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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air quality during the 2008 Beijing Olympics: secondary pollutants and regional impact" *by* T. Wang et al.

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Reply to anonymous referee #3' comments

"The manuscript "Air quality during the 2008 Beijing Olympics: secondary pollutants and regional impact" by Wang et al. describes and interprets measurements obtained from three ground sites near Beijing. These observations provide insight into the efficacy of drastic pollution control measures and are of great interest to the scientific community. Extracting the changes in secondary pollutants that can be attributed to the control measures implemented in Beijing is extremely challenging. The influence of urban emissions must be isolated from meteorology and the regional background. The authors correctly note the importance of meteorology and regional influences upon pollution levels, but I do not think these influences are adequately considered. This pa-

per is very broad and far-reaching, and I think that some of the conclusions require improved quantitative analysis to be justified. Specific recommendations are given below."

Reply: As the referee pointed out, it is indeed very difficult to isolate the effects of local emission, regional sources and changes in meteorology. Our measurement data clearly illustrate the impact of meteorology and regional sources, and we attempted to use the observations to estimate such impacts (for ozone and CO) by comparing the data from the upwind and urban sites. Comparing the data from 2005 and 2008 at a downwind site gives evidence for increasing regional emissions in the past three years. These initial results from our measurements clearly point out the importance in regional sources and meteorology. We agree with the referee that more quantitative analysis will be needed (as we have indicated in manuscript), we will modify relevant sentences such that the conclusions are directly/better supported by the analysis of our data.

The responses to the specific recommendations are listed below.

(1) Section 2.2: Does the NOy detector include particulate nitrate? NOy is never defined. This becomes important later. This section ought to at least mention detection limits and time resolution. I assume that all the measurements are above detection limits, but this needs to be stated explicitly.

Reply: the NOy detector does include particulate nitrate. We will add the definition of NOy in the revised manuscript. (NOy=NO+NO2+HONO+NO3+PAN+HNO3+NO3 +HO2NO2+N2O5+other organic nitrates...). The detailed description of the detection limit of NOy (and other instruments) has been given in our previous publications (e.g, Wang et al., 2001). The ambient NOy (and other pollutants) concentrations in three sites are above the detection limit of the instrument(s). We will mention this in the method section.

(2) Section 3: Results and discussions are mixed together, and it is difficult to follow.

The results ought to be stated first, and then a separate section should discuss the findings. Section 3.1: The results section begins with a discussion of API. But the API is for PM10, which isn't even measured at these ground sites. Furthermore, the averaging time for the API isn't given, so I'm not sure how the hourly data and daytime data shown should be compared to API. I think this discussion of API ought to be moved to either the introduction or to the end of the paper, if it is used at all. I'm not sure where the measurements shown in Fig 3 come from – this should be stated clearly in text and figure caption. I don't understand the line that states most of NOy is oxidation products of NOx. It appears to be the other way around in figures 3 and 4. From those figures, NOx looks to be about 2/3 NOy. But again, NOy isn't defined, and I'm not sure if those figures can be compared. The changes in secondary pollutants are stated without uncertainties. It appears that the standard deviations of the mixing ratios are quite large, and would easily encompass these changes. The statistical significance of the changes should be discussed. Additionally, dilution and mixing are never mentioned. Everything is stated in terms of mixing ratios. Are there any ratios to conserved species that could be used to eliminate the possibility that the changes are caused by mixing? Perhaps CO/NOx ratios could indicate changes in the automobile emissions?

Reply: This paper present the first results from our observations at the three sites, and covering four somewhat different topics (i.e., apparent relationship of secondary pollutants and weather conditions in the three periods in summer 2008, regional contribution to ozone (and CO), ozone production efficiencies, and comparison of 2005 and 2008 results at the downwind site.) For each topic we first presented the data and went on discussing the result. It would be difficult to use an alternative approach (i.e., present the results altogether with discussions followed). Thus we would like to keep the current structure. It seems that the problem is mainly in Section 3.1, We will improve the logic flow and re-organize the material in that section.

We used the API to show the officially reported air quality at the Olympic site and to

illustrate that it does not properly represent ozone pollution. The API is based on the averages of 24-hr concentrations from noon of the present day to noon of the preceding day. It would be difficult to change current position of API in the paper, but we will reorganize that section to clearly indicate our intention for using API there.

Figure 3 is based on measurements at the urban site (CRAES), we will add this info. in the text and figure caption.

While NOx was 2/3 of NOy in period 1, it accounted for only 1/3 of NOy in period 2 when chemically processed regional air masses arrived at the site. We will modify the sentence.

The comments on the need to add uncertainty in Fig. 3 and 4 have also been raised by another reviewer. In the revised manuscript, the test of statistical significance will be given when comparing the differences in pollutants shown in Fig. 3 and 4. As indicated in the previously posted reply to referee 1 and 2, in spite of the apparent large standard deviations, the diffidence between the mean values for ozone, sulfate, nitrate, NOy, and NOx are significant at $P<0.01$ ($P<0.05$ for ozone). For the VOCs shown in Fig. 4, the significance levels in the differences are lower ($P>0.05$) due in part to the much sampler number of samples. But considering the measurements from other researchers and result from the emission inventories, we think the apparent decreases in the VOCs after control at our site are real. This will be added in the revised paper.

