Ozone Air Quality during the 2008 Beijing Olympics: Effectiveness of Emission Restrictions

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Abstract:

A series of aggressive measures was launched by the Chinese government to reduce pollutant emissions from Beijing and surrounding areas during the Olympic Games. Observations at Miyun, a rural site 100 km downwind of the Beijing urban center, show significant decreases in concentrations of O_3, CO, NO_y, and SO_2 during August 2008, relative to August 2006-2007. The mean daytime mixing ratio of O_3 was lower by about 15 ppbv, reduced to 50 ppbv, in August 2008. The relative reductions in daytime SO_2, CO, and NO_y were 61%, 25%, and 21%, respectively. Changes in SO_2 and in species correlations from 2007 to 2008 indicate that emissions of SO_2, CO, and NO_x were reduced by 60%, 32%, and 36%, respectively, during the Olympics. Analysis of meteorological conditions and interpretation of observations using a chemical transport model suggest that although the day-to-day variability in ozone is driven mostly by meteorology, the reduction in emissions of ozone precursors associated with the Olympic Games had a significant contribution to the observed decrease in O_3 during August 2008, accounting for 80% of the O_3 reduction for the month as a whole and 45% during the Olympics Period (8-24 August). The model predicts that emission restrictions such as those implemented during the Olympics can affect O_3 far beyond the Beijing urban area, resulting in reductions in boundary layer O_3 of 2-10 ppbv over a large region of the North China Plain and Northeastern China.

1. Introduction

Ozone is produced in the troposphere by photochemical oxidation of carbon monoxide (CO) and volatile organic carbon (VOCs), initiated by reaction with OH in the presence of NO_x. In surface air, ozone has an adverse impact on both humans and vegetation (NRC, 1991). Due to its relatively long lifetime, it can be transported over long distances from source regions, making ozone pollution an issue of global concern.

China's rapid economic growth in recent years has resulted in large increases in pollutant emissions (*Zhang et al.*, 2007; *Ohara et al.*, 2007) with important implications for ozone on both regional and global scales. Beijing, China's capital, is one of the world's largest metropolises with a population of over 15 million with a vehicle fleet of more than 3 million. Beijing's air quality problems were characterized historically by high concentrations of particulate matter and sulfur dioxides (*Hao and Wang*, 2005). In recent years, due to a rapid increase in vehicular emissions, ozone pollution has drawn increasing attention in Beijing (*Hao and Wang*, 2005; *Wang et al.*, 2006), especially in the period leading up to the Summer Olympic Games (August 2008) (http://www.nytimes.com/2007/12/29/world/asia/29china.html). Formulating a successful strategy to address O_3 pollution poses a difficult challenge as a consequence not only for Beijing but also for other regions of the developed and developing world.

To improve air quality during the Olympics (8-24 August 2008) and the Paralympics (9-17 September 2008), the Chinese government implemented a series of aggressive measures to reduce pollutant emissions in Beijing and surrounding areas for more than two months during the time periods of the Olympics and the Paralympics. From 1 July to 20 September 2008, all vehicles that failed to meet the European No. I standards for exhaust emissions (including light-duty and heavy-duty trucks and inefficient personal vehicles) were banned from Beijing's roads. Mandatory restrictions were implemented from 20 July to 20 September for personal vehicles, allowing them on roads only on alternate days depending on license plate numbers (odd-numbered vehicles on odd-numbered days and even-numbered vehicles on even-numbered days). As a result, traffic flows in Beijing urban areas were found to have declined by 22% during the

Olympics (Y. Wu, personal communications). In addition to traffic emission controls, other area and point sources in Beijing were placed under strict control during the same period. Power plants in Beijing were required to reduce their emissions by 30% from their levels in June when they had already met the Chinese emission standard. Several heavily-polluting factories were ordered to reduce their operating capacities or to completely shut down during the Games. All construction activities were placed on hold. Since it has been shown that Beijing's air quality problems also have regional causes (*Streets et al.*, 2007; *L. Wang et al.*, 2008), emission controls on large industrial sources were also applied in surrounding provinces (e.g., Inner Mongolia, Shanxi, Hebei, Shandong) and in the city of Tianjin. Traffic restrictions similar to Beijing's were instituted in Tianjin during the Olympics Games.

As a result of these initiatives, one would expect to see significant decreases in emissions of ozone precursors (CO, NO_x, and VOCs) and other key pollutants (SO_2 and particulates, for example) in Beijing. *Wang et al.* [2007] and *Cheng et al.* [2008] demonstrated that the four-day traffic restrictions in Beijing during the Sino-African Summit in early November 2006 resulted in significant temporary reductions in concentrations of NO_x and particulates in the city. Compared with the Sino-African Summit, the emission reductions during the Olympic Games were more aggressive, affecting more than the transportation sector and lasting much longer. The effect on ozone is an important research question as the dependence of O_3 production on NO_x and VOCs is significantly different between the so-called NO_x-limited regime and the hydrocarbon-limited regimes (*Sillman et al.*, 1990). Many previous studies investigated the nonlinear O_3 chemistry and challenges of combating O_3 pollution in cities of developed countries (*Sillman et al.*, 1995; *Murphy et al.*, 2006, 2007; *Harley et al.*, 2005; *Trainer et al.*, 2000). Compared with these studies, basic scientific understanding about surface O_3 and precursor emissions in Chinese cities has been minimal. For example, examining the day-of-week variations of O_3 provides a useful methodology to improve understanding of nonlinear ozone chemistry for many western cities and downwind regions (*Murphy et al.*, 2006, 2007). However, no such 'weekend' effect has been observed for precursor emissions in China (*Beirle et al.*, 2003), likely due to different

emission patterns related to social-economic factors in China. This suggests that it is essential to study in situ observations of important atmospheric species in China.

The present study will focus on the impact of the Olympics emission restrictions on air quality over Beijing, particularly on surface ozone in the summertime. Without a network of multiple observational sites over different parts of Beijing urban area, this study employs long-term, continuous measurements of O_3, CO, NO_y, and SO_2 at a suburban/rural site (*Wang et al.*, 2008b) located directly downwind of the Beijing urban area during summer months. Species correlations at the site will be used to infer 'top-down' constraints on the magnitude of emission restrictions during the Olympics. The nested-grid version of the GEOS-Chem global chemical transport model (*Wang et al., 2004a; Chen et al., 2009*) will be employed to interpret the observations and to evaluate the reductions in emissions during the Olympics.

The emission restrictions associated with the Olympic Games offer an invaluable opportunity to test our understanding of the chemistry and dynamics affecting ozone and its precursors in a major Chinese urban environment. The ability of chemical transport models (CTMs) to reproduce changes in tropospheric ozone arising in response to these emission changes provides an important test of these models.

We begin by introducing the Miyun site and the nested-grid GEOS-Chem model. The paper is organized then in two parts. The first is devoted to observational results. Trace gas concentrations and meteorological conditions measured at the Miyun site in August 2008 are compared with observations for Augusts of the two preceding years, demonstrating significant decreases in O_3, CO, NO_y, and SO_2 during August 2008. We show that the reduction in pollution levels during the Olympics, far exceeding the magnitude attributed to year-to-year changes in meteorology, reflects most a response to the emission reductions. Using species concentrations and their correlations observed at Miyun, quantitative estimates are derived for the magnitude of emission reductions for SO_2, CO, and NO_x during the Olympics employing a 'top-down' approach independent of any modeling or bottom-up information. The latter part of the paper focuses on a model-based analysis. The extent to which the 'top-down' estimates of emission reductions improves the performance of the model in simulating the observations at

Miyun provides an independent evaluation of the observational analysis conducted in the first part of the paper. Model sensitivity analysis is used to differentiate quantitatively between meteorology- and emission-driven changes in ozone during the Olympics. The impact of the emission reductions of O_3 at a regional scale is also predicted by the model. Concluding remarks are presented in Section 5.

2. Observations and Model

2.1 Surface Observations

The Miyun site (40º 29'N, 116º 46.45' E) is located at an elevation of about 152 m in Miyun County (population of about 420,000), about 100km northeast of the Beijing urban area. The terrain to the south of the site falls off gradually to about 90 m in a region characterized by a mix of agriculture and small villages. Mountains rise steeply to the north. There are no big point sources between the Beijing urban area and the site, nor close to the site in other directions. A map of the Beijing-Miyun region was shown in *Wang et al.* (2008b) (Figure 1) and is not reproduced here. The station was established through a collaboration between the Harvard China Project and Tsinghua University. The measurements began in November 2004 and include continuous observations of O_3, CO, CO_2, together with basic meteorological data (temperature, relative humidity, and wind speed and direction). Additional instruments measuring NO, NO_y, and SO_2 were added in 2006, with data collection for these species initiated in early 2007. The present study focuses on measurements of O_3, CO, NO_y, and SO_2 for July, August, and September (JAS) 2008 when the emission restrictions were in place. Mixing ratios of these species measured for the same period in 2006 and 2007 are used for comparison. Measurements in summer 2005 had many gaps due to instrumental problems and are not included for comparison. Figure 1 shows afternoon wind directions recorded at the Miyun site in August 2006-2008. The prevailing SSW-SW-S winds suggest that the Miyun site is located directly downwind of the Beijing urban area in summer. Miyun observations are representative therefore of plume conditions of Beijing urban pollution in summer.

