

## ***Interactive comment on “Use of a mobile laboratory to evaluate changes in on-road air pollutants during the Beijing 2008 Summer Olympics” by M. Wang et al.***

**Anonymous Referee #2**

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General Comments: This article describes measurements of a subset of generally traffic related aerosols and chemicals in the air made by a newly constructed mobile laboratory travelling in and around Beijing before, during, and after this city hosted the 2007 Summer Olympics and Paralympics. The extensive pollution control measures implemented by the Chinese government during this time might be considered one of the largest scale experiments in atmospheric chemistry conducted in recent times. The question as to what effect these pollution control measures ultimately had on measured air quality is highly appropriate for discussion in Atmospheric Chemistry and Physics, throughout the scientific community, and hopefully beyond. Within this framework, Wang et al presented a reasonably readable first draft. Due to generally

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missing information and for reasons of clarity, both grammatical and scientific, several more drafts are recommended before it is resubmitted for publication.

### Specific Comments:

1. More attention should be paid to the explanation of what pollution control measures were implemented, the goal of each measure, and the timing of implementation as it is central to most of the rest of the discussion. Merging of information from table 2 and table a1 as well as a translated version of the Chinese web-site documenting these measures would be very useful. Use of time divisions as set forth in table a1 might be rethought in favor of using the simple designation of before, during and after Olympics.
2. The overall description of inlets, instrumentation, and their performance needs a great deal of clarification and additional characterization. Without such work, it is difficult to assess the merit of these measurements.
3. The authors repeatedly imply that only a mobile laboratory is capable of assessing temporal and spatial variability. A good network of stationary measurements should also capture temporal and spatial variability. In fact, measurements from such a network of stations would be extremely helpful in showing when the mobile laboratory measured emissions from the vehicles directly surrounding it and when the measurements were more heavily affected by regional conditions. Such measurements would also help to show that their mobile measurements indeed reflect either temporal or spatial variability in a quantity that might otherwise be conserved over the extent of the study time and area.
4. A number of correlations were put forth with relatively little to justify their presence in the discussion. What am I to draw from tables 3, 4, and 5? The claim that the benzene/toluene ratio changed due to the cessation of painting activity, while possible, is not very well justified. Did somebody keep track of painting activity before, during and after the games? Fugitive solvent emissions are also claimed to be a source but somehow this is no longer considered a factor. Why?

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Technical Comments:

1. The authors wish to distinguish between larger particulate matter in the air and individual chemicals in the air and often use "aerosol phase" and "gas phase" to make this distinction. It might be more accurate to say aerosols and chemicals suspended in the air or something to this effect as they are both technically suspended in the "gas phase".

2. A large number of semicolons are used to separate ideas in sentences throughout this paper. Although their use is not necessarily grammatically forbidden, it does tend to produce very long sentences that can be difficult to understand.

12858 Line 3-4: "...innovative..." The word innovate implies something never before seen or that marks a significant departure from status quo. As the authors cite many examples of other mobile laboratories and do not clarify how their own laboratory marks a significant departure from previous efforts, a word such as versatile would be more appropriate.

12858 Line 9: "...ethyl benzene, m-, p-, and o-xylene...". It would be more accurate to say "the sum of ethylbenzene, m-, p-, and o-xylene" as the PTR-MS on board is unable to distinguish the individual isomers. Attention should be given to this distinction throughout as it is often misused.

12858 Line 12: "...Concentrations increased again after the control period ended..." Is this in comparison to the control period? Specify what increased with respect to what.

12858 Line 10-16: What is the normal variation in the concentrations of these chemicals over the time period in question for other years and at other times of the day?

12859 Line 13: The reference to Rogers 2006 here seems misplaced. Doesn't this have to do with measurements in Mexico City rather than Beijing?

12859 Line 14: "...formations..." Should be "...formation..."

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12859 Line 19: Is the citation for Hao correct here? The previous reference by Han has to do with cars up until 2020. Does Hao also fortuitously use the year 2020?

