waliguan (WLG) (36.28°N, 100.90°E, 3816 m asl), a WMO's GAW Baseline Observatory, which

indicated a summertime maximum [Tang et al., 1995; Ma et al., 2005]. Analysis of measurement of O_3 together with tracers of the troposphere and stratosphere such as water vapor, CO, non-methane hydrocarbons (NMHCs) [Wang et al., 2006] and Be^7 [Lee et al., 2004; Zheng et al., 2005] suggests a stratospheric source for the summertime O_3 maximum at WLG. A chemical transport model study by Zhu et al. [2004], however, concluded that the summer O_3 peak at WLG was resulted from the transport of anthropogenic pollution from eastern/central China and South Asia. Ma et al. [2005] pointed out the importance of convective processes in transporting upper-tropospheric/stratospheric air to the surface in summer. In the present study, we carried out meteorological simulation for the period of April–August 2003 when an intensive measurement campaign took place at WLG to better characterize air mass composition in western China [Wang et al., 2006]. We show that most of the high O_3 events during the study period were due to stratospheric intrusions, which were generally associated with strong upper-level jet streams. We further examine the mean zonal winds in spring and summer to compare the position of the subtropical jet stream in the two seasons and explain the dynamical cause of the summertime O_3 maxima at WLG.

2. Methodologies

2.1. Surface O_3 , CO, and Total O_3 Column Data

[4] We make use of hourly O_3 and CO data from surface measurements conducted during April to August 2003 to help identify high- O_3 events for comparison with dynamic model simulations. These gases were measured together with NO, NO_y , NMHCs, halocarbons, and fine aerosols in two intensive campaigns during the above period at WLG. The detailed description of instrumentation for O_3 and CO, as well as the interpretation of these data, is given by Wang et al. [2006]. Besides the surface observation, we also use the TOMS total O_3 data to study the column O_3 abundance during the study period.

2.2. Mesoscale Meteorological Simulation and Potential Vorticity Analysis

[5] We conduct mesoscale meteorological simulation using the PSU/NCAR MM5 model. Considering the accumulative errors of the model, we divided the whole period into subsections with 5-days intervals. The simulations were run in two interactive nested domains (DM1 and DM2) with the horizontal grid spaces as 81 km and 27 km, respectively (see Figure 1). The model has 26 vertical sigma layers divided from the ground level to the top pressure 50 hPa. Both the initial conditions and boundary conditions were generated from NCEP final analyses data on 26 pressure

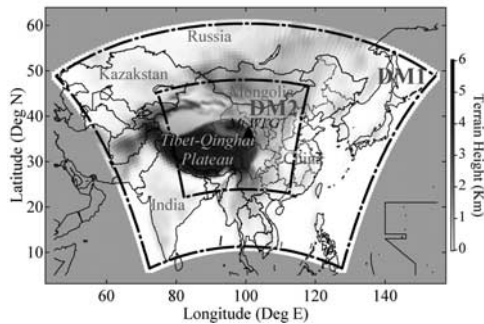


Figure 1. Topographical map showing the location of WLG and MM5 domains.

levels with a horizontal resolution of 1 degree, but were enhanced by using surface and radiosonde data of wind, temperature, and relative humidity with the LITTLE_R module of MM5. To decrease the forecast error, “analyses nudging” was employed in DM1 during the whole simulations. For physics parameterizations, we used the Blackadar scheme for boundary layer, Kain-Fritsch2 scheme for cumulus, and rapid radiative transfer model for longwave radiation. The similar modeling settings have been successfully applied in other studies [e.g., Ding *et al.*, 2004].

