

## ***Interactive comment on “Ozone air quality during the 2008 Beijing Olympics – effectiveness of emission restrictions” by Y. Wang et al.***

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We thank referee 2 for his/her helpful comments.

*1) The paper describes an O<sub>3</sub> control experiment that has numerous parallels to deliberate multiyear strategies and to weekend/weekday variations observed in many other cities. However, the paper does little to put its results in the context of these studies. From those studies we know quite a bit about the spatial extent of the combined and separate effects of VOC vs. NO<sub>x</sub> controls on O<sub>3</sub>.*

The VOC and NO<sub>x</sub> regimes of O<sub>3</sub> have been observed and studied in many cities in the US and other western countries. Murphy et al. papers are nice demonstration of using the weekend/weekday variations of precursor emissions and O<sub>3</sub> provides to

C2441

understand ozone chemistry for a US city. The emission reduction measures during the 2008 Olympics were formulated based on all the prior knowledge about O<sub>3</sub> chemistry derived from western studies, but it was largely unknown before the Olympics whether these costly emission control measures implemented in Beijing would be successful. After all, our basic scientific understanding about surface O<sub>3</sub> (concentrations, temporal and spatial variations etc), precursors emissions (NO<sub>x</sub> vs VOCs; anthropogenic vs biogenic, etc), and the role of meteorology in modulating O<sub>3</sub> in Chinese cities has been minimal compared with that for US cities.

Ideally, to study the impact of Olympics experiments on pollution, there would be a regional-scale comprehensive study which took measurements in different parts of the Beijing city and examined the changes and correlations in trace species concentrations in order to improve current understanding of O<sub>3</sub> chemistry in the region. The reviewer suggests in his/her later comments that we should look at other measurements in the city or some of his/her comments was based on the assumption that observations in other parts of the city were made readily available to our study. But unfortunately, this ideal case did not happen as everyone wishes in Beijing. Although several Chinese groups set up independent observational network and carried out in-depth analysis, sharing observational data between them were not efficient, or at least we did not have access to the data. Given the situation, our study uses long-term observations at a rural site downwind of Beijing and by comparing trace gas concentrations and meteorology before and during the Olympics in conjunction with thoughtful modeling exercise, the focus of our paper is to (1) investigate whether the costly emission control experiments during the Olympics were successful to reduce O<sub>3</sub>; (2) to partition the observed O<sub>3</sub> reduction between meteorology and emission reductions.

On weekend/weekday effect: For Chinese cities, it has been shown in a number of prior studies that weekend/weekday variations were not observed for precursor emissions such as NO<sub>x</sub> over Chinese cities (Beirle et al., ACP, 2003), likely due to different emission patterns related to social-economic factors in China. To our knowledge, there

C2442

is no study on the weekend effect on O<sub>3</sub> in Beijing or other Chinese studies so we cannot directly refer to western studies for VOC vs NO<sub>x</sub> controls on O<sub>3</sub> over Beijing. Brief investigation of our O<sub>3</sub> data at Miyun did not reveal any weekend/weekday effects. Our study examines the differences in monthly mean O<sub>3</sub> for Augusts of 2006-2008, not on the weekly scale. Thus we did not include any discussion of the weekend variation or absence of it in China, which would be a different topic worthy of in-depth analysis sufficient for another paper.

We have reorganized the introduction to elaborate the focus of our work (see revised paper). We added the following on weekend/weekday effect in the text:

(pg 3, line 19–pg 4, line 8) “The effect on ozone is an important research question as the dependence of O<sub>3</sub> production on NO<sub>x</sub> and VOCs is significantly different between the so-called NO<sub>x</sub>-limited regime and the hydrocarbon-limited regimes (Sillman et al., 1990). Many previous studies investigated the nonlinear O<sub>3</sub> chemistry and challenges of combating O<sub>3</sub> 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<sub>3</sub> and precursor emissions in Chinese cities has been minimal. For example, examining the day-of-week variations of O<sub>3</sub> 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<sub>3</sub>, CO, NO<sub>y</sub>, and SO<sub>2</sub> at a suburban/rural site (Wang et al., 2008b) located directly downwind of the Beijing urban

C2443

area during summer months.”