The O_3 and CO instruments and the site details are discussed in *Wang et al.* (2008b).

The NO_y and SO_2 instruments are outlined here. The NO_y mixing ratio is measured by the chemiluminescence method (Thermo Environmental Instruments 42C-Y). Sample air is drawn first into an inlet 6 m above the ground and then split into two parallel channels. The NO_y channel uses a heated molybdenum converter to reduce all forms of NOy to NO. The catalyst is preceded by as short a section of Teflon tubing as practical to minimize loss of HNO_3 and other surface active compounds on tubing walls before they reach the catalyst. The instrument response and catalyst efficiency are calibrated every 6 hours by introducing NIST traceable standard NO and n-proply nitrate into the sample air in sequence. The SO_2 mixing ratio is measured by a pulsed fluorescence method (Thermo Environmental Instrument 43CTL). Sample air is drawn from the same inlet as CO and O_3, and a mass flow controller upstream of the instrument maintains constant flow in the system. The instrument zero is determined every 2 hours by passing sample air into a denuder coated with sodium carbonate. A calibration sequence is implemented every 6 hours by introducing NIST traceable SO_2 standard into sample air. The mixing ratio is computed by subtracting the zero offset from ambient signal voltage and multiplying by the instrument gain.

2.2 Model description

The nested-grid GEOS-Chem model developed by *Chen et al.* (2009) is employed in the present study. The GEOS-Chem global 3-D model for tropospheric chemistry is driven by meteorological data assimilated by the Goddard Earth Observing System (GEOS) at the NASA Global Modeling and Assimilation Office (GMAO). The present study uses GEOS-5 meteorology covering the period from Dec 2004 to present. The meteorological data include 3-D fields updated every 3 hours for surface fluxes and mixing depths and every 6 hours for other variables. The horizontal resolution is 0.5º latitude by 0.667º longitude, with 72 levels in the vertical extending from the surface to 0.01 hPa. The lowest 2 km is resolved into 14 layers with midpoints at altitudes of 70, 200, 330, 460, 600, 740, 875, 1015, 1157, 1301, 1447, 1594, 1770, 2000 m for a column based at sea level. For inputs to the global GEOS-Chem model, the horizontal resolution of the meteorological fields is degraded to 2º latitude x 2.5º longitude or 4º latitude x 2.5º

longitude due to computational limitations. Details of the degrading process are provided by *Wang et al.* (2004a).

The structure of the nested-grid GEOS-Chem model involves a window with a uniform horizontal resolution of 0.5^o x 0.667^o embedded in a low-resolution (4^o x 5^o) global background. The nested-grid GEOS-Chem retains the generic high horizontal resolution of the GEOS-5 data over the nested regional domain. For the present study, the nested domain is set at 70°E-150°E and 11°S-55°N and includes all of China, its neighboring countries, and a significant portion of the northwestern Pacific (*Wang et al.*, 2004a; *Wang et al., 2004b*; *Chen et al.*, 2009). The high-resolution regional simulation is coupled dynamically to the low-resolution global model through lateral boundary conditions that are updated every three hours.

The GEOS-Chem model includes a detailed tropospheric O_3-NO_x-hydrocarbon-aerosol simulation. The aerosol and oxidant chemistry are coupled through the formation of sulfate and nitrate, heterogeneous chemistry, and aerosol effects on photolysis rates. Photolysis frequencies are computed using the Fast-J radiative transfer algorithm (*Wild et al.*, 2000) which allows for Rayleigh scattering as well as for Mie scattering by clouds and aerosols. Simulation of wet and dry deposition follows the schemes developed by *Bey et al.* (2001). Application and evaluation of the model over China have been described by *Wang et al*. (*2004a; 2004c*). Anthropogenic emissions of NO_x, CO, SO_2, and VOCs over the nested East Asia domain were taken from *Zhang et al.* (2009) for the year 2006. Since our analysis focuses on the differences in model results over Beijing between 2007 and 2008, 2007 is chosen as the base year with which to represent emissions from Beijing. Anthropogenic emissions for Beijing in 2007 are taken from detailed inventory work carried out by researchers at Tsinghua University. The inventory for Beijing was developed bottom-up and has a spatial resolution of 4 km x 4 km. It was compiled from detailed energy statistics for Beijing, road network databases, locations of power plants and large industrial facilities, population distribution, and surveys of other key parameters related to activity rates. Emission factors for pollutants were obtained from a detailed technology-based approach reflecting rapid renewal of combustion equipment and processes, combined with field measurements of representative combustion types

(S.X. Wang et al., Emission reductions and air quality improvements of air quality control measures during the 2008 Olympics in Beijing, *Environ. Sci. Tech.*, submitted, 2009).

3. Air quality improvement during the Olympics

As summarized in the introduction, some emission-reduction measures started later than others, although all were in place during the time period of the Olympics (8 – 24 August, 2008). Therefore, in order to evaluate the aggregate effects of the emission-reduction policies, most of the analysis below will focus on pollutant concentrations for August 2008. We showed in a previous study that O_3 peaks in June at Miyun and that mean daytime O_3 in August is on average 10 ppbv lower than that in June (*Wang et al.,* 2008). Other observations for Beijing have shown similar seasonal patterns in surface O_3 (*Ding et al.,* 2007; *Lin et al.,* 2008). To minimize the compounding effects of this natural seasonal variability of O_3 and other species, our analysis compares trace gas levels in August 2008 to the same periods in 2006 and 2007, rather than comparing August to June or other months of 2008. No 'weekend effect' has been observed for precursor emissions in China (*Beirle et al.,* 2003) and we did not find a 'weekend effect' on O_3 observed at Miyun. Therefore, we did not distinguish weekend from weekday observations in the following discussion.

3.1 Trace gas concentrations

Figure 2 presents mean daytime mixing ratios of O_3 (2a) and CO (2b) as observed at Miyun in summer (July-August-September; JAS) 2006-2008. In JAS 2008, mean daytime mixing ratios of CO and O_3 dropped significantly compared to the same months in 2006 and 2007. The mean daytime mixing ratio of CO was 350 ppbv in August 2008, about 150 ppbv (or 30%) lower than for August in 2006 and 2007. The mean daytime mixing ratio of O_3 decreased by about 15 ppbv between Aug 2006-2007 and Aug 2008, from 65 ppbv to 50 ppbv. During the Olympics (4 August – 24 August 2008), daytime O_3 averaged 42 ppbv. The decreases in O_3 and CO, compared to the same months in 2006-2007, are larger than the magnitude of interannual variations observed at Miyun. In

2006 and 2007, mean O_3 in August was found to be comparable to or even greater than that in July, while in 2008 the pattern was reversed, with O_3 much lower in August.

Figure 2c presents hourly mixing ratios of O_3 observed at Miyun in August 2006-2008. It is clear that O_3 was lowest in 2008 for almost every day in August except the few days at the beginning of the month, which were attributed to unfavorable meteorological conditions (to be discussed in Section 4.2). The decreases in August 2008 were most significant in the afternoon when photochemical production of O_3 is most active. Mixing ratios of CO peak in the afternoon in summer, similar to O_3, indicating the arrival of urban pollution plumes (*Wang et al.*, 2008). The decrease in afternoon mixing ratios of O_3 in August 2008 indicates reductions in chemical production of O_3 in urban pollution plumes. The reduction in O_3 was not only reflected in mean concentrations but also in peak concentrations. The number of hours with 1-hr average concentrations of O_3 exceeding 102 ppbv (Chinese air quality standard for ozone) decreased from an average of 25 hours in August 2006 and 2007 to only 3 hours in August 2008.

Figure 3 displays afternoon mixing ratios of O_3, CO, NO_y, and SO_2 observed at Miyun in August 2006-2008 as a function of wind direction. For NO_y and SO_2, data are available only for 2007-2008. Lower mixing ratios of all the species were observed for all wind directions in August 2008. The largest reductions in trace gas concentrations were observed for air masses arriving from SW, SSW and S, i.e. from the Beijing urban area. Table 1 compares mean afternoon mixing ratios of the trace gases associated with SSW-SW-S winds observed at Miyun between August 2007 and 2008. For these air masses, the relative reductions in mixing ratios of SO_2, CO, NO_y, and O_3 from August 2007 to August 2008 are 61%, 25%, 21%, and 26%, respectively.