12859 Line 21: "...on-road automobiles in Beijing emitted..." Presumably these automobiles still emit now. Include the year/years from this study in which automobiles emitted.

12859 Line 22: "...Furthermore, chemical mass balance modeling with VOC observations during 2002-2003 showed that automobile exhaust was responsible..." Change to: "Chemical mass balance modeling using VOC observations made between 2002 and 2003 showed..."

12860 Line 4: What are the Euro IV standards?

12860 Line 7: "...the web site <http://www.bjepb.gov.cn/bjhb/tabcid/68InfoID/15395/ftid/40Defa> The listed link is, as one might expect, in Chinese. As APC is an English language publication, it would be useful to distill the contents of this web site into a table/time line in English and to tell the user ahead of time that the provided link is to a Chinese language web site. Although tables 2 and A1 purportedly do this, it would be helpful to combine them such that the time divisions made by the authors are more easily cross referenced with respect to the regulations imposed and their date of imposition. The divisions of table A1 don't seem to add very much to the simple before, during and after- analysis.

12860 Line15: "...in situ rapid response..." Add a comma: " in situ, rapid response..."

12860 Line16: "...of traffic emissions; such data cannot be obtained by stationary monitoring sites." Rework the second half of this statement which is misleading and get rid of the semicolon. It is possible to obtain data like that reported using identically equipped stationary monitoring sites spread densely over the study area. Since this infrastructure does not exist, a mobile laboratory is one alternative to obtaining the information. One might write something like: "Such data cannot be obtained by currently

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existing and equipped stationary monitoring sites."

12860 Line16: "Rarely has the use of a mobile laboratory for temporal and spatial analysis been reported." Is there a reason for this? Maybe it is difficult to link changes in measured parameters unambiguously to either temporal variations in the study area, spatial variations in the study area, or very local emissions without having additional information from stationary measurement sites or other mobile labs simultaneously in motion.

12860 Line26: "...in situ, on-road..."

12860 Line 27:"...aerosol phase..." Remove the word "phase".

12861 Line 15: "...were deployed..." The word "deployed" should be used in conjunction with some indication of what was deployed where. This makes it sound as if the UPS system was only sometimes built into the van and at other times, only the generators were built into the van.

12861 Line 21: "The gaseous pollutant inlet..." How long was it? What was the air flow through it. What is the typical residence time of a chemical prior to reaching instrumentation. Why was there a glass manifold? Where was the glass manifold (in the van I presume)? Was there a pump attached? If residence time was variable and depended on the speed of the platform, how were line-losses characterized as a function of the speed of the vehicle? How was the residence time reported on 12862 Line 8 derived? Did you inject gaseous/aerosol standards and watch to see how long it took to detect them while travelling at speed? Did you simply calculate based on the flow rate of gases you believe to be passing through the tubes? If the flow through the lines is variable, it is highly advisable to study particle/gas/chemical losses in these lines as a function of the speed of the van. Why is no such study reported?

12861 Line 25: "Airflows containing particles were forced..." How were they "forced" to enter the cone? Later, there are two inner diameters listed. Exactly which part is being

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described here? Figure 1 leaves a lot to the imagination. How do you isokinetically feed a flow from the cone shaped part into the other section. What is the advantage to having an inlet that requires you to drive at 60 km per hour? It seems to me that being in traffic requires constant adjustment of speed to safely navigate streets and this limits your "versatile" laboratory to only those roads on which it is possible to travel at this speed. Does this mean that only the data taken at this speed is valid? If speed is such an important factor, why don't you report a full characterization of your inlets at a variety of speeds? Why isn't there a plot of average speed for the many measurement trips reported in this work?

12862 Line 12: "..were mainly research-grade commercial instruments..." The words "mainly research grade" are meaningless. Leave them out or clarify exactly what it is about each instrument that makes it suitable for research and others not. Do you simply wish to make the distinction between home-built instrumentation and commercial instrumentation?