(pg 8, line 15-18) “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<sub>3</sub> observed at Miyun. Therefore, we did not distinguish weekend from weekday observations in the following discussion.”

*2) The authors begin by discussing non-linearities in the chemistry and then proceed in their observational and modeling analysis as if these non-linearities are non-existent and irrelevant. There are a number of papers in the literature suggesting that the spatial scale of the authors’ model is too coarse to capture the non-linear effects of chemistry on ozone. Murphy et al. 2006 and 2007 ACP and ACPD show an example of the nonlinear feedbacks on chemistry of other species. These ideas and prior results should inform the authors’ analysis of changes in ambient concentrations of SO<sub>2</sub>, NO<sub>y</sub>, CO and O<sub>3</sub>.*

We agree with the referee that we didn’t analyze the nonlinear ozone chemistry in the main part of the paper. As stated in the introduction, the focus of the paper is to demonstrate that the emission reduction measures during the Olympics were successful to reduce O<sub>3</sub> and to separate the relative roles of meteorology vs emission changes. We thus removed the two sentences in the introduction about nonlinear ozone chemistry.

Our model has a spatial resolution of 0.5 degree x 2/3 degree, roughly 40km x 50 km over China, typical for regional-scale models. We agree with the referee that our model’s scale is too coarse to resolve the heterogeneity in emissions and thus non-linearity in ozone chemistry at urban scales and we didn’t try to push our model to do that. Instead, we compare our model with observations made at a rural site (Miyun) 100 km downwind of Beijing urban region.

*3) The paper needs to more clearly define who/where the controls were designed to benefit and to show that Miyun is indeed representative of the chemistry at the location(s) that were the target. To this end, the paper should include a map of the region*

C2444

*with approximately 150-200km dimensions that clearly marks the boundaries of the region referred to as Beijing, indicates the major stationary sources and roads and points out the locations of the Olympic venues. Miyun is 100 km from Beijing according to the text. Based on prior analyses of the spatial extent of the weekend effect and on the NO<sub>x</sub> dependence of the sign of the weekend effect on O<sub>3</sub>, Miyun might reasonably be expected to have experienced lower ozone while the core of the Beijing metropolitan area experienced higher ozone. That scenario occurs in many other cities and has been described thoroughly in the literature.*

A map of the region was included in our prior paper in ACP (Wang et al., 2008b). We pointed the readers to this reference. In the paper, Beijing refers to the urban regions within the Sixth Ring Road (about 40km x 40km domain) where the Olympics venues are. The site is located in the Miyun County, which belongs to the Beijing metropolitan administrative area. There are no major source regions between Beijing and Miyun or close to the site in other directions. Our analysis of flow patterns showed that the Miyun site is downwind of Beijing in August, measuring therefore aged pollutions coming out of Beijing. We added more site descriptions in the text (pg 5, line 9-15):

“The Miyun site (40° 29'N, 116° 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.”

Ozone concentrations at Miyun are expected to be different from those around urban centers. Without access to urban O<sub>3</sub> data in Beijing, we cannot tell whether O<sub>3</sub> at Miyun would be lower than that over Beijing urban center as suggested by the reviewer. Titration of O<sub>3</sub> by fresh-emitted NO in urban areas (NO+O<sub>3</sub> → NO<sub>2</sub> + O<sub>2</sub>) could make urban O<sub>3</sub> lower than rural O<sub>3</sub>. As urban pollution transports downwind toward rural

C2445

areas and NO<sub>x</sub> levels becomes lower, O<sub>3</sub> production efficiency increases leading to more ozone production per unit of NO<sub>x</sub>. There are many papers in the literature on the differences in concentrations and chemistry between urban O<sub>3</sub> and rural O<sub>3</sub> downwind. This topic, however, it is outside the scope of our paper. The purpose of our study is to evaluate the effect of emission reductions on pollution levels during the Olympics through analysis of observations before and during the Olympics at a fix rural site sampling aged urban pollution from Beijing.

