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Atmospheric Chemistry and Physics Discussions

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.

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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"

Response: We appreciate the referee 1 for precious comments to improve the quality of our paper. We have made extensive revisions according to your suggestions. Details

of the changes can be found in the following responses.

"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, 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."

Response: Accepted. We have thoroughly discussed the causes of the fluxes differences in the two days. In the section 3.3, three major impacting factors were attributable to the result: variations of emission patterns during the control and non-control period, variations of transport directions and dilution process on the way. Firstly, to explore the emission changes, we added maps of emission inventory in the emission control and non-control period and the difference in between. A clear reduction in SO2 emissions in Beijing and neighboring provinces can be identified. This is in agreement with the relatively low SO2 influx on 20 August (control period) and high SO2 influx on 4 September (non-control period). Secondly, we used NOAA backward trajectory model to trace the sources of SO2. Air masses on 20 August were derived from the area between Beijing and Tianjin while the source on 4 September was found in the south mainland of China. Anthropogenic emissions in this area were ten-fold higher than the emissions in Tianjin. Thirdly, meteorological conditions on 20 August (e.g. low wind speed, relatively high humidity) were favariable to the transformation and deposition of SO2 comparing with that on 4 September. Meanwhile, updraft emissions from high stacks (e.g. power plants) were not completely mixed down to the surface in the unstable boundary layer (e.g. 20 August). Thus, the SO2 concentrations from sources were partially diluted. All the aforementioned factors resulted in ten-fold difference in flux in the two days. Detailed discussions please see section 3.3.

"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. Compari-

son 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."

Response: Thanks for the suggestion. Actually, the intention of this study is trying to characterize the spatial distribution of SO2 and identify possible causes leading to the increase of SO2. We do not intend to quantitatively estimate the effect of emission control on the variations of SO2 because it requires more systematic measurements in both temporal and spatial scales. We want to find out and explicitly explain the reasons behind the potential pollution events in the sampling days. However, we agree that the spatial distribution of emission inventory and their emission changes is helpful to understand the sources attributed to the SO2 variations. Therefore, we incorporated our measurements to the emission inventories which were supported by Beijing EPB. Detailed discussions are available in section 3.3.

"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."

Response: In Eqn. 1, the sin(theta) indicates the wind vector which contribute to the influx of SO2 while the cos(theta) indicates the wind vector which has no contribution to the influx of SO2. The principle has been drawn in figure S5. In particular, if the sin(theta) approaches 1, this means that the wind vector is directly towards Beijing and therefore, contributes most of the SO2 flux to Beijing. Similarly, when the sin(theta) approaches 0, no SO2 flux is expected to transport into Beijing. To better understand

the figure and equation, we add the location of Beijing in figure S5.

For the study of the regional transport in the southeastern area(route 1 and 2), what we calculate are the total SO2 flux to Beijing, we didn’t define background concentrations. This is commonly used by several studies as well(Wang et al., 2006; Johanssen et al., 2009; Rivera et al., 2009). In the southeastern area (route 5), we found the emission sources were complex, including both local and regional transport emissions. To distinguish the overlay of local emission from regional transport, we use a simple spline interpolation to separate the SO2 from local emission and regional transport. To avoid misunderstanding, we use the “regional transport” instead of “regional background” on page 16479 line 5.

"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."

Response: Accepted, we addressed the following sentences:” since boundary layer on 20 August was not fully developed during the morning sampling, upwind SO2 sources from power plants may not have fully mixed down to the surface, the detrainment of SO2 into the free troposphere of elevated sources from power plant stacks may lead to potential bias of SO2 detected by the measurements.”. However, detrainment and entrainment were more effective in proximity to power plants. Since most power plants emissions surrounding Beijing were strictly regulated during the Olympics, this kind of emission was more possible by long range transport. Therefore, this effect was expected small due to downdrafts in long distance (Krautstrunk et al., 2000).

"I can recommend a few changes that should help improve clarity and logical devel-

opment: - 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."