12862 Line 13: "...with an emphasis on high time resolutions." Reword: "..with preference for instruments having high time resolution.

12862 Lines 14-18: Is this referring to all trace gas measurement devices? If so, the PTRMS should also be listed here. You go on to describe the working principle of the PTRMS but don't describe the working principles of the instruments used to measure NOx, CO, and SO2. Why not? The reference Fortner 2009 does not belong in a list of references describing the operational principles of a PTRMS. Leave it out in line 24.

Line 24: "...xylene..." This should be the sum of the xylene isomers and ethylbenzene.

Line 27: Why is Fortner 2009 cited here? To the best of my knowledge they don't provide an in depth description of catalytic converter design which is the information that a user would be expecting in the reference cited at this point. Any comment as to the reliability of background determination in PTRMS instruments? What about identification of the signals related to benzene, toluene, and xylenes etc. and the chemical itself. A

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longer discussion is given in Rogers 2006 but none here.

12863 Line 1-5: What is an EPA TO-15 standard? From a cursory look at the EPA document itself, it seems that it presents methods for canister analysis of air samples rather than a method of producing standards. Were the standards used for this work collected in canisters? Did you collect the canisters yourselves? Where did you send them for analysis? Why is this paragraph grouped under instrumentation and not under 2.3 Quality Assurance and control?

12863 Line 5-10: Previously, you described two sample inlets. Here you introduce a third. Describe all sampling lines together in one section.

12863 Line 10: What does a TI 3550 measure and why does it have a diffusing charging sensor? It doesn't seem to be in any of the tables of instrumentation and it is difficult to tell why this sentence is here.

12863 Line 14: Was this computer the same as the IPC mentioned earlier?

12863 Line 14: Revise sentence: "A freely rotating, high-resolution video camera installed atop the mobile laboratory continuously provided multi-angle views of on-road conditions to identify potential emission sources."

12863 Line 16: What particular model of Motorola GPS did you install?

12863 Line 23: "...and could be used to represent meteorological conditions along the 4th ring road..." What is meant by use of the word "represent"? From Figure 2 it seems that these stations were relatively close to the ring road. These were simply meteorological conditions measured within X km from the ring road. Are the authors implying that only meteorological conditions measured directly on the road are actually applicable for analysis? Again, this relates to the implication in the introduction of this work that stationary measurements cannot provide the same data as that of a mobile lab. This wording also goes against the claim that meteorological conditions were mostly homogeneous over the city of Beijing.

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12863 Line 26: Once again, the authors use the phrase "gas-phase instruments" without including the PTR-MS. These belong together throughout the article. Please check this carefully throughout.

12864 Line 3: "Difference between the calibration results and the concentrations of the standards was less than 4%." I think this phrase is supposed to mean that NO, CO and SO<sub>2</sub> values output while measuring the standard gas were only in error from the quoted value delivered with the standard by 4%. Please clarify this.

12864 Line 7: The limit of detection is printed as 0.3 ppbv for BTEX. Is this value good for benzene, toluene, xylenes+ethylbenzene individually or for the sum of all of them? Previously, it was stated that you only calibrated from several ppbv to several hundred ppbv. Later in the paper, values ranging from 0.4 to 0.6 are reported. How does this relate to the statement that "Gas-phase instruments were automatically zeroed and calibrated to 80 % of the detection range....".

12864 Line 11 to 19: A great deal more information is required if you are going to make such a comparison. Exactly which instruments are installed at the PKU laboratory? How are they calibrated? How far was the PKU lab sampling line separated from the mobile lab sampling line? If the mobile lab was parked near the lab (and not moving at 60 km/ h), how exactly did the inlets on the mobile lab function? What were the line losses like?

