

## ***Interactive comment on “Carbonaceous species in PM<sub>2.5</sub> at a pair of rural-urban sites in Beijing, 2005–2008” by F. Yang et al.***

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Authors thank the editor and two anonymous reviewers very much for their comments on our manuscript. Their general and specific comments for revision are very valuable for the improvement of the paper. We have addressed all the comments point-by-point as below. In addition, this revised form has improved significantly in terms of grammar and the general use of the English language with the aid of native speakers (including one of our co-authors).

Referee #1 (specific comments)

1. page 8724, line 23-29, the method used to analyze OC, EC concentration is different to the normal TOT or TOR protocol, could the authors give more information on the

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comparability of this method to the normal ones? It's of concern because comparison was made in the manuscript between the data of this study and the data published earlier or data from other groups.

Response: Thanks for the nice comments. We add more information about this method in section 2.2 and Table 1. The comparison results for a NIST reference material (RM 8785) between the EnCan-total-900 and the other two widely used methods are showed in Table 1, indicating that the results from the three methods are comparable. The detailed information of this method regarding accuracy, precision and linearity are referred to our early publications (Huang et al., 2006; Chan et al., 2010). These results were obtained via a gravimetric approach. As added in the revised version, this method has been used in an international comparison activity for black carbon measurements as one of thermal/optical methods (Hammes et al., 2007) applying to the samples with high carbon mass loadings.

2. page 8727, line 7-11, the authors used the more positive stable carbon isotope value of EC in winter as an indicator of the enhanced contribution from coal combustion. And in page 8728, the authors also used stable carbon isotope value as evidence to show the decrease in gasoline and diesel consumption during the 2008 Olympic Games. It seems that the carbon isotope value of EC is quite important for the explanation of the author's findings and I think the authors have had the data in hand. I think it would be very helpful to improve the manuscript if the dataset of carbon isotope be included.

Response: Thanks for the suggestion. To properly report the isotope data, the methodology, including traceability, calibration and standard measurements, should be included together with the ambient measurements, which is beyond the scope of the current paper. It is planned that the whole  $\delta^{13}C$  dataset from TH site from 2005-2009 (one more year to be measured) will be published in another manuscript. We refer the stable carbon isotope results to Huang et al. ( $^{13}C/^{12}C$  isotopic constraints on inter-continental transport of fossil fuel CO<sub>2</sub> & BC aerosols. 2010 NOAA ESRL Global Monitoring Annual Conference, May 17-19, 2010. Boulder, Colorado, USA. Available at

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<http://www.esrl.noaa.gov/gmd/annualconference/previous/2010/pdfs/6-LinHuang.pdf>) in the revised version. Similar findings from a short-term study on winter/summer difference of stable carbon isotopes in PM<sub>2.5</sub> from Chinese cities by Cao et al. (2011) is also cited.

3. page 8728, line 28-29, the authors said “these indicate that local emissions play a more important role than regional transport for contribution to fine carbonaceous particles in Beijing”. I don’t understand why? When the urban site had similar concentration with the suburban site, one could not conclude that the local contribution was more important, maybe the opposite was also reasonable.

Response: Thanks for the comment. To make the sentence clearer, we reword it as “Furthermore, these changes and different seasonal patterns of rural/urban carbonaceous species imply that local emissions play a primary role in contribution to high fine carbonaceous loadings in urban Beijing”.

4. page 8732, line 14-16, the authors stated that “the fractions of TCM in PM<sub>2.5</sub> mass at TH and MY in 2008 were significantly less than those measured in Beijing before”. In my opinion, this is the most important conclusion of the manuscript. I think the authors should make more explanations on that. Firstly, the authors should make sure the analyzing method is not the cause of the mentioned difference. Secondly, the authors should discuss if such a trend exist in the year of 2005-2008 using their own data set.

Response: Thanks for the nice suggestion. According to the suggestion, we add the discussion on the trend of TCM in PM<sub>2.5</sub> mass during 2005-2008, which is consistent with the long-term trend. A comparison between different methods has been added as Table 1, indicating that the methods used for this and the previous studies are comparable. The fact, i.e., the relatively higher concentrations of OC and TC in NIST urban reference by EnCan-total-900 than those by IMPROVE would also strengthen the trend (discussed in the paper) instead of weaken it.

5. Comments on language usage: The English writing should be greatly improved.

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Many of the sentences are very difficult to follow in the current state. Below are some examples: 1) page 8725, line 26-27, what’s the meaning of “Each season presented both high and low OC levels at the rural site over the four study years”? 2) page 8729 line 26 to page 8730 line 2, “Not surprisingly given the large differences regarding to carbonaceous particles emissions related anthropogenic and natural activities between the paired urban and rural sites, there are striking difference in their OC and EC levels and variation patterns. In the urban area, about 80% of the energy consumption is concentrated, and the overwhelming majority of soaring population of motor vehicles in recent years (3.50 million in 2008 compared to 2.58 million in 2005) is used”. It is really a headache to read so “complicated” sentences.