3.2 Meteorological conditions

Production and transformation of O_3 depend critically on meteorology and weather patterns. In this section we compare the meteorological parameters measured at Miyun for Augusts of the three years investigated in this study. As illustrated in Figure 1, the prevailing winds were from SSW and SW in August 2007 and 2008 and from S and SSE in August 2006. However, mixing ratios of O_3 and CO associated with S and SSE winds

in August 2006 were comparable to those associated with SW and SSW winds (c.f. Figure 3a and 3b), with both representing polluted air masses from urban areas to the south of the site. Therefore, we conclude that there were no significant changes in the characteristics of air masses associated with the prevailing winds arriving at the site over Augusts of the three years.

Temperature and relative humidity (RH) are two key meteorological parameters measured at the site. Temperature controls key chemical reactions. RH is an indicator of water vapor content in the air with respect to saturation levels. Reaction of O (^{1}D) with H_2O is the dominant pathway for chemical losses of O_3 in the troposphere ((^{1}D) + H_2O → OH, with subsequent reactions OH + O_3 → HO_2 + O_2 and HO_2 + O_3 → OH + 2O_2). RH is closely associated with weather patterns. RH is typically higher on cloudy and precipitation days than on sunny conditions. As cloudiness and precipitation are unfavorable for photochemical production of ozone at the surface, RH tends to be negatively correlated with O_3 (Davis et al., 1999; Elminir, 2005). Our prior study (*Wang et al.*, 2008b) discussed the negative correlation between RH as an indicator of cloudiness and O_3 in summer 2006 at the Miyun site.

Although wind direction measured locally at the site is not equal to the direction from which air masses originate, grouping observations for the whole month by wind direction can still give some statistical association with the origins of air masses. Figure 4 presents temperature and RH observed at Miyun in August 2006-2008 as a function of wind direction. Northeasterly winds (NNE, NE, ENE) were sampled 10% of the time in August 2008, more frequently than in either August 2006 or 2007 (2% and 4% of the time, respectively) (Figure 1). As shown in Figure 3, the mean mixing ratio of O_3 in NNE-NE-ENE air masses dropped from 50 ppbv in August 2006-2007 to 35 ppbv in August 2008, with relatively smaller or no changes for CO, NO_y, and SO_2, suggesting that the reductions in O_3 may be attributed to differences in meteorological factors such as temperature and relative humidity (RH) rather than in precursor emissions. For air masses from the NNE-NE-ENE directions, the mean temperature was 4^o C lower in August 2008 as compared to August 2006-2007, while RH was higher by 15% (Figure 4). Since lower temperature and higher RH are normally considered meteorological

conditions that are not conducive to photochemical production of O_3, this meteorological difference could account for reduced O_3 levels in NNE-NE-ENE air masses sampled in August 2008 relative to those in 2006 and 2007. However, winds from the NNE-NE-ENE sector are infrequent at the site in August, and lower O_3 mixing ratio for this sector relates to short-term, day-to-day, variability in meteorology and thus can only account for 2 ppbv of the reduction in monthly mean O_3 for August 2008. The effect on other species is even smaller. Therefore, the large decreases observed for O_3 and other species in August 2008 (e.g., a 15 ppbv reduction in O_3) would have to be related to the majority of air masses from SSW-SW-S-SE directions, where significant reductions in emissions took place in the Beijing urban area during the Olympics.

For SSW-SW-S-SE air masses, mean temperature and RH in August 2008 were not significantly different during Augusts of 2006 and 2007. For these air masses, mean temperature was 27^oC in August 2008, as compared to 26^oC and 29^oC in August 2006 and 2007, respectively. Average RH's in August 2006, 2007, and 2008 were 70%, 50%, and 60%, respectively. The difference in the mean mixing ratios of O_3 between August 2006 and 2007 is -4 ppbv, corresponding to a difference of -3^oC in mean temperature and +20% in RH. By comparison, the difference in mean O_3 between August 2008 and 2007 is -15 ppbv, despite only a -1^oC difference in mean temperature and a +10% difference in RH. This suggests that inter-annual variations in temperature and RH could not be the only explanation for the unusually low concentrations of O_3 and other species observed at Miyun in August 2008.

Figure 5 presents ozone observations in August 2006-2008 as a function of other meteorological variables (temperature, RH, and wind speed). Each of the meteorological variables has similar ranges for Augusts of the three years, except for the lack of high wind speed sector (> 4 m/s) in August 2008 (to be discussed below). For each of the sectors of temperature, RH, and wind speed shown in Figure 5, mean O_3 levels of Augusts 2006 and 2007 were always higher compared with August 2008. The differences tend to be larger at higher temperature and lower RH, which are typically favorable meteorological conditions for ozone pollution. This suggests that meteorology cannot be the only factor contributing to the reductions in O_3 in August 2008.

Mean daytime wind speed was 1.3 m/s in August 2008, slightly lower than that of 1.7 m/s in August 2006 and 2007. For SSW-SW-S-SE winds, mean speed in the afternoon was 1.6 m/s in August 2008 as compared to 2.3 m/s in August 2007. If slower southwesterly winds could be interpreted as indicating slower and less efficient transport of pollution from the Beijing urban region, this in combination with reductions in urban pollutions during the Olympics offers a plausible explanation for lower concentrations of O_3 and other pollutants observed at Miyun in August 2008.

We showed in a previous study that optically thick clouds associated with summer monsoonal rainfall have a significant radiative impact on O_3 at Miyun (*Wang et al.*, 2008). Cloud optical depth (COD) retrieved from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument aboard the Aqua satellite (*Platnick et al.*, 2003; MYD08_M3, MODIS level-3 monthly global product at $1^o x 1^o$ resolution) showed slightly lower COD for August 2008 (COD=17.7), as compared to August 2007 (COD=22.6). Therefore, interannual variations in precipitation and COD were ruled out as the key factors responsible for lowering O_3 as observed in August 2008.

3.3 Species correlations and 'top-down' estimates of emission reductions

In this section, we employ an observation-based approach to derive quantitative estimates on the magnitude of emission reductions for SO_2, CO, and NO_x during the Olympics. First, the relative reduction in emissions of SO_2, the shortest-lived chemical species of the three, is estimated from observations. The reductions in emissions of CO and NO_x, relative to SO_2, are inferred subsequently using the dCO/dSO_2 and dNO_y/dSO_2 correlation slopes observed at Miyun.

Due to the relatively short lifetime of SO_2 (a few hours in summer considering both gas phase and aqueous phase reactions), background SO_2 concentrations at Miyun are lower than concentrations of CO and NO_y. As illustrated in Figure 3, for the relatively clean air masses from the northeast sampled at Miyun, mean mixing ratios of SO_2, NO_y, and CO are about 0.5 ppbv, 5 ppbv, and 200 ppbv, respectively. Given the negligible background level for SO_2, the difference in mixing ratio from the urban pollution plumes between August 2007 and 2008 can be assumed to be caused by changes in emissions and

variations in the chemical lifetime of SO_2. One can expect that the chemical lifetime of SO_2 and its transport time from the Beijing urban area to the site depend on meteorological conditions. To separate the effects of meteorology on the lifetime of SO_2, the S-SW-SSW air masses sampled at Miyun during August 2007 and 2008 were divided into several data intervals according to their temperature, RH, and wind speed. Under the constraint that each interval contain at least five observational data points for each month to allow for statistical representation, we identified 6 intervals by temperature, 6 by RH, and 4 by wind speed, as illustrated in Figure 6a, 6b, and 6c, respectively. Mean mixing ratios of SO_2 were calculated for each interval as well as the relative difference (RD) in SO_2 between August 2007 and 2008. The mean RD averaged for all the intervals is assumed to indicate reductions in emissions, while the variance about the mean represents random variations in the chemical lifetime and transport time of SO_2 at Miyun. We found that the mean RD in SO_2 (weighted by the frequency of the meteorology classes in Figure 6) was 40% and the variance in SO_2 lifetime was 10%, representing the uncertainty in our estimate of emission reductions. We estimated therefore that during the Olympics period (i.e. August 2008), SO_2 emissions in Beijing were reduced by 60% (±10% uncertainty) compared with the same month the year earlier.

Figure 7 shows scatterplots of NO_y versus CO (7a), SO_2 versus CO (7b), and SO_2 versus NO_y (7c) observed at Miyun during August 2007 and 2008. As we are interested primarily in pollution from the Beijing urban area, the figure presents only data for the SSW-SW-S air masses sampled at Miyun. Linear regressions for the observations are obtained with the reduced major axis (RMA) method that allows for uncertainty in both variables (*Hirsch and Gilroy*, 1984). The three species are positively correlated at Miyun, suggesting that they originate from co-located sources. The enhancement ratios between two species (e.g., dCO/dSO_2), derived from the slope of the regression line, provide useful constraints on their emission ratios. Since the Miyun site is somewhat removed from fresh urban emissions, the enhancement ratios measured at the site will be affected also by differences in lifetimes between the species. In the following analysis, focusing on relative instead of absolute changes in enhancement ratios between August 2007 and 2008, we shall assume that the lifetime ratio between the species does not change

significantly between the two periods, given that meteorological variability influences the lifetime of all the species.