12864 Line 25: Does sampling only at 16:00 in the afternoon introduce a bias into your measurements? Can we expect to see other results at different times in the day? This highlights a great disadvantage to having a sampling inlet that only functions or is characterized for movement at 60 km/hr.

12865 Line 2-4: Why not put a N, S, W, E on Fig. 2 to show this. Also, on the corners that run diagonally, to which quarter do they belong (see for example N.E. corner on Figure 2...does that belong to the north or to the east)? What is the goal in dividing into 16 equal sections? This has to do with binning of measurements for purposes of

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correlation later. Please state this somewhere.

12865 Line 14-16: Why not discuss this or present it when the Chinese web site was given? This seems out of place here.

12865 Line 20: "...benzene, toluene, BTEX..." State either "sum of benzene, toluene, xylenes and ethyl benzene" or simply "BTEX" but not both.

12865 Line 21-23: There seem to be at least two different categorizations of the data with respect to pollution controls during the Olympics. One with three divisions (before, during, after) and a second with a number of divisions (as per table A1). These need to be reconciled with one another. This information also needs to be reconciled with Table 2 that actually lists the control measured implemented and date of implementation. It would be wise to discuss all of this at one point early on so that readers know what is going on. In addition, in Table A1, the row with national holidays comes as a bit of a surprise after what is stated in the text. In addition, the Post Olympics period should be listed as 20 Sept -???. We are still currently in the post Olympic period and so was the 6th of October.

12866 Line 1: 60 km/hr ...what was the deviation from this value? How often did you deviate. By what amount did you deviate?

12867 Line 4: "These differences suggest that the pollutants were influenced by different emission sources and subsequently by different control measures." The chain of logic isn't entirely clear here. The fundamental observation is that for the suite of chemicals observed, PM, BTEX, and NOx had the same mixing ratios before and after the Olympic controls. CO went down post olympics in comparison to pre-olympics while SO2, BC, and PM1 actually went up post Olympic compared to pre-olympics. While it may be that these chemicals have different sources and these different sources might be differently affected by pollution control measures, the way this is worded is a bit odd.

12867 Line 6-21: First of all, within the error bars it can certainly be said that over the

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three averaging periods there are differences. Except for BC and PM1, there are clear differences between pre, during, and post olympics. There is, however, some significant variation within each of these time periods. Discussion moves on to prevailing wind directions during this time period. However, Figure 5 shows only wind roses averaged over the entire measurement period, demonstrating that prevailing wind directions across Beijing were relatively homogeneous. The conclusions about pre-, during, and post-olympics are simply not pictured. It would be nice to see the average wind roses for each of these time periods in order to confirm the author's hypothesis that winds from the southwest resulted in generally higher loading of pollutants following passage of air over the city during the pre-olympics but not during or after the olympics.

12868 Line 2 to Line 13: Although it is interesting to look at correlations, it helps the reader along to say why we expect to see one in the first place.

12868 Line 15: "..By selecting the high-frequency distributions of RH and T among the whole data, the ranges in RH and T were chosen as 35-62% and 25-35 C, respectively." Please clarify how the data was treated here. 12868 Line 18-23: "However, benzene concentrations varied in different periods..." Yes, this was noted previously in section 3.1. Why are you repeating it here? This has nothing to do with correlations that were being discussed. Consider cutting this entire paragraph and restructuring the section having to do with correlations into a more cogent discussion. If you have nothing to say about correlations, then leave its discussion out entirely.

12869 Line 11: Has the abbreviation LDV been introduced before this point? The same comment applies to HDV which appears slightly later.

12869 These two paragraphs could be greatly condensed. Describe the postulated sources of each of the chemicals measured. Discuss how this relates to the correlations actually seen. Dividing the chemicals up as was done seems counterproductive and somewhat misleading. Essentially all of the chemicals/aerosols have some connection to traffic. The question is how much is traffic and how much is due to other

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factors.

12869 Line 22: "Since industrial activities have been slow down..." Change to "Since industrial activities were curtailed and biomass combustion was strictly prohibited..."