*4) The paper uses the word “effectiveness” to describe the response of ozone to the control strategies implemented, but it does not give a clear definition of the word. Presumably what we want to know is how much ozone was reduced below a dangerous threshold in locations where athletes and tourists were most likely to spend their time during the Olympics?*

As ozone is toxic, there is no known “safe” threshold of O<sub>3</sub> associated with negligible risk (Bell et al., Environ. Health Perspective, 2006). Because of the nonlinear ozone chemistry, reducing ozone precursors does not guarantee the reduction of ozone. So we use the word “effectiveness” to indicate that first, ozone levels were successfully reduced during the Olympics, and second the reduction were significant, averaging 15 ppb for monthly daytime mean in August 2008. We can define the “effectiveness” by using Chinese air quality standard (1-hr mean 102 ppb), as stated in the text: “The number of hours with 1-hr average concentrations of O<sub>3</sub> 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.”

*5) The paper would be much more convincing if it used the same methods to pursue an analysis of a control situation without implantation of control strategies. For example, May, June or October 2008 could be analyzed in parallel with August to show how the accuracy of the approach for months where no controls were implemented.*

O<sub>3</sub> and its precursors have natural seasonal cycles due to lifetime changes with sea-

C2446

son. In our prior paper [Wang et al., 2008b], we showed that in 2006, mean O<sub>3</sub> in June at Miyun were 20 ppb higher than that in August. We compare the same months of different years to minimize the seasonal effects. We already stated this explicitly in the text (pg 8, line 9-15):

“We showed in a previous study that O<sub>3</sub> peaks in June at Miyun and that mean daytime O<sub>3</sub> in August is on average 10 ppbv lower than that in June (Wang et al., 2008b). Other observations for Beijing have shown similar seasonal patterns in surface O<sub>3</sub> (Ding et al., 2007; Lin et al., 2008). To minimize the compounding effects of this natural seasonal variability of O<sub>3</sub> 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.”

*Other issues to be addressed: CO is a nice tracer of anthropogenic VOC reactivity but not of total VOC reactivity. The paper should describe the fraction of reactivity that is biogenic. A 15 ppb decrease in O<sub>3</sub> from meteorological factors alone is unusual. If there is precedent for this please cite. Could the decrease instead be due to biogenic emissions?*

We did not make any attempt in the paper to link CO emissions with VOC sources or VOC reactivity. Estimates on VOCs emission reductions during the Olympics were adopted from a bottom-up study, not scaled from CO.

We do not know which part of the text the reviewer refers to when commenting on 15 ppb decrease of O<sub>3</sub> due to meteorology. 15 ppb change in O<sub>3</sub> is not unusual for day to day variability of O<sub>3</sub> at our site (Wang et al., 2008b), and without manmade emission changes, the day-to-day variation of O<sub>3</sub> should be largely attributed to meteorology. We found that the O<sub>3</sub> levels for NNE-NE-ENE winds were 15 ppb lower in August 2008 than 2007 and attributed this to meteorology. As the NNE-NE-ENE flows were sampled only on a couple of days in August of the two years, as we show in the wind rose, the 15 ppb decrease in O<sub>3</sub> relates to day-to-day variations in meteorology. We've clarified

C2447

this in the text (pg 11, line 5-8):

“winds from NNE-NE-ENE sector are infrequent at the site in August, and lower O<sub>3</sub> 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<sub>3</sub> for August 2008.”