The dCO/dNO_y enhancement ratio at Miyun is 37.5 mol/mol in August 2008, similar to the value of 36.5 mol/mol observed in August 2007, indicating that the fractional reduction of emissions was similar for CO and NO_x in August 2008. In contrast, the dCO/dSO_2 and dNO_y/dSO_2 enhancement ratios in August 2008 are significantly larger than values observed in August 2007. This suggests that the fractional reduction of SO_2 emissions in August 2008 is much greater than that for CO or NO_x, resulting in the observed increases in the enhancement ratio of CO/SO_2 and NO_y/SO_2.

The relative reduction in CO and NO_x emissions can be inferred using the dCO/dSO_2 and dNO_y/dSO_2 enhancement ratios observed at Miyun. As shown in Figure 7, dCO/dSO_2 increased from 54.7 (±3.1) mol/mol in August 2007 to 93.2 (±7.5) mol/mol in August 2008. Given a 60% reduction in SO_2 emissions derived above, this suggests a 32% (±14% uncertainty) reduction in CO emissions during the Olympics. Similarly, dNO_y/dSO_2 increased from 1.7 (±0.1) mol/mol in August 2007 to 2.7 (±0.2) mol/mol in August 2008, suggesting a 36% (±14%) reduction in NO_x emissions. The relative emission reductions derived here using atmospheric measurements can be regarded as 'top-down' in contrast to the 'bottom-up' method based on analyzing changes in energy consumption or emission factors. Our 'top-down' estimate suggests that in August 2008, total emissions of SO_2, CO, and NO_x in Beijing were reduced by 60%, 32%, and 36%, respectively, compared to the same month the year before.

Our results suggest that the emission control on SO_2 was most effective during the Olympics. Emissions of SO_2 originate largely from coal-burning point sources such as power plants and industrial boilers, while sources of CO and NO_x are more diversified. Transportation accounts for a large fraction of emissions for both CO and NO_x, while the power sector is a greater contributor to both SO_2 and NO_x. The flue gas desulphurization (FGD) equipment installed on power plants in Beijing and mandated to operate at full capacity since Jun 2008 (Wang et al., submitted manuscript) can remove over 95% of SO_2 from smoke stacks, whereas NO_x-control technologies with the same effectiveness such as SCR (Selective Catalytic Reduction) are not widely installed because of high cost

involved. The low-NOx burner technology adopted in current Chinese power plants can remove NO_x emissions only by 30% at most (Zhao et al., 2009). Control measures targeted at the power sector would therefore be more effective in reducing SO_2 relative to NO_x, resulting in increases in the NO_x/SO_2 ratio. The traffic restriction during the Olympics would decrease the CO/NO_x ratio because of the strict ban placed on old, inefficient vehicles that failed to meet the European No. I standards for exhaust emissions. However, control on power plant emissions would increase the CO/NO_x ratio as power plants are minor sources for CO. As a result, dCO/dNO_y enhancement ratio observed at Miyun did not change significantly from August 2007 to 2008, leading to comparable estimates on emission changes for CO and NO_x.

Researchers at Tsinghua University conducted a detailed bottom-up study of Beijing emissions during the Olympics period based on roadside traffic monitoring, emission measurements at the smoke stacks of selected power plants, statistics on industrial output reductions and plant closures, and other information on activity levels and emission factors (*Wang et al.*, submitted manuscript). Their estimated emission reductions were 58%, 51%, and 55% for SO_2, CO, and NO_x. Our estimate of the reductions is consistent with the bottom-up estimate for SO_2 but is lower for NO_x and CO. The discrepancy may be attributed to certain types of emissions not included in the bottom-up study, such as biofuel combustion in rural areas surrounding Beijing, biological emissions of NO_x from soils, and CO produced from decomposition of VOCs. These types of emissions, which are important sources for CO and NO_x but not for SO_2, are unlikely to have been impacted by measures taken to reduce emissions of pollutants during the Olympics. Allowing for their contributions in the bottom-up study, the estimated emission reductions for CO and NO_x would have been lower. Our 'top-down' estimates are based on observational data, accounting for the composite impact of all the emission sources.

The bottom-up study estimated that VOCs emissions in Beijing were reduced by 59% during the Olympics compared with August 2007. As VOCs species were not measured at Miyun, we adopted the bottom-up estimate in the model simulation discussed below. Emission reductions for other regions were taken from bottom-up estimates by researchers at Peking University (*S.Q. Zhang*, personal communications).

The top-down and bottom-up estimates of emissions of different species combined here are referred to as the Olympics emissions in what follows.

4. Model Analysis

In this section, we first evaluate the performance of the nested-grid GEOS-Chem in simulating the changes in O_3 and other species observed at Miyun during the Olympics. The model will evaluate the top-down emissions and simulate the impact of emission reductions on the regional atmosphere.

4.1 Model evaluation of emissions

Figure 8 presents day to day variations of afternoon mean mixing ratios of O_3, CO, NO_y, and SO_2 at Miyun in August 2008. Observations are indicated by the red lines. Significant day-to-day variations were observed for all species at Miyun, with higher pollution levels during the first five days of August. Since all emission reduction measures had been put in place before 1 August 2008, emissions were expected to stay relatively constant throughout August except on the opening day of the Olympic Games (8 August), which was declared a public holiday for Beijing. Emissions from the transportation and industry sectors were likely lower on that day. Observations at Miyun indicate that mixing ratios of O_3, CO, NO_y, and SO_2 were lower apparently on 8 August compared with the day before, although the impact is expected to have lasted for at most a couple days. Day-to-day variability in mixing ratios at Miyun presumably reflects changes due to variations in meteorological conditions and chemical lifetimes of relevant chemical species.

In Figure 8, model results computed using the standard emissions for 2007 are displayed in black while those using assumed Olympics emissions are displayed in blue. With the standard 2007 emissions, the model captures well the temporal variability of all four species observed at Miyun, with correlation coefficients (r) ranging from 0.56 to 0.8. This suggests that the day-to-day variations in individual species are driven primarily by changes in meteorology and chemistry, features that are accurately reproduced by the

model. However, absolute values of model results obtained using the standard emissions are significantly higher than observational results for all the species, confirming the benefit to air quality of the emission restrictions implemented during the Olympics. The biases between model and observation average are +25 ppbv (+41%) for O_3, +70 ppbv (+21%) for CO, 3.5 ppbv (+37%) for NO_y, and 2.7 ppbv (+113%) for SO_2.

Adopting the Olympics emissions in the model results in a significant decrease in the mixing ratios of O_3, CO, NO_y, and SO_2 simulated for Miyun (Figure 8), thus reducing the model bias by more than a factor of two for all species. After implementing the Olympics emission reductions, mean mixing ratios simulated by the model are reduced by 15% (or 12 ppb) for O_3, 24% (96 ppb) for CO, 36% (4.8 ppb) for NO_y, and 42% (2.1 ppb) for SO_2, compared with those based on the standard emissions, with no significant biases between model and observation for CO, NO_y, and SO_2. The resulting decreases in CO, NO_y and SO_2 mixing ratios simulated by the model correspond very well to the 'top-down' estimates of emission reductions derived in the previous section using Miyun observations, e.g., a 32% reduction in CO emissions from Beijing resulting in a 24% reduction in CO mixing ratios at Miyun as simulated by the model, providing an independent evaluation on the observation-based approach. The ability of the model to simulate day-to-day variations in observations is also improved by adopting the Olympics emissions. The correlation coefficients between model and observation for O_3, NO_y, and SO_2 increase to 0.86, 0.75, and 0.65 from their corresponding values of 0.81, 0.56, and 0.54 in the standard simulation. We conclude therefore that the use of the Olympics emissions significantly improves the performance of the model in simulating the observations at Miyun.

Model sensitivity analysis was conducted to evaluate the relative contribution to air quality improvements in Beijing during the Olympics of local versus regional emission restrictions. We found that 80% of the concentration decreases simulated at Miyun during the Olympics resulted from a reduction in emissions from Beijing, with regional emission reductions accounting for an additional 20% decrease.

4.2 Influences of meteorology and emissions

Since the model has been demonstrated to reproduce well the variability in meteorology and chemistry, model sensitivity analysis was conducted to quantify the extent to which meteorological conditions were responsible for the improvement in ozone air quality in Beijing during the Olympics. The model was spun up for three months from 1 March 2006, and results for 1 June 2006 were saved to provide the initial conditions for subsequent simulations. The model was run from June to August for each of the three years (2006-2008), all using the same initial conditions obtained from the spin-up for 1 June 2006 and using the standard emissions for 2007. The Olympics emissions described above were then adopted to drive the model simulation for August 2008 only. Hourly model outputs for the Augusts of the three years were used for analysis.

