ROLE OF REGIONAL ANTHROPOGENIC EMISSIONS IN THE SUMMRER SURFACE OZONE MAXIMUM IN THE NORTHEASTERN TIBETAN PLATEAU-MODELING STUDIES

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1. INTRODUCTION

As one of the key species in global-scale atmospheric oxidation capacity and climate change (Akimoto, 2003) the response of tropospheric ozone (O_3) to anthropogenic forcing in remote regions is currently a topic of active research and debate in global change research. Recently, the role of anthropogenic forcing in a unique seasonal pattern of surface O_3 at a remote site in the northeastern edge of the Tibetan Plateau (TP) is being argued (Zhu et al., 2004; Ma et al., 2005; Ding and Wang, 2006). In contrast to most remote stations in Northern Hemispheric mid-latitudes, observations at Waliguan (100°54'E, 36°17'N, 3810 m), the highest WMO GAW's remote station, showed a broad summer maximum pattern which has also been frequently found at rural sites in industrialized areas (Zhu et al., 2004). This pattern was distinctly different from the spring-maximum found on the southern part of the TP (Bonasoni et al., 2008). Zhu et al.(2004) suggested that long-range transport of anthropogenic pollution from eastern/central China, Central/South Asia and even Europe caused the distinct seasonal pattern. However, Ding et al. (2006) argued that stratosphere-to-troposphere exchange (STE) contributed to higher summertime surface O_3 using meteorological simulations. Ma et al (2005) also suggested that summer intensive convections on TP easily transported high ozone in upper troposphere/lower stratosphere to the surface.

Regional chemical transport model is an ideal tool because it is able to track more information about contributions of various physical (e.g. advection and diffusion) and chemical processes to pollutants. In this study, we used two regional models (CMAQv4.7 and the nested air quality prediction modeling system (NAQPMS)) to simulate the seasonal pattern of surface ozone at Waliguan. The purpose is to investigate why the maximum of surface ozone at Waliguan appears in summer.

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2. MODEL DESCRIPTION AND SETTINGS

CMAQv4.7 modeling system released by the US EPA (Byun and Ching, 1999) is a multi-scale (urban and regional) and muti-pollutant air quality model. In this simulation, it utilizes SAPRC-99 scheme for gas-phase chemistry, and AERO5 module is used for aerosol calculation. For advection and diffusion, it applies PPM and acm2_inline scheme, respectively. For vertical resolution, the CMAQ has the σ-P coordination system up to 50hpa and 27 vertical layers. The horizontal resolution is 80km.

NAQPMS is a fully modularized three-dimensional system with various options for representing the physical and chemical processes describing regional- and urban-scale atmospheric pollution. The NAQPMS has been developed at the Institute of Atmospheric Physics (IAP), Chinese Academy of Science. NAQPMS utilizes Carbon-Bond Mechanism Z (CBM-Z), which is composed of 133 reactions for 53 species. Compared with Carbon-Bond IV, the CBM-Z extends the framework to function properly at larger spatial and longer time scales (Zaveri and Peters, 1999). For aerosol dynamics, NAQPMS applies a chemically speciated and size-resolved aerosol module CAM (Gong et al., 2002). The advection scheme employs a simplified but accurate mass-conserving, peak-preserving, mixing ratio-bounded advection algorithm (Walcek and Aleksic, 1998). The dry deposition module was updated to Wesely's scheme (1989). Since 1995, the NAQPMS has been used successfully to model dust events (Wang et al., 2000), the

transport of and chemical processes within polluted air masses (Sulfur, O3) (Li et al., 2007), and the interactions between mineral aerosols and acid rain over East Asia (Wang et al., 2002). Vertically, the model uses ten terrain-following layers, five of which are within the lowest 1 km above the surface. Vertical grid spacing is increased gradually from 50m at the surface to 2000m at the top (20 km a.s.l.). The horizontal resolution is 81km.

A tracer-tagged method is introduced into NAQPMS to track the contributions of photochemically produced ozone in China (CHN) and stratospheric intrusions to surface ozone at Waliguan. Fig.1 shows the CHN in this study. The details were described in Li et al. (2008).

Both CMAQ and NAQPMS are driven by meteorological fields calculated by WRF-ARW3.0, with NCEP/NCAR ds083.2 reanalysis data sets at six hour intervals. The initial, lateral and top boundaries of both models are derived from monthly averaged climatological outputs of a global chemical transport model (MOZARTV2.4) with 2.8° resolution (Horowitz et al. 2003). Anthropogenic and biomass burning emissions are provided by RETRO project for the year 2000 (Schultz et al. 2007); natural emissions are GEIA for the year 1995(Guenther et al. 1995). Fig.1 shows the distribution of NOx ($NO+NO_2$) in this study. And the location of Waliguan is also shown.