12870 Why do you need to make new divisions in time? If you insist on doing so, what do I have as far as pollution controls to distinguish between these periods? For instance, what is the difference between full scale control and full scale control during olympics. Aren't these identical? If not, why? It seems to me that the authors wish to demonstrate the correlation between vehicle numbers/hr and measurements. With this goal in mind, a simple time binning system should suffice where the binning has no connection to the application of regulations and simply has a good number of points that show trends clearly. It also seems as if this graph is the centerpiece of the discussion of the various measurements..

12871 Line 8: "...other emission sources, such as industry emission and biomass burning..." Change to "...other emission sources, such as industrial activities and biomass burning..."

12871 Line 11: "BC and SPM1 have weak correlations with CO and NOx, with correlation coefficients  $r$  having values less than 0.43... was stronger ( $r$  values greater than 0.45)"

12871 Line 13: "...since the SO<sub>2</sub> was strictly regulated in Beijing, the only possible remaining source of this chemical is from outside the city."

12871 Line 23-28 and 12872 Line 1 to 7: Was the choice of August 8 really based on Figure 7 or did it come from the change in the benzene/toluene ratio pictured in figure 8?

12872 Line 6-7: "...were mainly come from vehicular sources, most likely HDVs with diesel fueled engine." Revise to: "...mainly came from vehicular sources,..."

12872 Line 11: "This ratio is useful for estimating the photochemical age of an air mass

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(Khoder, 2007) and is therefore another indicator of the effectiveness of traffic control measures." Explain why an indicator of age indicates effectiveness immediately rather than waiting until the end of the paragraph.

12872 Line 18: "...variation in the B/T as is shown in Fig 8." Change to "...variation in the B/T ratio as is shown in Fig. 8."

12872 Line 19: "...which allows it to record emission plumes and reflect traffic-related factors such as traffic density, vehicle types, fuel composition..." Change to: "...which allows it to register passage of different emission plumes reflecting traffic-related factors such as traffic density, vehicle types, fuel composition..."

12872 Line 28: "This suggests that the lower B/T ratio before 4 August may have been caused by the heavy..." The authors present no evidence that reduction in painting activity rather than fugitive emissions or some other factor played any sort of large role. Do you have records showing how much painting was done as a function of time? Why the focus on painting?

12873 Line 6-8: "Studies earlier in this century have examined traffic-related BTEX behavior in the city of Beijing (Barletta et al., 2005; Song et al., 2007; Wei et al., 2007)..." Change to "Previous studies have examined traffic-related BTEX behavior in the city of Beijing (Barletta et al., 2005; Song et al., 2007; Wei et al., 2007) generally using roadside measurements employing gas chromatograph/mass spectrometry (GC-MS) or GC-flame ionization detection (GC-FID). Table 6 compares the B/T values obtained from these studies with our own work."

12873 Line 17: The authors say that they simply measured their values closer to the source and that this accounts for observed differences. Could there also be an instrumental contribution from the use of the PTRMS for measurement as opposed to GC-MS which is hinted at earlier in the paragraph? Could it be a change in the fuel mixture or solvent usage compared with previous studies? In short, why does it have to be due to proximity to the traffic? If it is proximity to the traffic, then how can your

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measurements be looked at spatially and temporally? If you are mainly influenced by local emissions, you won't have much to say about spatial or temporal variations over the entire study area because they would be drowned out or combined with a large dose of local emissions. Did you look at the traffic directly around you to find out what kind of correlations exist?

12873 Line 25: "...model stimulation and stationary measurements and real-time regional observation is needed to validate these results." Change to: "...model simulations and stationary measurements. Real time regional observations are needed to validate these predictions."

12874 Line 3: "...Ring Road on 6 August; prevailing winds were from the southwest and south." Change to "...Ring Road on 6 August. Prevailing winds during these measurements were from the south and southwest."