We define the daily O_3 anomaly in August 2008 as the deviation of afternoon-mean O_3 from its mean values in 2006 and 2007. The daily O_3 anomaly calculated from the model using the same standard emissions for August 2006-2008 can be thought of as representing the change in O_3 during the Olympics that is not related to emission restrictions, and we refer to it as the meteorology-driven anomaly. The O_3 anomaly derived from model results using the Olympics emissions for August 2008 is called the composite anomaly as the model in this case takes into account both meteorology and emissions specific to August 2008. The difference between the composite anomaly and the meteorology-driven anomaly is regarded to be the emission-driven anomaly. Figure 9a compares the observed daily O_3 anomaly at Miyun (solid line) with the composite anomaly simulated by the model (dashed line). The daily anomaly in both model and observation ranges from -60 ppb to 40 ppb. Positive anomalies were observed to occur frequently before 8 August, after which, negative anomalies prevailed. The model was found to reproduce well the daily O_3 anomaly observed at Miyun in August 2008: the correlation coefficient between model and observation is 0.83. The mean O_3 anomaly simulated by the model is -11.2 ppb, consistent with the mean of -12.7 ppb reflected in the observational data. With the model reproducing well the observed anomaly, we assume that the model can do a satisfactory job in distinguishing the meteorology-driven anomaly from the emission-driven anomaly, which cannot be separated in observational data.

Figure 9b presents the meteorology-driven anomaly (solid line) and the emission-driven anomaly (dashed line) predicted by the model. The meteorology-related anomaly has significant day-to-day variations, ranging from -40 ppb to +40 ppb. In contrast, the emission-driven anomaly is always negative, ranging from -20 ppb to -5 ppb, confirming the benefit of emission restrictions in reducing O_3 pollution over Beijing regardless of meteorological conditions. Compared with the meteorology-driven anomaly, the variability in the emission-driven anomaly is much smaller. The good temporal correlation between the meteorology-related and the composite anomaly indicates that the anomaly on individual days is mostly driven by meteorology. Table 2 summarizes mean anomalies for different days in August 2008. The large positive meteorology anomaly during the first week of August (+25 ppb) indicates that high O_3 levels during this period were largely meteorology driven, when the atmosphere was stagnant with weak southwesterly winds and high temperature. Although the emission-driven anomaly (-11 ppb) cannot fully compensate for the meteorology-driven anomaly during this period, it reduces the composite anomaly (+14 ppb) to 60% of the meteorology anomaly, suggesting the benefit of reducing emissions of O_3 precursors during polluted days. During the Olympics (8 August – 24 August), both the meteorology-driven and emission-driven anomalies are negative, averaging -12 ppb and -10 ppb respectively. The meteorology-driven anomaly appears to account for a slightly larger fraction (55%) of the composite anomaly than the emission-driven anomaly (45%) during this period. However, the difference between the two anomalies is only 2 ppb, within typical error bounds of chemical transport models for O_3 simulation. Averaged for the whole of August 2008, however, the mean emission-driven anomaly is -8.9 ppb, accounting for 80% of the composite anomaly and larger than the meteorology-driven anomaly (-2.3 ppb) by a factor of 4. We conclude that although the day-to-day variability in ozone is driven mostly by meteorology, the reduction in emissions of ozone precursors associated with the Olympic Games is responsible for at least half of the observed decrease in O_3 during August 2008.

4.3 Regional impact of emission reductions

Figure 10a shows monthly mean afternoon O_3 averaged over the planetary boundary layer (PBL; 0-2 km) simulated by the GEOS-Chem model using the standard emissions for August 2008. The model predicts that high ozone levels exceeding 70 ppbv are located over the North China Plain (32^o-40^oN, 110^o-120^oE). Ozone mixing ratios are relatively low in south China and northeastern China. Previous studies showed that the Beijing urban area was in a VOC-limited regime (*Chou et al.*, 2009), while the surrounding suburban and rural areas were NO_x-limited (*Wang et al.*, 2006). As a result of the combined effects of both NO_x and VOCs emission reductions during the Olympics, use of the Olympics emissions in the model decreases the simulated O_3 mixing ratios over Beijing and the surrounding regions. Figures 10b and 10c display the spatial distribution of the resulting reduction in O_3, averaged separately over the PBL and over the free troposphere (2-6 km) respectively. As expected, the largest reduction is found over Beijing, averaging about 12 ppbv for August 2008. Because of the control on regional emissions as well as the relatively long lifetime of O_3, the impact of the imposed reduction in emissions is found to extend far beyond the Beijing urban area, covering a large region over the North China Plain and Northeastern China. Both the magnitude and spatial extension of the simulated reductions are larger to the northeast of Beijing than to its southeast, reflecting the direction of the prevailing winds during this season. Within the PBL, the areas with mean O_3 reductions exceeding 4 ppbv extend northeastward from Beijing to about 45^o N in Jilin province and southeastward to about 37^o N in Hebei province. The reduction in the FT is about 50% less than in the PBL. The 2 ppb reduction isopleth in the FT extends northeastward from Beijing to about 45^o N in Jilin province and southeastward to about 37^o N in Hebei province.

5. Concluding Remarks

To improve air quality during the Olympics (8-24 August 2008) and the Paralympics (9-17 September 2008), a series of aggressive measures was implemented by the Chinese government to reduce pollutant emissions in Beijing and surrounding areas, in place for more than two months during the interval of the two Games. We

conclude that the emission restrictions were notably successful in improving air quality over Beijing. In August 2008, significant reductions in mixing ratios of O_3, CO, NO_y, and SO_2 were detected at Miyun, a rural site located 100 km downwind of the Beijing urban center, based on comparison with comparative data for August 2006-2007. The mean daytime mixing ratio of O_3 was reduced by about 15 ppbv in August 2008 from 65 ppbv in August 2006-2007, while daytime O_3 averaged only 45 ppbv during the time period of the Olympics (4 August – 24 August 2008). The decrease in O_3 was most significant in the afternoon when *in situ* photochemical production of O_3 is most active. The reduction in O_3 was reflected not only in mean but also in peak concentrations.

In August 2008, the relative reductions in daytime mixing ratios of SO_2, CO, and NO_y observed at Miyun amounted to 61%, 25%, and 21%, respectively as compared to the same month a year earlier. Concentrations of the three species are positively correlated at Miyun, indicating that they originate from co-located sources. While there is no significant change in the dCO/dNO_y enhancement ratio from August 2007 to 2008, the dCO/dSO_2 and dNO_y/dSO_2 enhancement ratios in August 2008 are significantly larger than values for August 2007, suggesting that the relative reduction of SO_2 emissions is much larger than that of CO and NO_x. Control strategies targeting the power sector reduced emissions of SO_2 and NOx, but most effectively for the former, resulting in increases in the NO_x/SO_2, CO/SO_2. and CO/NO_x ratios. Because of the strict ban placed on old, inefficient vehicles, the traffic restriction tended to decrease the CO/NO_x ratio, compensating for the effect of power sector emission control The changes in the enhancement ratios, after excluding the impact of the variability of SO_2 lifetime, indicate that the relative reductions in emissions of SO_2, CO, and NO_x in August 2008 correspond to 60%, 32%, and 36%, respectively, as compared with the same month the year earlier. Our 'top-down' estimate of the reductions is found to be consistent with the bottom-up estimate for SO_2 but is lower for CO and NO_x. The combination of the top-down and bottom-up estimates on emissions of different species is used to define the Olympics emissions used to drive the model simulation.

The nested-grid GEOS-Chem model with $0.5^o x 0.667^o$ horizontal resolution over China is found to reproduce well the day-to-day variations in O_3, CO, NO_y, and SO_2

observed at Miyun but significantly overestimates mixing ratios derived using the standard 2007 emissions. Adoption of the Olympics emission reconstruction in the model leads not only to significant reductions in model biases but also to improvements in the temporal correlations between model and observations. Analysis of meteorological conditions observed at Miyun for the three years covered by the present observations and interpretation of the observations using the model both suggest that the reduction in emissions of ozone precursors associated with the Olympic Games made a significant contribution to the observed decrease in O_3 during August 2008, accounting for 80% of the O_3 reduction for the month as a whole and 45% during the Olympics Period (8-24 August). Because of the controls on regional emissions as well as the relatively long lifetime of O_3, the model predicts that emission restrictions can affect O_3 far beyond the Beijing urban area, resulting in boundary layer O_3 reductions of 2-10 ppbv over a large region of the North China Plain and Northeastern China.

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Tables

Table 1 Mean afternoon mixing ratios (ppbv) of SO_2, CO, NO_y, and O_3 associated with SSW-SW-S winds observed at the Miyun site in August 2007 and 2008.

	August 2007	**August 2008**	**Reduction (%)**
SO_2	6.2	2.4	61.3
CO	468	352	24.8
NO_y	11.7	9.2	21.4
O_3	78	58	25.6

Table 2 Observed and modeled anomaly of afternoon O_3 (ppb) for August 2008 and its decomposition averaged for different days in August.

