

## ***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) Prior studies (Table 3) indicate the importance of cooking contributions to organic carbon in Beijing. It is noted in the experimental section that cholesterol was not detected in most samples; however, it is reported in Table S2 as a quantified value for both PKU and Yufa sites. For the observed cholesterol levels, what concentration of cooking-derived organic carbon is estimated on average? What contribution from cooking might be expected for specific dates when cholesterol concentrations were

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greatest? How do other molecular markers that are characteristic of cooking (i.e. fatty acids, also reported in Table S2) provide insight to this source? How do CMB model results change when cooking tracers are included?

We agree with the referee that 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 by using other tracers, such as Oleic Acid. 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  $R^2 < 0.80$ , the sum of square residual Chi-square value  $\chi^2 > 4$ ). 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. We made some clarification in the revised manuscript to explain why we did not use 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,  $R^2 < 0.80$ , and  $\chi^2 > 4$ ). Thus to make the results more reliable, cooking was not considered in the model. However, if cooking or other sources contributed to OC, they would be apportioned as other OC."

2) Table 1: Describe the calculation of the measurement uncertainty (or clearly indicate that this is the standard deviation, standard error, as applicable).

We thank the referee to point this out. The uncertainty stands for standard deviation. We have clarified this in Table 1.

3) Table S1 is not useful, as fitting requirements are summarized in section 2.3.

We agree with the referee, and Table S1 has been deleted.

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4) References to CMB profiles appear erroneous or incomplete. For example, Zheng et al., 2005 is a source apportionment paper and not a primary article reporting a coal burning profile. Justification of the selection of profiles is needed, especially for biomass burning. Why is a profile for fireplace combustion of wood in the United States used, when open-burning from nearby provinces is expected to be the source of levoglucosan? What more representative profiles are available? And how do CMB results change with selection of biomass profile (i.e. sensitivity test).

We thank the referee to point this out. Because fully developed local source profiles for coal burning of Beijing are not available. The only available profile is from Zheng et al.'s (2005) work. In their work the coal profile was obtained from the analysis of direct emissions from the burning of Datong coal in a small cooking oven in a house in Yungang, China, which were collected by placing the inlet of the sampler into the diluted smoke plume. We add the explanation in the text to this as following: "Because there is no well-developed source profiles for coal burning of Beijing, source profile for coal combustion from Zheng et al. (2005)'s work was used in this study. The profile was obtained from the analysis of direct emissions from the burning of Datong coal in Yungang, China."

We did the sensitivity test of the CMB model results by verifying the input biomass burning profiles. The different input source profiles varied the biomass burning contribution by the factor of 0.75-1.33, and other source contributions were impacted within 10%. The reason for choosing source profile from fireplace is because open biomass burning was strictly constrained in and around Beijing area during the measurement period. Another reason for choosing fireplace profile is because we want to use the same source profiles as previous work (Zheng et al., 2005) to make the results comparable. However, our conclusion was not precise that we can only conclude the biomass burning (not specifically open biomass burning) from nearby province can effectively impacted the air quality of Beijing. The sensitivity test and reason for choosing biomass burning source profile was added in the session "2.3 Source apportionment" as following:

“The sensitivity of the CMB model results was tested by verifying the input biomass burning profiles. Four biomass burning profiles were used, including one from open biomass burning (Lee et al. 2005), two from wood stoves (Fine et al., 2004; Wang et al., 2009), and one from fireplace (Schauer et al., 2001). The different input source profiles varied the biomass burning contributions by the factor of 0.75-1.33. Other source contributions were impacted slightly (within 10%). The contributions were highest when open biomass burning profile was used. However, open biomass burning was strictly constrained in and around Beijing area during the measurement period. In addition, the contributions were very close (by a factor of 1.09) when wood stove and fireplace source profiles were used. In this study, fireplace profile was used because we want to use the same source profiles as previous work (Zheng et al., 2005) to make the results comparable. This sensitivity test convinced the CMB results were reasonable and reliable.”

5) Additional section in methodological description is needed discussing the statistical analyses used in comparing data across locations and time periods. It is suggested that section “2.4. Statistical analysis” follow the methodological description of source apportionment.

We agree with the referee. An additional section “2.4 Statistical analysis” was added in the text, and a summary of statics from t-test was also added in the supplement material. “2.4 Statistical analysis Statistical analyses were used to compare data across locations and time periods. A pair t-test was used to compare the data between two sites. To evaluate the emission control effectiveness, F-tests and t-tests were employed to first qualitatively test whether the pollution concentrations during different periods have statistical differences (significance level 5%). F test is used to decide whether two groups have statistically different variances so that proper t-test can be chosen. T-test was employed to determine whether there are statistical differences between controlled and non-controlled periods. Then the mass concentrations of specific sources, as well as their contributions to total OC, were compared directly between dif-

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ferent periods, to quantitatively determine the variation of different source. A summary of the statics from t-test was listed in Table S1 in supplement material.”

6) What accuracy and precision is expected for PAH concentrations (page 32891 lines 21-23) and hopanes concentrations (page 32892, lines 12-15)? It is questionable whether all of the reported digits are significant.