[6] Potential vorticity (PV) analysis is a widely used approach to view the dynamics related to stratosphere-to-troposphere exchange (STE) because PV, as the product of absolute vorticity and thermodynamic stability, is a conserved quantity in adiabatic frictionless flow, and it is greater in the stratosphere than in the troposphere [Hoskins *et al.*, 1985]. *World Meteorological Organization* [1986] defines the so-called dynamical tropopause by the 1.6 PVU isosurface ($1 \text{ PVU} = 10^{-6} \text{ K kg}^{-1} \text{ m}^2 \text{ s}^{-1}$). In this study, we use GrADS (The Grid Analysis and Display System) to diagnose PV and plot isentropic PV maps from MM5 simulations.

2.3. Separation of Different Timescale Variations in Trace Gases and PV

[7] As most of the observed O_3 events at WLG typically lasted for several days, in order to see more clearly the dynamical processes contributing to these variations we applied a Kolmogorov-Zurbenko (KZ) filter to the data set to separate the components representing different timescale variations. The detailed description of the KZ filter is given by Zurbenko [1986]. Briefly, a variable A (in our case, mixing ratio of surface CO and O_3 , total O_3 , and PV) can be divided into three parts: $A = LA + MA + SA$, where LA , MA , and SA are the long-term, synoptic, and high-frequency components, respectively. A quintic fit to A was used to estimate LA , and a 25-hour moving average, repeated four times, was used to wipe off the scales lower than $25 \times 4^{1/2}$ hours (~ 2 days) yielding the term $LA + MA$ [Zurbenko, 1986]. The difference between the two gives the synoptic term MA with a scale range from about 2 days to weeks.

3. Results and Discussion

3.1. High O_3 Events and PV

[8] Figure 2a shows the mean tropopause height defined as 1.6 PVU and synoptic component of PV over WLG, and

Figure 2b presents the long-term and synoptic component of total and surface O_3 and CO at the site from April 18 to August 17, 2003. The surface O_3 data reveals that O_3 enhancements (lasting more than two days) frequently occurred at WLG in summer especially from mid-June to July. Figure 2b shows that except for one case in mid-June, most of these O_3 events were associated with negative anomalies of CO. The negative correlation between O_3 and CO and other anthropogenic tracers such as NMHCs and halocarbons [Wang *et al.*, 2006] suggests that these O_3 enhancements were not due to long-range transport of anthropogenic pollution. Instead, these surface O_3 events were accompanied by enhancements in total O_3 and positive anomalies of PV in the upper troposphere (marked as A–E in Figure 2a), indicating intrusions of stratospheric air [Hoskins *et al.*, 1985; Vaughan and Price, 1991].

3.2. Case Analysis

[9] To understand the mechanism(s) for transporting upper-level O_3 -rich air to the surface, we examined the detailed dynamics for all the identified stratospheric events and present here one case for June 18 as an illustration. Figure 3a shows the 340 K isentropic PV and horizontal wind vectors and backward trajectories ending at different heights over WLG at 1200 UTC 18 June. On this day a jet stream with wind speed larger than 50 m s^{-1} generated a deep upper-level trough over central China. The PV map shows that high PV values abutted upon the northern edge of the jet stream indicating strong stratospheric intrusions there. WLG was just located on the western side of the upper-level trough, where stronger subsidence often occurs [Davies and Schuepbach, 1994; Stohl *et al.*, 2000]. The back trajectories also show that the air masses observed on that day predominantly came from northwest and experienced an apparent subsidence before arriving over the site. The markers whose size represents PV values at two-hour intervals also suggest stratospheric origin of these air masses. In Figure 3b, we show the cross-section of PV, v - w wind-vectors, zonal wind (dashed lines, only for speed larger than 20 m s^{-1}) and clear-air-turbulence (CAT, solid lines, calculated using the RIP4 package) on longitude 100.9E, i.e., line AA' in Figure 3a, at 1200 UTC 18 June.