		August 1 - 28	**August 1-7**	**August 8-24**
Observed anomaly (composite)		-12.7	16	-26
Modeled anomaly and decomposition	**Composite**	-11.2	14	-22
	Meteorology-driven	-2.3	25	-12
	Emission-driven	-8.9	-11	-10

Figures

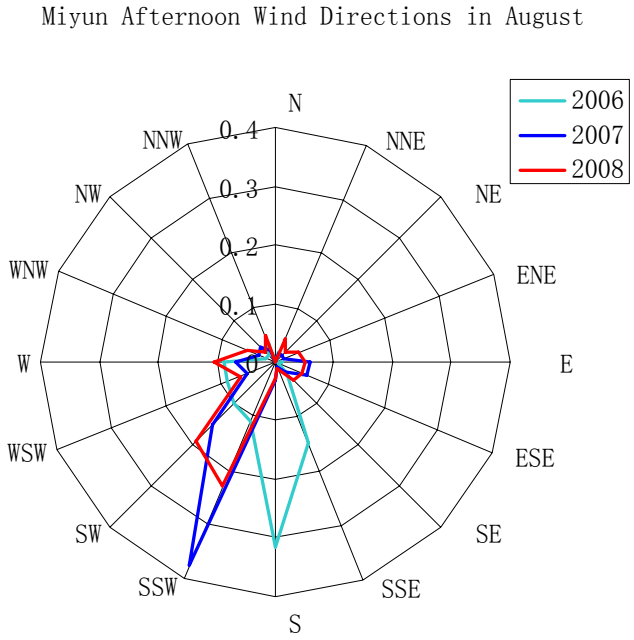

Figure 1. Wind roses of afternoon wind directions at Miyun in August 2006 (light blue), 2007 (blue), and 2008 (red). The radius indicates the frequency of wind observed in each direction.

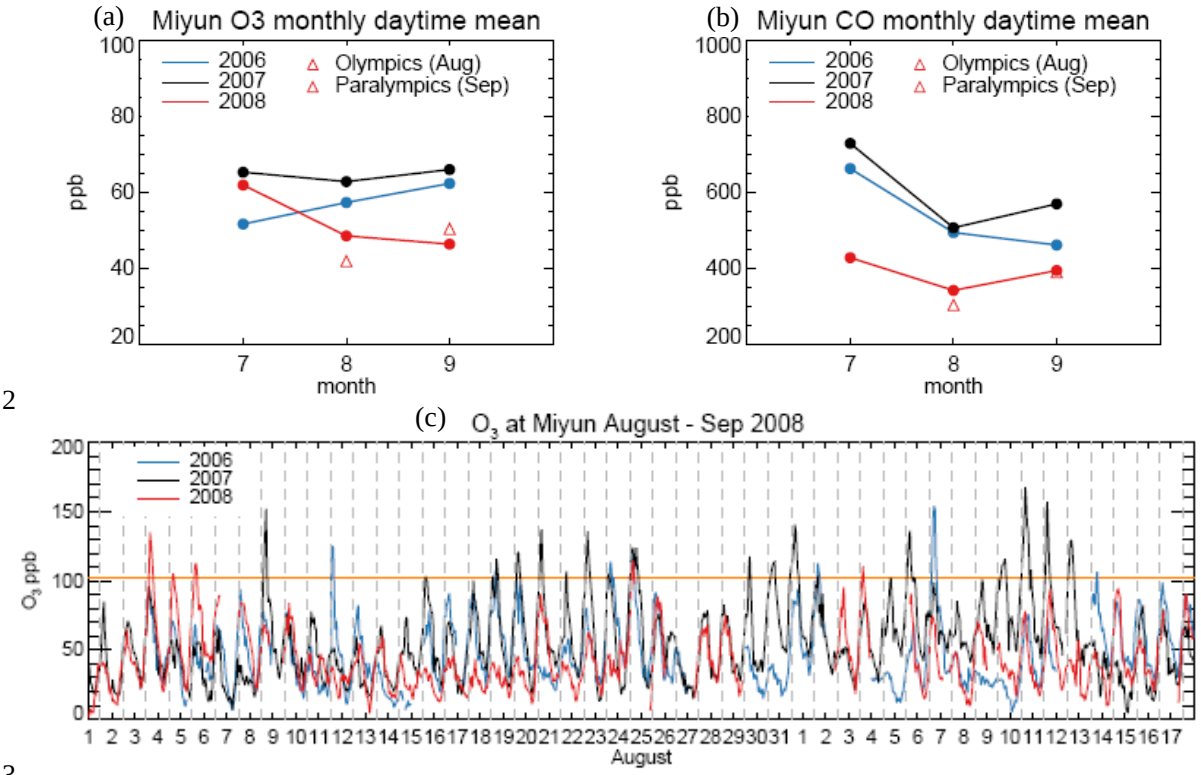

Figure 2. Daytime mean mixing ratios of O_3 (2a) and CO (2b) observed at Miyun for July-August-September of 2006 (blue line), 2007 (black line) and 2008 (red line). The triangles indicate the mixing ratios averaged for the exact time period of the Olympics Games (4-24 August 2008) and the Paralympics Games (6-17 September 2008). (2c) Hourly mixing ratio of O_3 observed at Miyun for the period 1 August – 17 September of 2006 (blue line), 2007 (black line), and 2008 (red line).

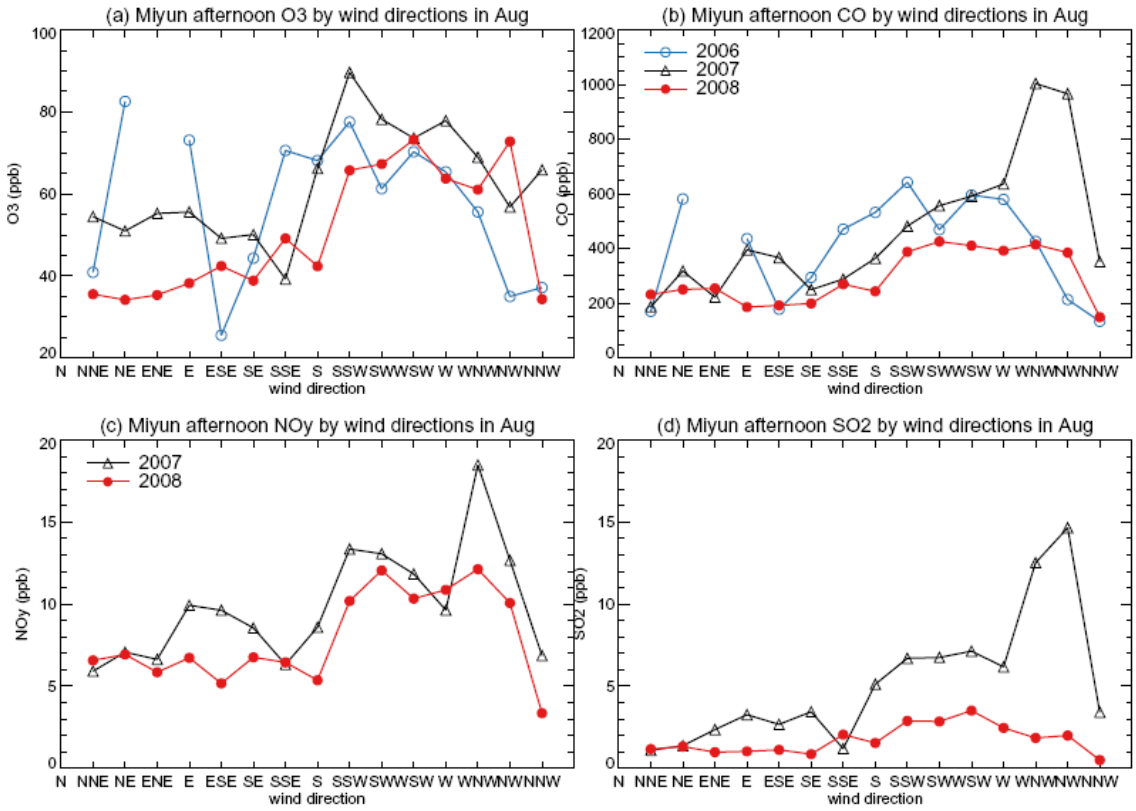

Figure 3. Afternoon mixing ratios of O_3 (3a), CO (3b), NO_y (3c), and SO_2 (3d) observed at Miyun in August as a function of wind directions. Data for August 2006 are displayed in blue, Aug 2007 in black, and Aug 2008 in red. Observations of NO_y and SO_2 were not available in 2006.

