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Interactive comment on "Using a mobile laboratory to characterize the distribution and transport of sulfur dioxide in and around Beijing" by M. Wang et al.

Anonymous Referee #1

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General comments.

This is an interesting paper that attempts to calculate SO2 mass transport on a regional scale in and around Beijing, China. Five days of surface measurements of SO2 from a mobile van are combined with modeled wind fields and boundary layer heights to derive mass fluxes along transects defined by the highway network around Beijing. An additional factor the authors touch on is the emissions reductions mandated around the 2008 Olympics time period.

I found the logical development difficult to follow. Data and derived flux values for the different days are presented, but since multiple variables (wind speed, source region,

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emissions source strength) vary independently on each day, it was not easy to understand the reasons leading to the factor of over 10 in flux into Beijing presented in the final analysis. This range is sufficiently large to determine whether regional transport is significant, or is a minor player, in the SO2 budget of Beijing.

It is also not clear from this paper alone how, or where, or by how much the mandated emissions reductions during the Olympics actually affected SO2 emissions. Comparison to the 2007 annual inventory is appropriate, but the spatial distribution of emissions changes could lead to a very different source pattern during the 4 days of mobile lab sampling, and need to be mentioned and incorporated somehow.

Clarity is also lacking in the application of the flux calculation (Eqn. 1) to the measured data. At times the value for sin(theta) approaches zero, so a different approach is used, but is not compared to the days for which sin(theta) approaches 1 and for which the two approaches should be identical. I did not understand how the background concentration was defined, and it seemed to be defined differently for different cases (e.g., the local flux calculated for the Jingshi Highway vs. that calculated for "regional transport" to the Shijingshan district). I would recommend that the data for each calculated flux for each transect on each day be shown in a figure, including the choice of background adopted for that particular transect.

Comparison of derived flux on 20 August, which is characterized as "low" to that on 4 September characterized as "high" is not well supported. This compares data taken during a morning drive (8 AM - 1 PM on 20 August) including times when the boundary layer was not fully developed, to data taken during the afternoon (2 PM - 6:30 PM on 4 September). Since upwind SO2 sources may include power plants which may not have fully mixed down to the surface in the morning of 20 August, this represents a potential source of bias that needs to be addressed.

There are several other inconsistencies that make this overall not ready for publication in its present form. The relevant information is presented, but scattered through different tables and figures so that it was difficult to follow the development of the text.

I can recommend a few changes that should help improve clarity and logical development:

- Present the 5 days of mobile SO2 data, and note the dependence of measured concentration on source strength, transport direction, and dilution due to variable meteorology on each day. Then discuss each factor separately, before attempting to calculate a flux into the Beijing area.

- Variability in source strength: Present/discuss the spatial distribution of annuallyaveraged SO2 emissions, and then note the magnitude, location, and timing of changes expected due to the mandated controls due to the Olympics. It could be that controls altered the spatial distribution of SO2 emission around Beijing that the annual averages presented in Table 4 do not reflect the relative contributions of the source regions during this period. A focused discussion is needed, one that does not assume in-depth prior knowledge of this issue.

- Variability in transport direction/source region: Present /discuss the Hysplit trajectories and WRF fields. (I think there is an error in the Hysplit back-trajectories – they should correspond to the actual mobile van sampling times, which are different (at present the trajectories in Fig. S6 are both initialized at 1400 UTC). I think they should be initialized at 0200 UTC for Aug. 8 and at 0800 UTC for Sept. 4. If that is correct, the 48-hour back-trajectory for Aug. 8 looks quite different than presented in Fig. S6, and suggests southerly transport from eastern mainland China rather than from the Bohai Sea and over Tianjin – i.e., more similar upwind direction (but shorter fetch) to that observed on Sept. 4.

- Variability in dilution: The derived fluxes will depend linearly on the wind speed at time of emission, not time of measurement. An additional factor, detrainment into the free troposphere of surface sources (and equivalently, entrainment into the surface layer of elevated sources of SO2 from power plant stacks) is not discussed and should

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be added.. A focused discussion of the assumptions, and resulting dependencies on dilution, inherent in the flux analysis is necessary before calculating fluxes.