12874 Line 12: "...SPM1, indicating that they were from similar combustion sources."

12874 Line 14: "...PKU site, while background increases with broad peaks..." Change to "PKU site. Background increases with broad peaks..."

12874 Line 16: "...from local industrial sources lying a relatively short distance from the road (approximately 12 km). Along Jingshi Highway, the peaks likely reflect sources lying within a larger radius (30 km), showing the effects of regional transport."

12874 Line 21: "This study demonstrates that the mobile laboratory is a..."

12874 Line 23-24: Perhaps revise in this way: "The mobile laboratory collected data that could not have been obtained through currently existing and equipped stationary observation sites in the Beijing area."

12875 Line 10: "...gradually after the Olympic regulations were lifted, especially after the traffic control period..."

12875 Line 13: "...were shown afterward, a trend also reflected in the number of HDVs

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on the road."

12875 Line 15: "...indicated that they had similar local emission or regional transport sources which were later controlled."

12875 Line 19: "The B/T ratio, regarded as a source indicator..."

12875 Line 20: "in the period after 8 August."

12875 Line 21: I do not like how this is stated. While it might be true that this ratio changed due to a change in the amount of painting occurring before and after the Olympics, there is no hard evidence for it. All that can be said is that the ratio changed and there are a number of possible reasons for this change, one of which might be a change in the amount of painting.

12875 Line 26: "...implemented by the Beijing EPB were able to effect short term improvements in air quality."

12881: The Parameter listed for the PTRMS is "VOCs". However, no VOC mixing ratios other than those related to BTEX are listed. Change this entry to BTEX. In the detection limit column, the words "Common standard" are used. What is the common standard? Please report a number for the devices used or state what the common standard is.

12882: "Before" is not a starting date. Where were the heavy industrial polluters relocated? Who were the heavy industrial polluters? Where was the desulfurization facility installed? "50% of government cars were not allowed to drive. Diesel and heavily polluting vehicles not allowed to drive in Beijing. Only those vehicles meeting emission standards equivalent to Euro II were allowed to enter Beijing." "Start of full scale control: Reduce or stop production at certain factories surrounding Beijing." "Extra 20% of governmental cars were not allowed to drive. Temporarily close some gas stations. Increase bus fleet and transit frequency. Lifting of regulations adopted from July 20."

12886: 0.49+-0.55 not 0.49~0.55

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12887: Revise this table. Consider putting the national holidays as a foot note. The post olympics period comprises everything from 20 Sept on to today.

12888: Consider making a close-up picture of the inlets and give a better description of each of the three inlets. Make sure these descriptions are together. Why is the PTRMS attached to the aerosol sampling inlet in the picture of the van in Figure 1 although it is making measurements of gas-phase molecules rather than aerosols?

12889: "The 4th Ring Road was chosen as the sampling route. For analysis purposes, this route was binned into 16 sections of equal length seen here colored red and blue." "The red stars show meteorological stations and places where traffic speed was recorded."

12890: In the figure caption, describe the meaning of the white areas. The symbol for micrometers is a u rather than the greek letter mu. How many bins are there per Cardinal direction? Figure labels of x axis don't seem to be aligned.

12893: "...each of the road lines shown in Fig.2." Does this second sentence mean that you only included points for which there also existed a point for RH and T?

12894: "...on policy changes; before full scale control..." Units of traffic speed are "units/h". Is "km/h" intended? Where does the 8th of August fall in this plot?

12895: Units of traffic speed are "units/h". Is "km/h" intended? Where does the 8th of August fall in this plot?

12896: Units of traffic speed are "units/h". Is "km/h" intended? Where does the 8th of August fall in this plot?

12897: "The ratio of benzene to toluene for all cruises. The light grey line represents minute averages. The black line with symbols shows daily averages. The red line shows the average ratio as calculated for the stable period (from ??? to ????).

12898: "...The long dashed line in (b)...."

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