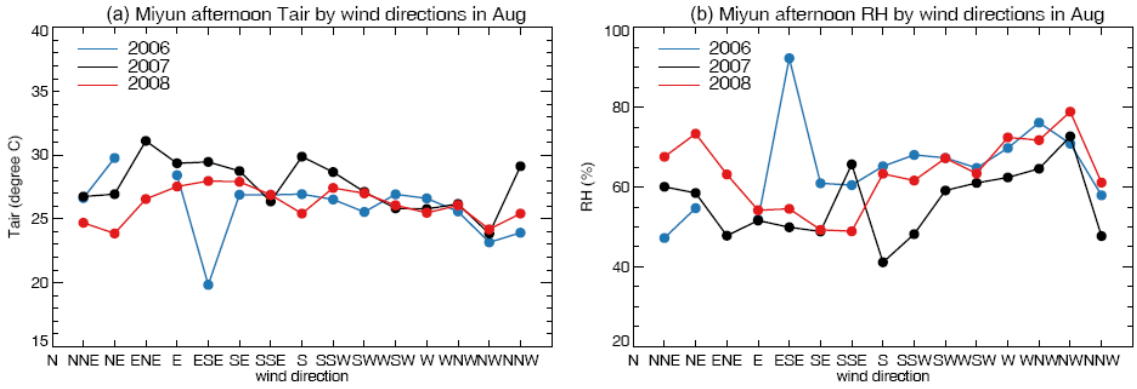

Figure 4. Air temperature and relative humidity observed at Miyun in August as a function of wind directions. Data for August 2006 are displayed in blue, Aug 2007 in black, and Aug 2008 in red.

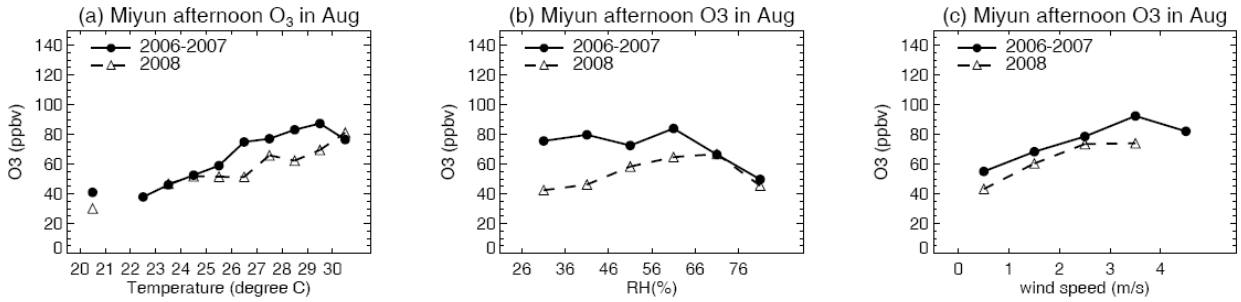

Figure 5. Mixing ratio of afternoon mean O_3 observed at Miyun in August as a function of temperature (a), RH (b), and wind speed (c). Data averaged for Augusts of 2006 and 2007 are indicated in solid black line, for August 2008 in dashed black line.

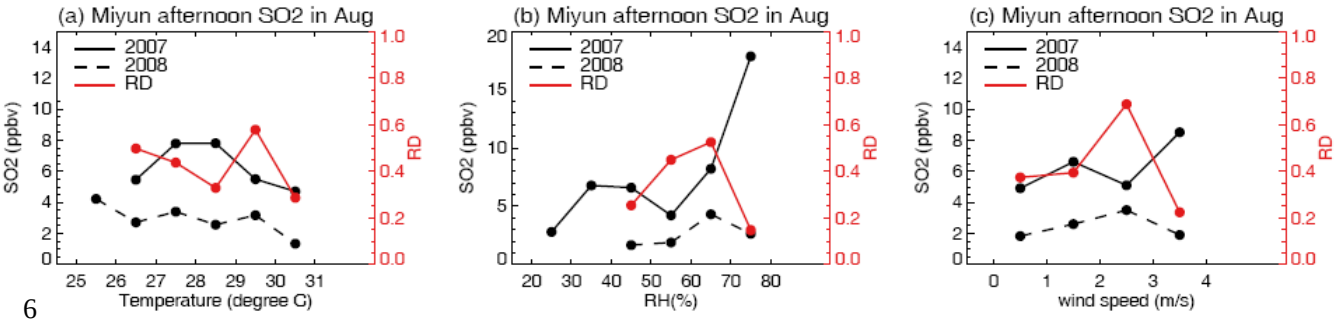

Figure 6. Mixing ratio of SO_2 observed in SW-SSW-S air masses at Miyun in August as a function of temperature (a), relative humidity (b), and wind speeds (c). Data for August 2007 are indicated in solid black line, August 2008 in dashed black line, and the relative difference (RD) between August 2008 and 2008 in red line.

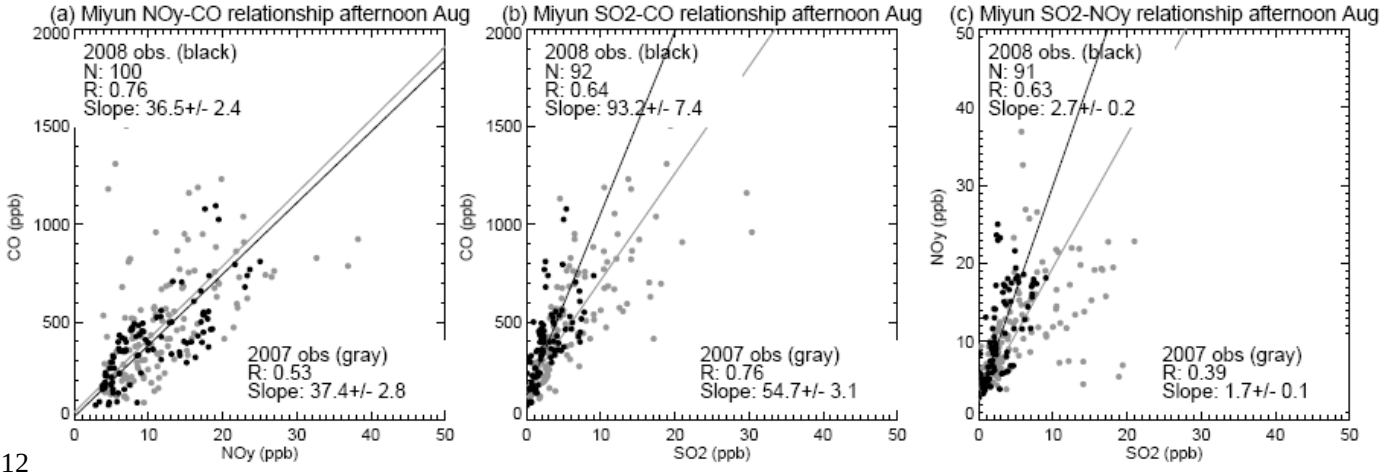

Figure 7. (a) NO_y-CO relationship in afternoon observations at Miyun in August 2007 (gray dots) and August 2008 (black dots). Each point refers to hourly mean mixing ratios. Correlation coefficients (R) and slopes of the reduced major-axis regression lines are shown in inset; (b) same as (a), but for SO_2-CO relationship; (c) same as (a), but for SO_2-NO_y relationship.