- Once these issues are addressed, then pick and contrast the data from days that most nearly illustrate a change in only one variable. For example, are there two periods that sampled the same source region, with similar meteorology, but under different emissions scenarios (pre- and post Olympics?)?

At present the large differences in derived flux between the 5 days are not quantified in terms of possible differences in sources, transport, or dilution. Further, reaction during transport of SO2 is not considered explicitly, but must have differed between the days – to what degree is not known or discussed.

Finally, Although the differences in derived SO2 flux are large – factors of ~ 10 – between the different days, in an absolute sense they are relatively small (a flux of 2 kg/second is roughly equal to 2e25 molecules per second, which is equivalent to a single large coal-fired electric utility power plant. My impression is that the derivation of flux over the spatial scales in this paper has uncertainties on that order, so that the differences between days are not especially large. Certainly the differences between the various Ring Road fluxes on a single day (Table 3) approaches the differences between different days. A more careful error discussion and propagation should help to establish the significance of these findings.

The English usage is mostly excellent but there are still several areas that could use some editorial input from a native speaker.

Some specific comments are below.

p. 16469 line 26: comparison of AM to PM data is difficult without establishing that any transported SO2 aloft from distant, elevated sources has actually mixed down to the surface, esp. for the data on Aug. 20 that begin at 8 AM. What PBL height was chosen for this flux calc? The PBL height changes rapidly over time during this period,

as shown by Fig. S3.

p. 16471 line 20 and following: a more thorough discussion of detrainment/entrainment issues for the various days is necessary – these processes would bias the flux calcs. differently for different source types (surface vs elevated sources).

p. 16472 line 25: Please develop an estimated uncertainty for the wind speed, PBL height, and degree of entrainment/detrainment for the combination of WRF and surface measurements. These errors will directly affect the accuracy of deriving fluxes from the input data.

p. 16473 line 15: Cite White et al., Science, 1976 for the flux calculation.

p. 16473 line 23: Vi is the wind speed at time of emission , not measurement, and the inherent assumption is that it is constant between emission and measurement.

p. 16474: The API quantity is introduced but is distracting. This value is nonlinear in concentration, is derived from consideration of 5 pollutants (O3, NOx, SO2, CO, and PM10) and in fact is not correlated with SO2. Not sure if its introduction here is useful to the discussion.

p. 16478, line 5: Why is the PBL height so different on 11 September?

p. 16480 line 2: Please quantify instead of stating "... we are confident that our flux calculations are reasonable with acceptable uncertainty."

P. 16480, line 7: Reference should be to Lu et al., (2010).

Fig. 2: Please note the mobile van sampling days on the time series – Aug 6, 20, and Sept. 3,4 and 11. During what period, and approximately by how much was the overall SO2 reduction taken around the Olympics? Please indicate on the figure.

Fig. 3: Are these 24-hour trajectories? Please include in the caption.

Fig. 5: Change caption to note the SO2 values were measured by the mobile van.

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Please remove the grid cell lines, which are very distracting, and make the wind barbs larger for clarity. Consider a larger map to provide context for the data – I found this map hard to reconcile with the different areas shown in Figs. 1, 3, 6, and 8, and even harder to deduce what part of the region was shown. Perhaps provide more road/city outline information in each map, for those not as familiar with the geography?

Fig. 6: larger wind barbs for clarity.

Fig. 7: Please rescale Y-axes to show all plots from 0 to 40 ppbv. Sept. 3 has by far the highest concentrations of SO2, but I could not easily find a derived flux elsewhere in the text?

Fig. 8: Show a larger map or an inset – context is lacking, it took a while to understand where these data were in relation to Beijing. The wind barbs are indecipherable small. Would an average wind vector be more interpretable? The Shijingshan and Jingshi Highway sections appear to have significantly different derived fluxes, implying very different choices of backgrounds – please graphically indicate what the background choices were in the figure.

Fig. S3: Indicate what the drive times were for the August 20 plot – I think this started relatively early in the AM when the PBL had not fully developed. What value was used for the flux calc? Please also show similar data for the other two mobile van sampling days. What led to the very low PBL height on Sept. 11?

Fig. S4: Not sure what geographic region these plots correspond to. Please overlay some relevant map data – Beijing, coastline, roads, etc.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16465, 2011.