Response: Accepted this good suggestion. We have tried to re-write our paper fully according to the suggestion. The paper starts from descriptions of SO2 spatial distribution incorporation with the wind field in the five observation days(section 3.2), then provide insight discussions on the emission changes of SO2, variability of transport directions and dilution processes(section 3.3). Detailed information please see section 3.2 and 3.3.

"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."

Response: Accepted. The issues leading to the differences of derived flux have been addressed in section 3.3. Moreover, reactions during the transport have been discussed as well in section 3.3. Since quantifying the difference of transformation process during the two days requires highly advanced instruments such as AMS and needs complicated simulation with chemical reaction models, we only provided reasonable explanation for the effect of transformation on the difference of SO2 flux in days.

"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) approach the differences between different days. A more careful error discussion and propagation should help to establish the significance of these findings."

Response: Accepted. We have discussed thoroughly the uncertainty of flux calculations in section 3.5. The overall error was estimated approximate 31%. Moreover, the derived SO2 fluxes in the two days are roughly equivalent to the reported emission rate from power plants or industrial complex. The influxes have been compared with the annual emissions in Beijing in both control and non-control period, indicating the transport of SO2 is of pivotal importance to the SO2 concentration in Beijing. We also retrieved the total SOx using SOR index from measured SO2 which implied comparable results to the annual emissions in the surrounding city or province of Beijing (section 3.4).

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

Response: The manuscript has been corrected by native speaker.

"Specific comments: 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."

Response: Accepted. The potential bias has been addressed after the flux calculation, please see the response to former question. The PBL height for the flux calculations on the ring roads were 885, 950 and 1025 meters.

"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). "

Response: Accepted, we have discussed it in section 3.3 and potential bias has been

estimated in section 3.5.

"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."

Response: Accepted. We wrote section 3.5 to fully estimate potential bias of SO2 influx calculation in terms of accuracy of wind field in comparison to station network, vertical profiles of winds and SO2 distributions, updraft effects on high stacks, PBL modeling regarding to lidar, temporal fluctuations of wind field in the sampling period. The overall error is about 31%.

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

Response: Accepted.

"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."

Response: Accepted. The assumption has been mentioned in the manuscript.

"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."

Response: Accepted. We delete the text and figure related to API.

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

Response: The low boundary layer on 11 September is mainly due to the strong inversion layer and the weak wind near the surface.

"p. 16480 line 2: Please quantify instead of stating "... we are confident that our flux

calculations are reasonable with acceptable uncertainty.""

Response: Accepted. The uncertainty has been assessed and the overall error is about 31%.

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

Response: Corrected.

"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."

Response: Completed accordingly.

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

Response:Corrected.

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

Response: Accepted. The grid cell lines have been removed and wind barbs have been enlarged. A map background has been added to provide geographical information. Besides, we marked the route numbers on each figures so that readers can make sense of the mobile sampling routes relating to the geographical locations.

"Fig. 6for : larger wind barbs clarity."

Response: Accepted. The grid cell lines have been removed and wind barbs have been enlarged.

"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?"

Response: Corrected.

"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."

Response: A map has been added and wind barbs are enlarged. As illustrated in the text, the emission sources in the Shijingshan and Jingshi highway include both local and regional transport sources. Since the local emission sources in Shijingshan district haven't been geographically mapped, the intention of figure 8 is trying to identify and demonstrate the exists of them and to calculate their fluxes individually.

"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?"

Response: Corrected. The indications for the sampling times were highlighted. The PBL height for the flux calculations on the ring roads were 885, 950 and 1025 meters. Unfortunately, the data from lidar measurements were only available in those three days. We think from the comparisons of the three days, the WRF model predicts generally well of the PBL. The cause of the low PBL on 11 September has been explained in previous reply.

"Fig. S4: Not sure what geographic region these plots correspond to. Please overlay

some relevant map data – Beijing, coastline, roads, etc."

Response: The map cannot fit to fig. S4, we therefore added supporting information including the mobile sampling routes and marked the cities around.

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