We thank the referee to point out this. The significance digits of the data have been revised.

7) Discussion of PAH distribution (page 32891, lines 25-end): Citation needed for discussion of ring-number distribution and its relationship to temperature.

We thank the referee to point out this, we have added the reference and revised the text as following: “This distribution was consistent with the previous work and was attributed to the volatility of PAHs and the high ambient temperature in summer time (Wang et al. 2009).” 8) Levoglucosan concentrations (page 32892, lines 5-10) – are these to be nano-grams per cubic meter? If micrograms per cubic meter, they account for more than the observed PM organic carbon.

We thank the referee to point this out. The unit should be ng/m<sup>3</sup>. The text has been revised.

9) Hopane concentrations (page 32892) – do the relative amounts of hopanes provide insight to their sources? They can also be emitted from coal combustion, which is known to be an important source in the region.

We did not think about this before, and thanks for pointing this out. This should be clarified in the text. The hopane concentration can to a certain extent to indicate the vehicle emission, and coal burning can also effect particulate hopane concentration. However, in summer of Beijing, coal burning is not very significant, so the main source of hopanes was from vehicle emission. We have revised the text as following: “Hopanes are not abundant in atmospheric particles, but they can well indicate the vehicle emission (Si-

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moneit, 1986). Although coal burning can also emit particulate hopanes, their contributions were not as high as vehicle. In addition, in summer of Beijing, coal burning is not very frequent, so the main source of hopanes was still from vehicle.”

10) Table 3 should explicitly report SOC sources (as suggested on page 32893, line 7). Likewise, a citation to Guo et al., 2012 should be included.

We agree with the referee. The SOC contributions were added in Table 3, and the citation to Guo et al. 2012 was also added.

11) The calculation of the uncertainty in source apportionment results needs to be described in detail – is this standard error, 95% confidence intervals, or propagated measurement/model uncertainty?

We thank the referee to point this out. The uncertainty is the standard deviation of the source contribution from different samples. We have clarified this in the text and related table.

12) Variation of organic particle sources – the F test (page 32895, line 7) is used to compare the variances of measurements. How and why were F-tests used to compare values across time periods? How were mean contributions of sources quantitatively compared? Summary statistics from which the authors draw conclusions should be incorporated into the Supplemental Information.

We thank the referee to point this out. There is a mistake here. We want to say the F-test and the t-test were used. In our work, F test is used to decide whether two groups have statistically different variances so that proper t-test can be used to qualitatively judge whether the contributions between different periods have statistical difference. In other words, the t test results can qualitatively determine whether the emission control had statistically effect on reducing particle pollution. A summary of statistics are listed in the supplemental information.

13) Biogenic vs. anthropogenic SOA – Need to be more quantitative about increases

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in biogenic SOC during the control period; “little higher” is insufficient (page 32897, line 17). Was this difference in biogenic SOC statistically significant? Data is needed to support the conclusion that “emission control constrained anthropogenic SOC”.

We agree with the referee. The statistical result showed the difference between anthropogenic SOC was not significant, but biogenic SOC was. The increase of biogenic SOC is about 8%. The ratio of anthropogenic to biogenic SOC was employed to explore whether the control measures had impact on anthropogenic SOC formation. This ratio was 2.0 in non-control period which is higher than that in controlled period (1.7), implying the emission control in a certain extend constrained the anthropogenic SOC formation. The related text was revised as following: “A t-test result showed that the anthropogenic SOC contributions did not have significant differences between controlled and non-controlled period. However, the biogenic SOC contributions such as isoprene SOC had statistical difference, with an increase of 8% during controlled period. For biogenic VOC emission cannot be controlled, the biogenic SOCs were considered to be affected mainly by weather conditions. The ratio of anthropogenic to biogenic SOC can simply explore whether the control measures had impact on anthropogenic SOC formation. This ratio was 2.0 in non-control period which is higher than that in controlled period (1.7), implying the emission control to a certain extend constrained the anthropogenic SOC formation in urban area. Wang et al. (Wang et al., 2010a) reported the toluene mixing ratio decreased during the traffic control period. The lower precursor concentrations may lead to the lower anthropogenic SOC concentration. However, it is difficult to quantitatively evaluate this reduction.

14) Conclusions – do the authors have any recommendations of how to reduce SOC pollution?

From our data, it is difficult to tell how to reduce SOC pollution. However, we give some suggestions. Since weather condition was the major influencing factor on SOC formation, reducing oxidant concentrations such as O<sub>3</sub> may be a good way to reduce secondary particle pollution. To improve the regional air quality, especially reduced the

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SOC pollution, more strict control measures should be implemented in a larger regional scale in the future.

Minor typos- Entire manuscript would benefit from copy-editing with attention to grammar. Page 32886, line 17: precious -> previous Page 32887, line 15: chromatograph -> chromatography; ionic compounds -> ions Page 32891, line 1: n-alkanoic -> n-Alkanoic We really thank the referees for his/her careful review of our paper. All the suggestions are very useful to improve our paper.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/12/C12911/2013/acpd-12-C12911-2013-supplement.zip>

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