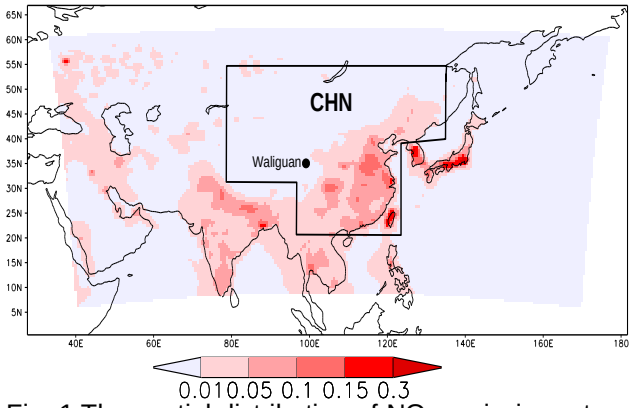

Fig. 1 The spatial distribution of NOx emission rate (shaded, μg/m2/s) in this study. The solid marks mean the location of Waliguan. Also shown is the tagged ozone production region CHN (China).

This simulation was performed from October 2000 to December 2001. The first 3 months were regarded as the spin-up period to reduce the influence of initial conditions.

3. COMPARISON OF MODELING RESULTS AND OBSERVATIONS

Fig.2 shows the observations and simulated results of NAQPMS and CMAQ at Waliguan in 2001. For NAQPMS, the model matched the observed surface O3, and PAN generally to within reasonable errors. Particularly, the simulation of surface O_3 compared more favorably to observations in summer months than in winter when there was a 5-10 ppbv underestimation. CMAQ was not able to reproduce summer surface ozone maximum at Waliguan. This was because of the overestimation of ozone concentrations in winter-spring. In March-June, ozone simulated by CMAQ reached 70-80 ppbv, which was ~20 ppbv and 25 ppbv higher than observations and NAQPMS. Overestimation of STE over TP in CMAQ was one of likely key causes. It's well known that the intensity of STE shows a maximum over North Hemisphere, particularly in high altitudes. Tibetan Plateau, the highest plateau with averaged elevation of approximately 4000m, was more sensitive to the intensity of STE than other places in the world. In summer, strong updrafts over TP (Ye et al, 1998) offset the effect of STE. As a result, the simulated ozone of CMAQ and NAQPMS and observations showed close to each other (~5 ppbv gap). The comparison of PAN seemed to support this assumption (Fig.2b). As a photochemical product, PAN from NAQPMS and CMAQ matched well, and showed high summer peroxyacetyl nitrate (PAN) with a mean value of 0.4-0.5 ppbv as observations, which was generally higher than other remote sites (0.01-0.36 ppbv).

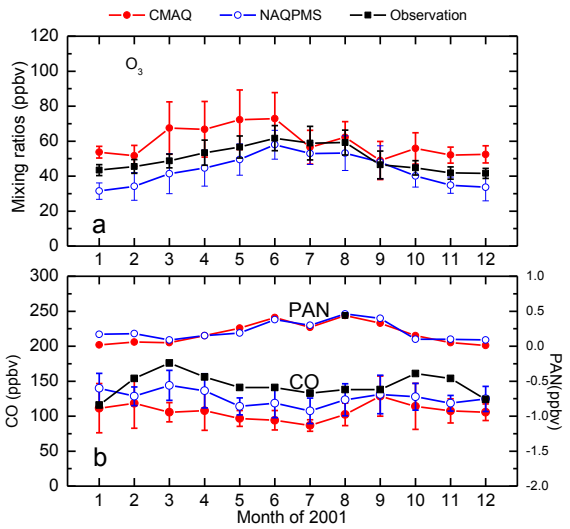

Fig.2 Observed and simulated surface O3 (a), CO and PAN (b) at Waliguan in 2001. Note that for observations, surface O3 is in 2001; PAN is in 2006 from Zhang et al. (2009); CO is in 2001 from The World Data Centre for Greenhouse Gases (WDCGG).

Fig.3 presents the seasonal distribution of surface ozone simulated by CMAQ in East Asia in 2001. Over the whole TP, ozone reached 65-70 ppbv in spring, while it ranged from 45-60 ppbv in summer.

