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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantitative evaluation of emission control of primary and secondary organic aerosol sources during Beijing 2008 Olympics" *by* S. Guo et al.

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We thank the referees for his/her careful and critical review of our paper. The followings are our responses to the reviewers' comments.

1) In general, careful revisions by a native English speaker would greatly benefit the quality of this paper. There are numerous grammatical errors (examples: pg. 32884 ln. 11 "of vehicle emission", ln. 18. "regional" should be "rural"; pg. 32885 ln. 8 "other" should be "rather", ln. 25 remove "the"; pg. 32886 ln. 17 "precious" should be "previous"). There are also umerous poorly formed or awkward sentences (examples: pg. 32885 ln. 1; pg. 32886, ln. 20-22; pg. 32887 ln. 8, ln. 16, ln. 19; pg. 32889 ln.

3-5). These examples are just a sample; a thorough review of the whole paper should be carefully done.

We thank the referee to point out this. We asked our native English speaking colleague to go through our manuscript.

2) pg. 32888 ln. 12: It is very unlikely that there was no cooking aerosol in Beijing during the study. Cooking aerosol is present in most studies in urban areas and comprises a major fraction of aerosol in Beijing in Wang et al. (2009). The majority of the observed OC at both sites was not apportioned. It is likely that cooking SOA is in that "Other OC" group and is still important for urban and rural OA.

We agree with the referee. In previous study, such as Wang et al. 2010, cooking contributed a significant fraction to organic carbon. We also tried to include cooking emission in the CMB model. The results showed that cooking can contribute 2.0$\pm$1.6% and 3.1$\pm$2.1% to organic carbon at PKU and Yufa, respectively. The highest contribution occurred in August 30th day-sample at PKU and July 27th day-sample at Yufa with the contributions of 9.5% and 11.3% to total OC. However, when we use cooking emission in the CMB model, the results for 32 out of 74 samples at PKU and 21 out of 99 samples at Yufa are not very reliable (the square regression coefficient of the regression equation R2 <0.80, the sum of square residual Chi-square value ïĄčÂň2 > 4. It is probably because Chinese cooking emission is very complicated. Thus, to make the results more reliable, we decided not use cooking in the model. However, even if cooking is not included in the model, its contribution can be included in other OC. As the referee mentioned, another reason may be because the primary cooking particles were aged and transform to secondary organic aerosols. We made some clarification in the revised manuscript to explain why we did not used cooking in the model as following: " In addition, when cooking was used in the model, the fitting results of some samples were not reliable (in 32 out of 74 samples at PKU and 21 out of 99 samples at Yufa, R2 <0.80, and ïĄč2 >4). Thus to make the results more reliable, cooking was not considered in the model. Another reason for this low contribution of cooking may

be because the primary cooking particles were aged and transformed to secondary organic aerosols. However, if cooking or other sources contributed to OC, they would be apportioned as other OC."

3) Table 3: Why are the SOA sources not included in this table?

We agree with the referee. The SOC contributions were added in Table 3.

4) Table 4: It would be very helpful if the excess non-apportioned OC was shown in this table as well. In line with the comment above, Tables 3 & 4 need to be more consistent. Same with figure 2; are the percentages shown of everything or just the apportioned aerosol?

We thank the referee to point this out. We added non-apportioned OC in Table 4. The percentages in figure 2 are of total measured OC. We added description in the figure caption.

5) p. 32891 ln. 15: The author uses carbon predominance index (CPI), which is not used in this field and the term is not explained in the references given. I suggest removing this comment. Additionally, It is worth mentioning recent work on primary emissions of VOCs from gasoline and diesel vehicles (P.N.A.S.) may point to the importance of diesel emissions with the high concentrations of C12-C22 alkanes.

We thank the referee to give this useful suggestion. We have removed the discussion about CPI and mentioned PNAS paper to explain the source of C12-C22 alkanes. Please see the following: "However, low carbon number species such as C19, C20 and C21 were also high especially at urban site. A recent work reported diesel and gasoline vehicles can emit large amount of C12-C22 alkanes (Gentner et al., 2012). The results of our work suggested fossil fuel combustion, especially vehicle emissions were important in Beijing."

6) Sec. 3.4: In general there are too many numbers in the text that are either shown in Table 4 or could be shown graphically with greater efficiency. Reducing the incidence

of numbers will greatly improve readability.

We thank the referee to point this out. We have removed some numbers, and left only important ones.

7) pg. 32897 ln. 19: As the authors say earlier on the page, SOA formation is very complicated. Due to the interconnectivity of anthropogenic/biogenic oxidation and SOA formation, simply comparing the ratio of anthropogenic to biogenic SOA is insufficient to assess the effect of control measures. The ballpark estimate does not consider the impact of differences in temperatures over this period (a driver of biogenic emissions); changes in NOx emissions (NOx levels affect the rate of oxidation & SOA formation); and the authors have not apportioned the majority of the OA. Whether this "other OC" aerosol is anthropogenic or biogenic could have a major effect on this basic calculation. Furthermore, toluene SOA is just a marker for one type of anthropogenic SOA from toluene and potentially similar aromatics. SOA from alkanes is not considered and cannot be assumed to scale with the toluene SOA marker, especially considering the differences in SOA from gasoline vs. diesel and the variable decrease in emissions from the two vehicle types during the study. So, it is inaccurate to say that the anthropogenic SOA is "constrained" during the control period. More advanced techniques and modeling needs to be done to study this. The authors should consider removing this portion of the paper.

We really agree with the referee. Actually, when the manuscript was under review for ACPD, one of the referees suggested us to discuss more about SOA. We also think from our results it is difficult to evaluate the emission control on SOA formation. The related text was removed, and we also suggested more advanced techniques should be used to investigate the effectiveness of emission control on SOA formation.

8) Table S2 gives PM (size cut needs to be specified) concentrations for a number of species that will be largely partitioned into the gas phase, if not entirely in the gas phase. These intermediate volatility organic compounds (IVOCs) needs to be better

labeled in the table to indicate that the reported concentrations are underestimates and that some of the reported mass may actually be vapors that condensed on the filters during sampling. Examples: naphthalene, methylnaphthalenes, dimethylnaphthalenes, acenaphthylene, acenapthene, & n-alkanes (12-22).

We thank the referee to point this out. Size cut was added and IVOCs compounds were marked in the table.

9) It seems inaccurate to call the first period the "non-control period" since control measures were in place. We agree with the referee. We have changed the period name to "Not-fully-controlled period (NC)".

Minor comments: 1) It may be more appropriate to use "fine mode" rather than just "fine" when referring to particulate matter. Furthermore, "particulate matter" or "aerosol" is more appropriate than "particles".

We have changed some of the "fine" to "fine mode", and "particles" to either "particulate matter" or "aerosol".

2) When referring to the extra material not in the paper, it is more appropriate to write "the supplementary material" rather than just "Supplement". The related text has been revised. Thanks for pointing this out.

3) pg. 32890 ln. 18-21: this is repeated from end of sec. 2.2. This part has been removed.

4) pg. 32888 ln. 21: I would not call the tracer-yield method "very useful". It is a basic method that offers a very rough way to estimate SOA. This statement has been removed.

5) pg. 32891 ln. 18: The statement about PAHs from volcanoes does not seem very relevant. The statement about volcanoes has been removed.

6) Tables 1-2: The format should have the month first before the year. The date format

has been revised.

7) Fig. 1: Label control periods Control periods have been labeled in figure 1. 8) Table S1 is almost completely redundant from the text in the paper; I suggest removing it. We agree with the referee. Table S1 has been removed.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 32883, 2012.