Dilution (both horizontally and vertically) can affect the ambient concentrations of pollutants, as an impact of meteorology. But we believe the variations in pollution levels in the three periods were mainly due to changes in air flow and rainfall, as clearly shown in the presented data. We did not have continuously measured conservative tracer that was not affected by emission control measures. CO/NOx in the rush hours at CRAES indicated a higher ratio in period 2 (43 ppbv/ppbv) compared to period 1 (26) and 3 (37). The apparent higher ratio in period 2 is consistent with the dominance of regional pollution which has different CO/NOx.

(3) Section 3.2: The regional contribution is assessed by comparing ozone upwind and downwind of Beijing in Figure 7. But what about ozone precursors? There is still plenty of NOx upwind of Beijing, so the upwind contribution is underestimated by ignoring further ozone formation. Also, there may be significant NO2 levels at all of the sites. Failure to account for this would underestimate ozone contributions from each site.

Reply: We agree. We will add discussion to point out that the estimate may have underestimated regional contribution to ozone (due to additional production of ozone).

(4) Section 3.3: The OPEs are listed without uncertainty, and I am not convinced there is a significant difference between any of the values. Nitrate is a sizable fraction of NOy. Does NOz account for nitrate? The similarity between the urban and downwind sites is striking, as the authors correctly note. Wind speeds are mentioned only once, where in section 3.4 they are stated to be below 1 m/s. With such low wind speeds, it could be possible that the air simply sloshes around between the urban and downwind site, which explains why they look the same. Further discussion of the wind speeds is necessary to accurately interpret the data.

Reply: The uncertainty for OPEs will be added, and the result has confirmed that the differences in our comparisons are significant. NOz includes nitrate.

The surface wind speeds at the mountain site were always low due to the influence of topography. Surface winds at flat land site in Beijing and the back trajectories indicate some movement of air during episodes; with speeds of 2-4 m/s (see Figure 1). We will add this info.

(5) Section 3.4: By comparing measurements from different years at a site downwind of Beijing, the comparisons include changes in meteorology, urban emissions, and regional emissions. The authors acknowledge that these 3 are tangled together, but still try to separate the regional influence. Wouldn't it be easier to examine data from a site unaffected by Beijing to examine the regional background? I think the conclusions reached in section 3.4 would require an entire paper to justify.

Reply: The discussions in section 3.4 attempted to explain why some species have decreased but others increased from 2005 to 2008. After considering the three factors, the most likely reason is the increases in regional emissions between the two periods. We agree that addressing regional emission changes during 2005-2008 would better use concurrent upwind and urban measurements, which we do not have. Nonetheless results from section 3.2 and 3.4 do point out the important role of regional sources in secondary pollutants during the Olympics and this section provides evidence of increase in regional emission between 2005 and 2008. We will modify the conclusion in Section 3.4, as follows.

"The increased regional emission during 2005-2008 inferred from the above analysis and the regional contribution to the very high concentrations of secondary air pollutants after the drastic control measures in the summer of 2008 suggest that more stringent control of the regional emissions will be needed in order to significantly improve the air quality (especially ozone and secondary aerosols) in Beijing and the surrounding regions."

(6) Figures 1: It is very hard to see the symbols on the right panel. They should be made larger and in different colors. Also, could the scale be changed to include XCC and HSZ? Fig 2: c) should be split into c), d), and e). The axes need to be labeled for the last time series. And the averaging time should be stated for panel c. Also, is the wind direction an average over 4 hours, or an instantaneous measurement taken once every 4 hours? Fig 3: Where were these data obtained? Date format on fig should be changed to be consistent with the rest of the paper. Fig 5: Put markers to indicate HSZ, CRAES, and XCC on map. Fig 6: what is red star in b)? Include markers as in Fig 5. Fig 7: Is NO2 large? Does O3 titration cause the difference between HSZ and CRAES in the late afternoon? Fig 8: The upwind site is very polluted. The authors note this, but I think the source of the pollution ought to be identified as much as possible. Is it from the previous day's Beijing emissions? Is it all local?

Reply: The suggested improvements on figures will be adopted, except that the scale

in the right panel of Fig. 1 will be not changed as it would otherwise be difficult to see the details of the urban areas. The wind data were instantaneously taken every 4 hours. The data in Fig. 3 are from CRAES. The red star in Fig. 6 indicates the center of Beijing, and we will remove it since the markers for the three sites will be added. The average NO2 at CRAES in late afternoon (1600-1700) during the eight high-ozone days was 9-12 ppbv, which is 9-10 ppbv higher than that of the downwind HSZ site (2-3 ppbv), so the difference in ozone ($\sim$20 ppbv higher at HSZ) should not be due solely to titration. Upwind sources causing high concentrations of CO are emissions from cities, townships, and agriculture burning in the flatland areas of the North China plains. We will include this info. Finally, we would like to point out that the present paper gives an overview of the first results; many interesting and specific features of the data at each site (which the referee has mentioned in several places) are being examined in on-going effort, and we will present these results in separate publications soon.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 12433, 2010.