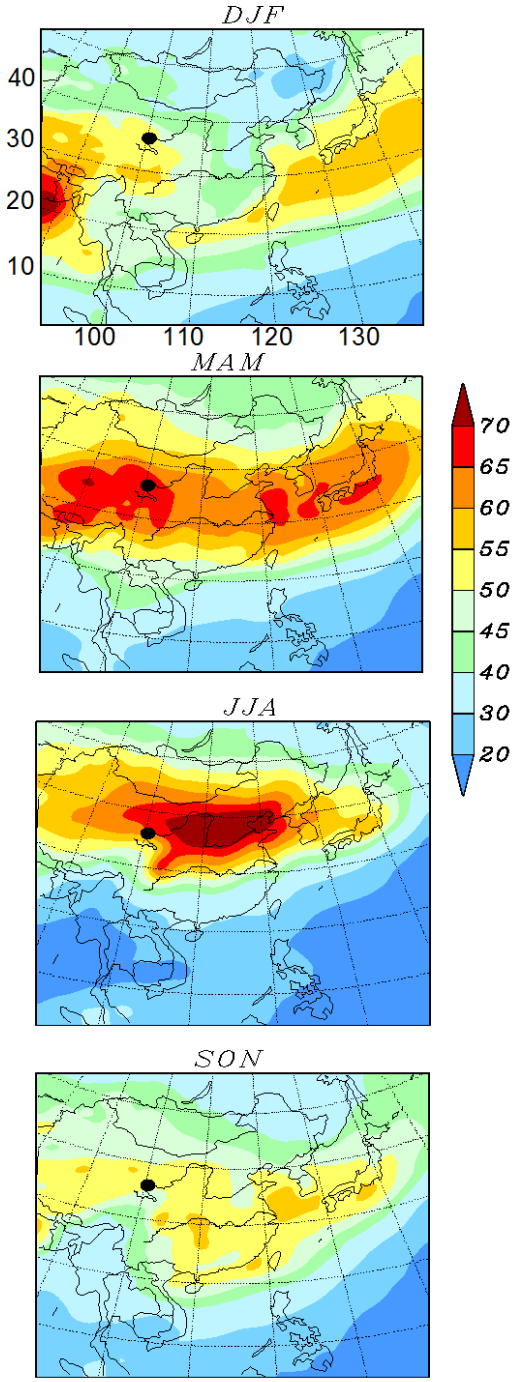

Fig.3 Spatial distributions of surface ozone concentrations (ppbv, shaded) simulated by CMAQ in 2001. (DJF: winter; MAM: spring; JJA: summer; SON: fall)

4. ROLE OF CHINA ANTHROPOGENIC EMISSIONS IN THE SURFACE SUMMER O_3 MAXIMUM AT WALIGUAN

As stated in section 1, a tracer-tagged module in NAQPMS was used to track the contributions of different origins to ozone. Fig. 4 shows the contributions from stratospheric intrusion, lateral boundary and ozone production in China to surface ozone at Waliguan. Clearly, the transport of photochemically produced O3 in China was the most important mechanism for the summer maximum, leading to seasonal mean O3 mixing ratios of 23.7 ppbv in summer which was 3-5 times that of spring. In contrast, contributions from lateral boundaries were minimal in summer at 16.8 ppbv. Summer O3 resulting from STE was only 4.7 ppbv and less than spring.

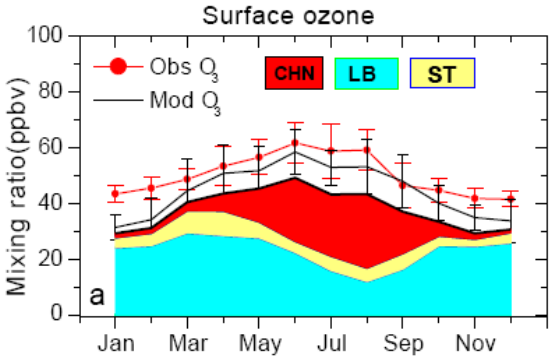

Fig. 4 NAQPMS simulated and observed surface ozone (ppbv) with contributions from stratospheric ozone (ST), lateral boundary (LB) and ozone production in China (CHN).

Summer intensive long-range transport from ozone photochemical production in China was caused by a unique weather pattern in TP. Under the Asian summer monsoon, TP experiences a strong near-surface convergence and upper layer divergence driven by elevated surface heating and low air density in summer (Ye et al., 1998). Near-ground strong divergence (positive values, net outflow) around the plateau shown in Fig.5 indicates that large amounts of air were transported towards the convergence zone in the plateau to balance the low-level cyclonic vorticity. The prevailing easterly winds in PBL (Fig. 5) transported pollutants over North Central China (NCC, 100-110oE, 30-40oN), a polluting region with a summertime intensive O3 photochemical production (5-15 ppbv/day) (Li et al., 2007), to the northeastern edge of the plateau in the favor of orographical and convective lifting in the eastern slope of the Tibetan Plateau. Fig.6b clearly

illustrates this close circulation in summer transported surface polluted air to Waliguan till 7 km a.s.l. In contrast, air in the eastern Tibetan Plateau came from the western upper troposphere and descended in central/east China in April (Fig. 6a).