*Since O<sub>x</sub> is so strongly affected by temperature, it would be useful to compare the correlation of O<sub>3</sub> with temperature in the different years to show that the slope is indeed different with the controls. Perhaps using that as a metric would allow the authors to incorporate ozone data from other sites in the study region.*

We showed in the text (Section 3.2) that there was less than 2 degree differences in mean temperature between Augusts of 2007 and 2008, while the O<sub>3</sub> levels differ by more than 15 ppb. As a result, the correlation slope between O<sub>3</sub> and temperature is expected to be very different with the controls.

The paper does not include data from other sites in the study region.

*Explain why RH is relevant to the analysis. Is it because of increases in O<sub>1D</sub> + H<sub>2</sub>O or as an indicator of stagnation or some other physically based reason.*

We added the explanation of RH in the text (pg 10, line 9-18):

“RH is an indicator of water vapor content in the air with respect to saturation levels. Reaction of O (1D) with H<sub>2</sub>O is the dominant pathway for chemical losses of O<sub>3</sub> in the troposphere ((1D) + H<sub>2</sub>O → OH, with subsequent reactions OH + O<sub>3</sub> → HO<sub>2</sub> + O<sub>2</sub> and HO<sub>2</sub> + O<sub>3</sub> → OH + 2O<sub>2</sub>). 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<sub>3</sub> (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<sub>3</sub> in summer 2006 at the Miyun site.”

C2448

*Non-linear chemistry will affect the location within the plume where HNO<sub>3</sub> is formed resulting in changes in the fraction of NO<sub>y</sub> deposited during transit from the source. This may or may not be important in the year to year changes reported.*

We do not have measurements for individual NO<sub>y</sub> species at the site, so cannot analyze this issue. But our measurements of CO, SO<sub>2</sub>, and O<sub>3</sub> all showed coherent year-to-year changes as NO<sub>y</sub>.

*The analysis does not appear to be accurate enough to prove or disprove sources of variability at the 20% level. I recommend omitting the claim that regional reductions are shown to be responsible for any ozone changes.*

The 20% reduction on O<sub>3</sub> from regional emission reductions was derived from model sensitivity simulations. When we adopted emission reductions for both Beijing and surrounding provinces, the O<sub>3</sub> level at Miyun in August 2008 was reduced by 10 ppbv in the model. In another scenario when we adopted emission reductions only from the surrounding provinces (i.e., no emission reduction over Beijing), we found only 2 ppb decrease in O<sub>3</sub> at Miyun. Thus we concluded that regional emission reductions were responsible for 20% reduction of O<sub>3</sub> during the Olympics.

*The non-linear effect of the emissions reductions should be most strongly seen in SO<sub>2</sub> and may explain the apparent difference between its emissions as inferred from the observations and as reported in the inventory.*

SO<sub>2</sub> emission reductions inferred from the observation-based 'top-down' method is 60%, and that reported from the bottom-up inventory is 58%. They are very close to each other, lending support for our top-down method.

*If the paper is to argue that ozone is reduced everywhere in the domain, it should at a minimum show using a box model, that O<sub>3</sub> at both Miyun and the center of Beijing is in a NO<sub>x</sub> limited regime or that the VOC reductions are so large as to overwhelm the enhanced ozone production if the photochemistry is in a VOC limited regime.*

C2449

Previous studies showed that Beijing urban area is VOC-limited [Chou et al., JGR, 2009] and surrounding suburban and rural areas are NO<sub>x</sub>-limited [Wang et al., 2006]. Both NO<sub>x</sub> and VOCs emissions were significantly reduced during the Olympics. In our model calculation of the regional impact of emission changes, we adopted 36

“Previous studies showed that Beijing urban area was in a VOC-limited regime (Chou et al., 2009), while the surrounding suburban and rural areas were NO<sub>x</sub>-limited (Wang et al., 2006). As a result of the combined effects of both NO<sub>x</sub> and VOCs emission reductions during the Olympics, use of the Olympics emissions in the model decreases the simulated O<sub>3</sub> mixing ratios over Beijing and the surrounding regions.”

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C2450