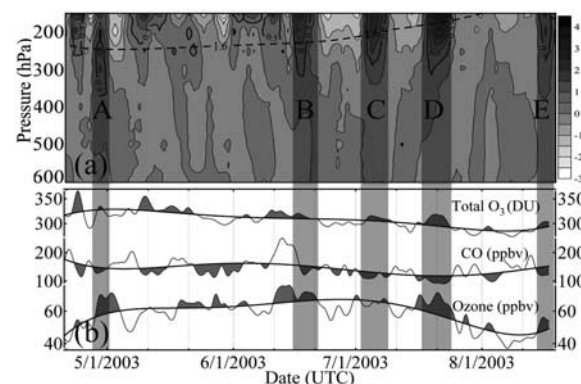


Figure 2. (a) The mean tropopause height (dashed line, 1.6 PVU isoline of PV long-term component) and synoptic component of vertical PV profile over WLG, and (b) the long-term and synoptic components of total and surface O_3 , and surface CO at WLG.

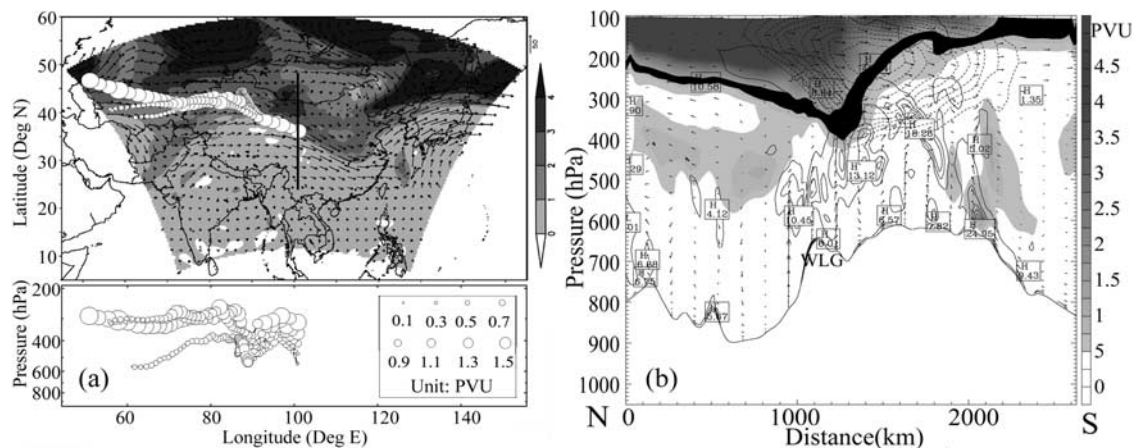


Figure 3. (a) The 340 K IPV map and back trajectories ending at 2–6 km over WLQ at 1200 UTC 18 June, and (b) cross section of PV, wind speed and clear-air-turbulence on 100.9 E at 1200 UTC 18 June.

The black stripline, representing PV values between 1.0–1.6 PVU, shows that on that day a stratospheric intrusion event occurred at the northern bottom of a jet stream with the core about at 200 hPa. This figure also shows that strong CAT existed in this event over the Qinghai-Tibetan Plateau, which could cause strong mixing in the “compressed” troposphere over the Plateau and further enhance the surface O_3 concentrations. The mechanism for the vertical exchange related to CAT has been extensively studied [e.g., Shapiro, 1980]. Analysis of other cases of intrusion during this study suggests similar dynamical characteristic in these events, i.e., the presence of a jet stream.