Response: The revised form has improved significantly in terms of grammar and the general use of the English language with the aid of native speakers, including one of our co-authors. The sentences mentioned above are also reworded to be clear.

Referee #2 (specific comments)

1. Lines 13-15 in Abstract. The “both” is an ambiguous word, and I am not sure it means OC and EC or minimum and maxima? According to the first paragraph on Page 8726, the maxima occurred in winter. Please clarify this description.

Response: Thanks for the comment. We reword this sentence to clarify the description in the revised form.

2. Lines 19-25 on Pages 8723. About the description of the urban and rural sites, more information should be presented such as the surrounding buildings and potential pollution sources around the two sites. These are very important to identify their representativeness for local or regional status.

Response: Thanks for the nice comment. The section of “Experimental” has been re-arranged. A new sub-section has been added as sampling location to provide more information about the two sampling sites, potential emissions and their representative-

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ness.

3. The last paragraph on Page 8724. About the thermal method used in the study, a more detailed description should be given since OC and EC results are sensitive to the measurement method, and if a comparison between this method and other widely-used ones (e.g. TOT/TOR) is discussed, the data here will be more cited by and compared with other similar studies, such as those listed in Table 1 (in which the analysis methods were not pointed out).

Response: Thanks for the comments. We add more information about this method in the revised version (i.e., the comparison results for NIST RM 8785 between different methods in Table 1). Please see the response to the specific comment #1 for referee #1, which should also addressed your concerns. We has added the information about the OC&EC analysis method in Table 1 (Table 2 in the revised version).

4. Lines 22-24 on Page 8726. "... reflect the emissions resulting from space heating practices, since industrial and transportation activities are relatively constant throughout the year". This is a specious sentence and should be carefully rewritten. The amount of industrial boilers and vehicles may keep constant all the year, but their OC and EC emissions can vary greatly due to the large temperature difference between summer and winter. For example, OC and EC emission factors from cold-start engines is much higher than from hot-start ones. For OC, much more organics stay in gaseous phase in summertime than cold seasons, and you cannot differentiate this aspect because only filter samples were collected in this study.

Response: Thanks for the thorough comments. The content has been reworded to address your concern. We reword the sentence as "The peak values of OC and EC concentration in winter at the TH site are likely attributed mainly to emissions resulting from commercial/residential heating using coal as the fuel". We also add two sentences as a supplementary (i.e., "In addition to greater coal-fired heating emissions in winter, vehicular cold starts significantly increase emissions of carbonaceous particles

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and precursors (Singer et al., 1999). Lower temperatures also lead to a gas-particle equilibrium shift with more SVOCs appearing in the particle phase in winter") in the revised next paragraph.

5. Lines 7-11 on Page 8727 and Lines 14-18 on Page 8728. About the stable carbon isotope data. The explanation of more positive stable carbon values by reduction of coal consumption and vehicle amount cannot be held if corresponding data for these emission sources are not presented. For example, if coal combustion and vehicular exhaust are the dominant contributor of EC in Beijing and their stable carbon isotope are similar, the reduction of consumption in 2008 can not explain the variation of isotope composition. A recent study by Cao et al. (2011, AE, 45: 1359-1363) may be referred to this aspect. Additionally, a detailed stable carbon isotope data for EC should be presented in the manuscript.

Response: Thanks for the comments. We refer the stable carbon isotope results to Huang et al. (13C/12C isotopic constraints on inter-continental transport of fossil fuel CO<sub>2</sub> & BC aerosols. 2010 NOAA ESRL Global Monitoring Annual Conference, May 17-19, 2010. Boulder, Colorado, USA), which presents the time series of  $\delta^{13}\text{C}$  values of EC at TH and is available at <http://www.esrl.noaa.gov/gmd/annualconference/previous/2010/pdfs/6-LinHuang.pdf>. Assuming very small isotopic fractionations in combustion processes, it is generally known that  $\delta^{13}\text{C}$  values of carbonaceous species from coal combustion and from vehicular exhaust are different ( $-24 \pm 1\text{‰}$  vs.  $-28 \pm 1\text{‰}$ ). We also have some source profile measurements in  $\delta^{13}\text{C}$  of EC, supporting that there is a difference in  $\delta^{13}\text{C}$  of EC between the two major sources. Please also see the response to the specific comment #2 for referee #1 addressing the similar concerns. Similar findings from a campaign study on stable carbon isotopes in PM<sub>2.5</sub> from Chinese cities by Cao et al. (2011) is cited according to the suggestion.

6. Lines 5-6 on Page 8732. Shanghai is located in the Yangtze Delta Region in east China, not south China.

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Response: Thanks for the correction. We change 'south China' into 'east China'.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 8719, 2011.

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