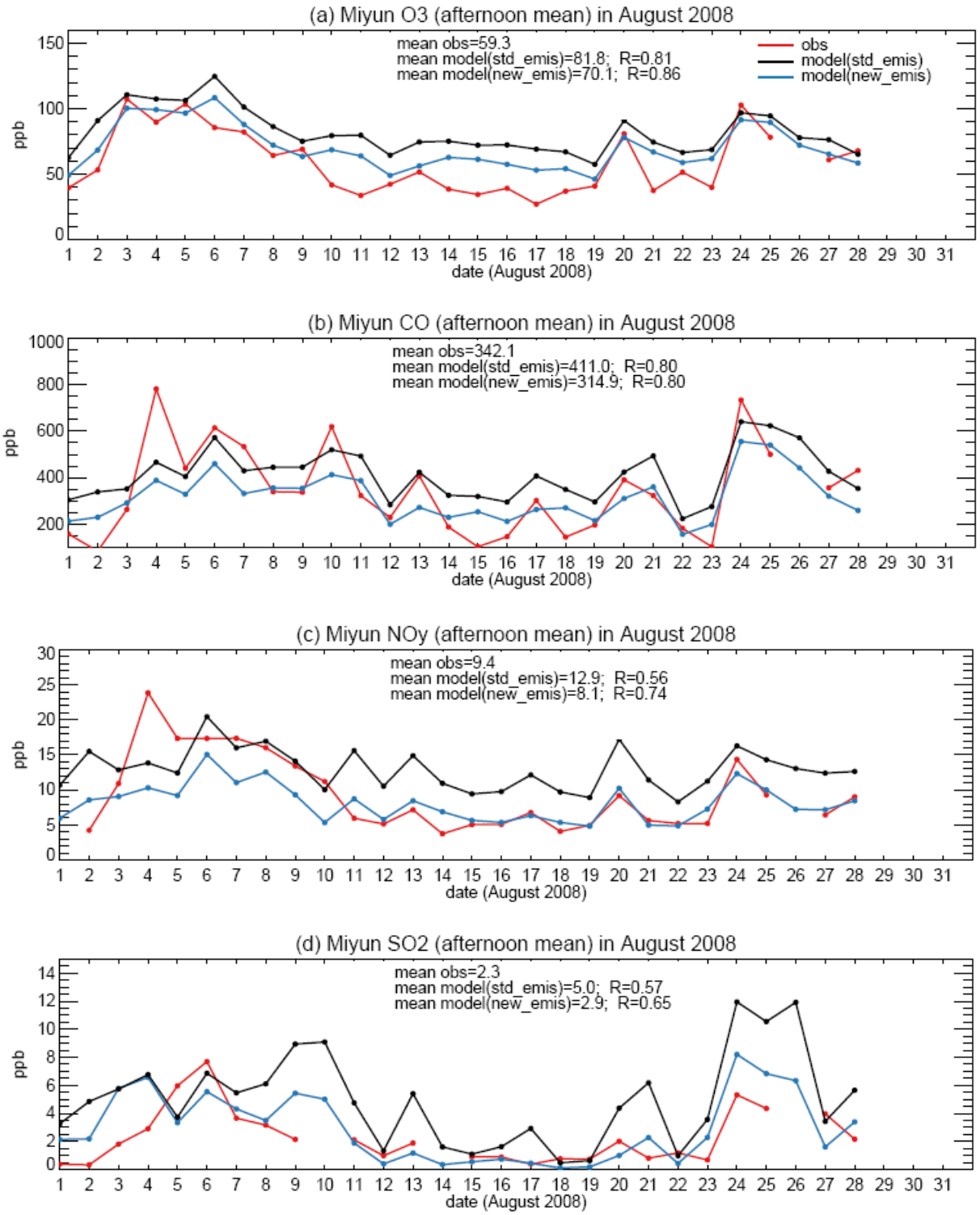

Figure 8. Day-to-day variations in O_3 (7a), CO (7b), NO_y (7c), and SO_2 (7d) at Miyun in August 2008. Observations are indicated in red, the GEOS-Chem model results using the standard emissions in black and the model results using the Olympics emissions in blue.

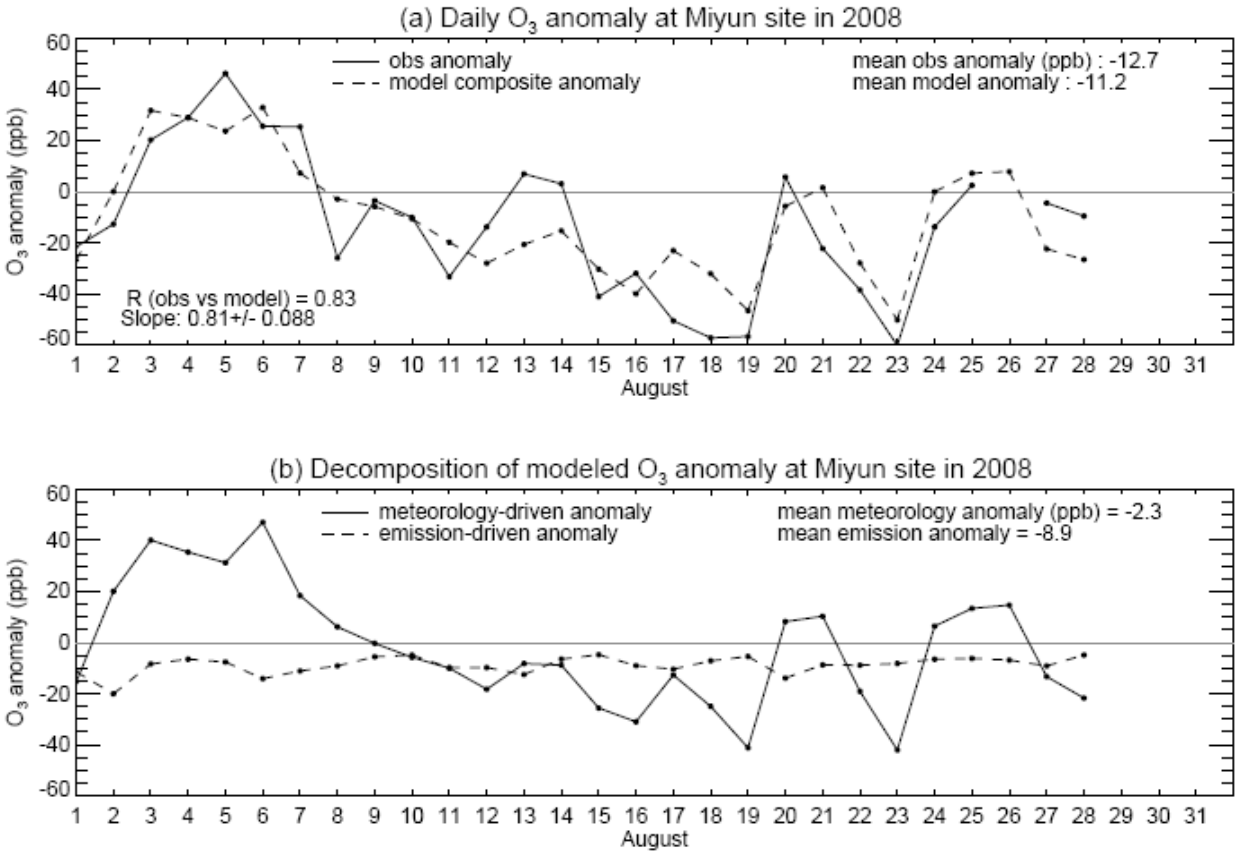

Figure 9. (a) Daily O_3 anomaly in August 2008 at Miyun. The observed anomaly is shown in solid lines and the modeled anomaly in dashed lines. The daily O_3 anomaly is defined as the deviation of afternoon mean O_3 in August 2008 from the mean values in August 2006 and 2007. (b) The meteorology-driven O_3 anomaly (solid line) and the emission-driven anomaly (dashed line) as simulated by the GEOS-Chem model.

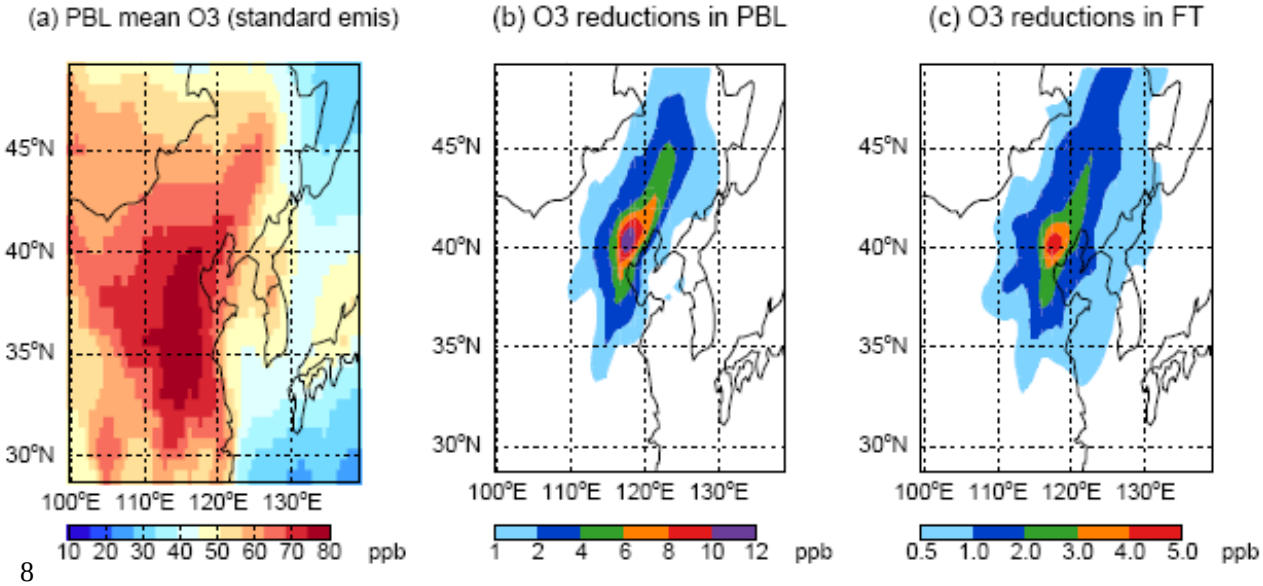

Figure 10. (a) Monthly mean afternoon O_3 averaged over the planetary boundary layer

(PBL; 0-2 km) simulated by the GEOS-Chem model using the standard emissions for August 2008; (b) Reductions in PBL O_3 indicated by model results using the Olympics emissions as compared with the standard emissions; (c) same as (b), but for O_3 reductions in the free troposphere (2-5 km).