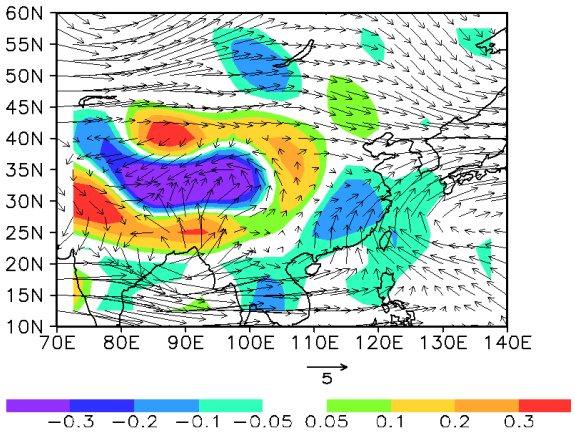

Fig.5 NCEP divergence (1×10^{-5} s^{-1}) at 700 hPa in summer (June-August).

5. SUMMARY

In this study, we used two regional chemical transport models to investigate why the maximum of surface ozone at Waliguan appears in summer, rather than spring-maximum at most remote stations in Northern Hemispheric mid-latitudes. The results showed that NAQPMS reproduced this distinct seasonal pattern, while CMAQ failed to reproduce it where a spring-maximum was found. CMAQ overestimated winter-spring ozone at Waliguan with a magnitude of ~20 ppbv than observations. Overestimation of STE over TP in CMAQ was likely one of key causes.

The tracer-tagged module introduced into NAQPMS was used to quantify impacts of anthropogenic emissions and stratosphere-to-troposphere exchange (STE) on surface ozone on TP. Model results clearly showed that the regional transport of photochemically produced ozone in China was significantly responsible for the summer maximum in the northern TP with a contribution of ~10-25 ppbv in summer, which was more than ~5 ppbv contribution of STE. The unique strong near-surface convergence and upper layer divergence driven by elevated surface heating and low air density over TP in summer was its dynamic cause.

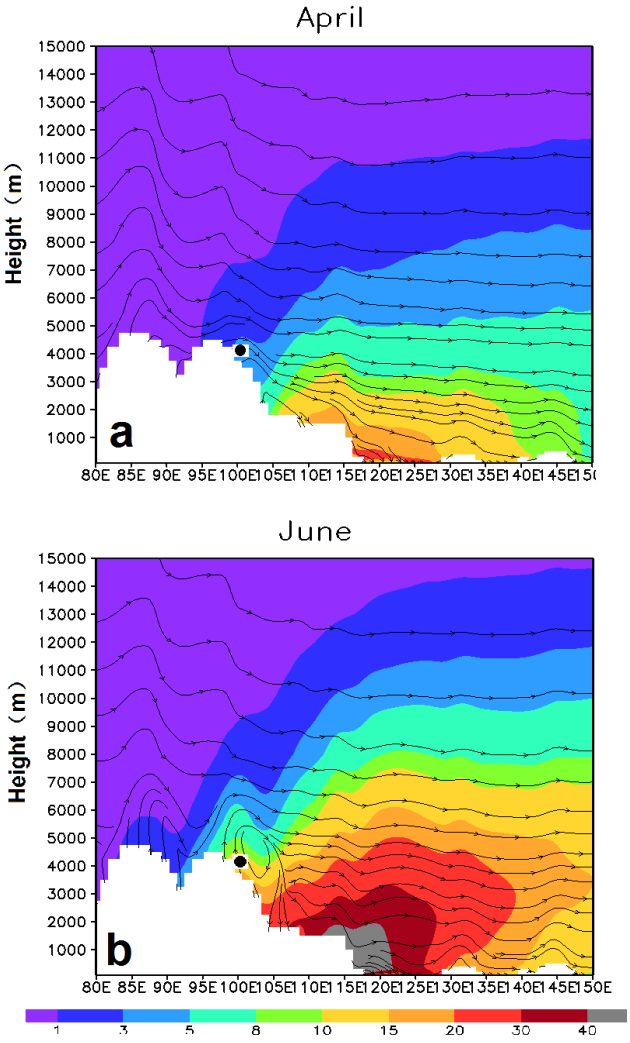

Fig. 6 The Longitude-Height distribution of tagged O3 (ppbv) produced in China and the flow streams along 37°N in (d) April and (e) June; white areas indicate topography. Waliguan is marked by solid circles.

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