3.3. Discussion on the Cause of Summertime Peak of Surface O_3

[10] The above analysis shows that the high O_3 events in summer were attributed to the stratospheric intrusions on jet stream. It has been known that STE reaches a maximum strength in spring contributing to the peak of surface O_3 concentrations in many mid-latitude locations of the Northern Hemisphere [Holton *et al.*, 1995; Monks, 2000]. But why doesn't O_3 peak in spring at WLQ? To help answer this question, let's examine Figure 2a again. Although the mean tropopause over this region was higher in summer than in spring (refer to the 1.6 PVU isoline), which has been used as evidence for a “weak” summertime STE [Zhu *et al.*, 2004], the synoptic component of PV shows that there are more cases of strong stratospheric intrusions in summer, suggesting that the mean tropopause level alone cannot represent the strength of STE. The more frequent intrusions in summer, together with the mixing induced by CAT and the stronger convective mixing due to the heating of the Tibetan Plateau, should contribute a stronger transport of O_3 -rich stratospheric air to the surface. Our finding on a stronger summertime STE over Mount Waliguan for 2003 is consistent with the work of Sprenger and Wernli [2003] who calculated the northern hemispheric climatology of STE with 15 years data. Figure 2 in their work shows that the net STE over central Eurasia is stronger in summer (July), which is quite different from other regions at the same latitude. Therefore the summer peak of surface O_3 at

WLQ and possibly also for a larger part of Central Asia is likely due to the stronger STE over the region.

[11] We next examine the dynamic cause(s) of the summertime maximum STE over this region. We have shown that all cases of stratospheric intrusion observed during our study have a common dynamical feature, i.e., they were all associated with the presence of an upper-level trough along a jet stream. Many others studies have demonstrated a strong relationship between STE and a jet stream [e.g., Appenzeller and Davies, 1992; Langford, 1999]. Figures 4a and 4b shows averaged horizontal zonal wind on 200 hPa (a level near the tropopause) for spring (March–April) and summer (June–July) 2003, respectively. A subtropical jet stream can be clearly seen in both seasons but with different patterns. In spring, the axis of the subtropical jet was situated at $25^\circ\text{N} \sim 30^\circ\text{N}$ but the jet stream was “broken” over India and was positioned along the southern side of the Tibetan Plateau, possibly due to the thermal effects of the Plateau on atmospheric circulation. Under such conditions, WLQ, which is located on the northern side of the Plateau, was less affected by the jet (and thus with fewer cases of stratospheric intrusions). In comparison, the subtropical jet moved northward (to about 40°N) in summer and was positioned as a straight line over the northern side of the Plateau (just over WLQ) with the strongest jet lying over central Asian countries such as Kazakhstan. (Analysis of multiple year data (2000–2004) gave a similar result on the different position of the jet stream in spring and summer). Since the case studies in the present work and other researches [Holton *et al.*, 1995; Tsutsumi *et al.*, 1998] have

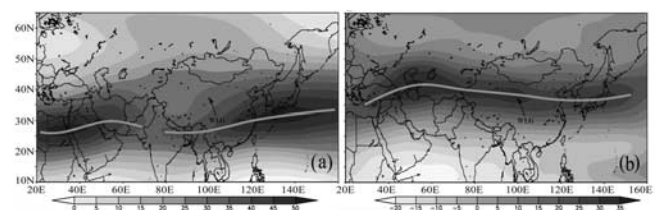


Figure 4. The averaged 200 hPa zonal wind in (a) spring (March–April) and (b) summer (June–July) in 2003.

shown that the strongest intrusion generally occurs below the northern side of a jet, a maximum of STE in central Eurasia is expected to occur in summer. This is different from the situation over eastern China and the larger eastern Asia, where the subtropical jet is positioned at a similar location in summer and spring but has a greater strength in spring [Tsutsumi *et al.*, 1998].

4. Conclusions

[12] Through meteorological simulations we have shown that the elevated surface O₃ concentrations observed at remote WLG during spring-summer 2003 were mostly caused by the downward transport of stratospheric air. Similar to many previous studies, the stratospheric intruding episodes were generally associated with the presence of a jet stream. We found more frequent intrusion of stratospheric air in summer than in spring in 2003. The stronger STE together with mixing due to clear-air-turbulence and stronger convective activities in summer over the Tibetan Plateau may have contributed to higher summertime surface O₃ concentrations over the Plateau. The different position of the subtropical jet in the two seasons results in the stronger STE in summer over western China and possible for larger parts of central Asia. Further studies are needed to quantify the contribution of the stratospheric intrusion relative to other sources to the summertime budget of surface O₃ at WLG.

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