

## *Response to the comments from Anonymous Referee #2*

The manuscript combined multiple-technique to study three types of haze pollution and the results are interesting and useful. This reviewer has only some minor comments for authors considering.

We thank the reviewer's hard work. We addressed the comments point by point as shown below.

Page 21721, lines 1-19 could be moved to supporting information.

Yes, thanks for the suggestion. We removed this part to the appendix.

Page 21722, lines 1-2 could be discarded since the two sentences are deviating from the main theme of discussion.

Yes, we have deleted these two sentences in the new version following the reviewer's suggestion.

Page 21722, lines 4-5, "The mass ratio of PM<sub>1</sub>/PM<sub>2.5</sub> ranged from 0.90 to 0.99, indicating particles tended to accumulated in smaller sizes." The sentence needs to rewrite because the meaning is not clear to this reviewer.

Actually, we wanted to emphasize that the aerosol mass accumulated in much smaller size, i.e., less than 1.0 μm. As most of the published research results in Shanghai and other regions of China mainly focused on PM<sub>2.5</sub> and/or PM<sub>10</sub>, the results of PM<sub>1</sub> shown in this study could shed some light on the further study of smaller particles, as particles with smaller sizes probably had more negative health effects. In the revised manuscript, we have revised this sentence to be "The mass ratio of PM<sub>1</sub>/PM<sub>2.5</sub> ranged from 0.90 to 0.99, suggesting that the mass of PM<sub>2.5</sub> was mainly dominated by smaller particles, e.g., PM<sub>1</sub>."

Page 21724, lines 1-4, the paragraph does not read well and the information presented is very negative. This reviewer believes that the authors are combining aerosol optical and chemical properties to support their classification later. This is fine, but it does not mean that the classification presented early is problematic. Rewrite.

Yes, we have revised the statement and make it more positive. This paragraph is rewritten to be "In the discussion above, the analysis of PM, pollutant gases and meteorological parameters had already shed some light on the pollution types of different episodes. And we will show more evidences of the optical and chemical properties in the discussions below."

Page 21724, lines 7-8, references are missing (e.g., Gao et al., 2011, AE, 45, 3069-3077).

Yes, we have added this reference into the revised manuscript.

Page 21725, lines 3-5, here are too speculated and more evidences are needed.

Yes, we added more quantitative analysis in Section 3.3 to support our speculation as shown below. The mean AI concentration during PE1 was  $5.48 \pm 2.52 \mu\text{g}/\text{m}^3$ , higher than the AI concentrations ( $2.51 \pm 2.52 \mu\text{g}/\text{m}^3$ ) in the winter (Huang et al., 2011, manuscript in preparation) when dust transport was almost free. Also, the fraction of mineral aerosol in TSP during PE1 reached ( $40 \pm 17\%$ ), also higher than that ( $\sim 20\%$ ) in the non-dust days in Shanghai (Fu et al., 2010). This result combined with the regional distribution of Angstrom exponent probably indicated that PE1 could be influenced by the transport of dust aerosol to some extent.

Page 21725, line 8, not “ANYWAY”, should be “However”

Yes, we corrected it following the advice of the reviewer..

Page 21725, lines 13-14, this reviewer cannot follow the logic and the potential impact of dust storm should be elaborated.

In this paragraph, we want to confirm that PE2 was indeed caused by the long-range transport of dust from the view of remote sensing technique. We have revised these sentences in the new version to be “Thus, the remote sensing analysis confirmed that PE2 was impacted by the long-range transport of dust. The dust storm had spread over almost the whole Northern China Plain and Eastern China as shown in Fig. 5d.”.

Section 2.5 is informative, but does not focus. Suggest making some revisions.

Yes, we made some revisions to make this section more focused.

Page 21735, What is the role of “The problem for the present and future would be nitrogen oxides.”? Lines 6-19 are deviating from the main theme and need to be reorganized.

The role of this statement aims to make suggestion on the future control of NO<sub>x</sub> emission for the policy makers of local government. In the future, the major source of NO<sub>x</sub> would be from vehicle emission. As shown in Figure 1 below, the increase trend of the annual tropospheric NO<sub>2</sub> in Beijing was consistent with the increase of vehicle numbers. In the eastern China, the annual tropospheric vertical columns concentration of NO<sub>2</sub> has been significantly increasing in recent years as shown in Figure 2 (Zhang et al., 2007). The vehicle numbers in Shanghai increased from 1 million in 2000 to 2.5 million in 2008, thus, the NO<sub>x</sub> emission in the future in Shanghai would increase further, which had been discussed in Section 3.5. The positive effect of mitigating SO<sub>2</sub> emission via the wide application of FGD in China would be probably offset due to the expansion of traffic system in the future. In the new version we make the statement of “The emission of nitrogen oxides would be the major pollutant to be concerned for the present and future” to emphasize the importance of mitigating the NO<sub>x</sub> emission, especially in the control of vehicle.

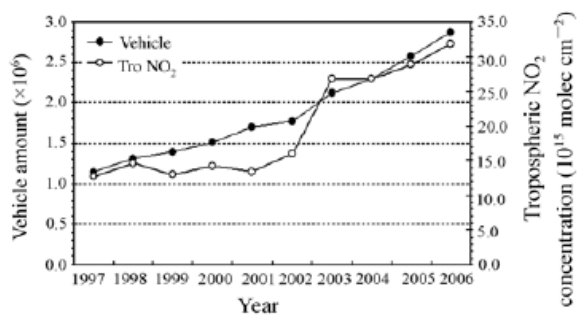


Figure 1. Annual variations of vehicle numbers and tropospheric NO<sub>2</sub> column concentrations in Beijing

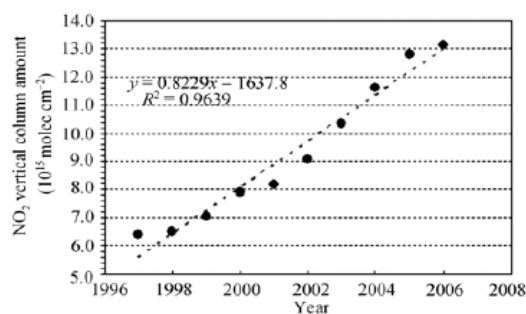


Figure 2. Annual trends of tropospheric NO<sub>2</sub> vertical column concentrations over Eastern China

Lines 6-19 mainly focused on the comparison between the results of this study and some previous research, which gave a picture of the trends of NO<sub>x</sub>. We still think that this part is in the main scope of this paper as we found that nitrate played a more and more important role in the formation of inorganic secondary aerosol. In the new version, we reorganize some paragraphs to make them more closely linked to the main theme of this paper.

Page 21738, “In this study, although there was no OC data of high time resolution available due to restrictions of instruments, we believed that CO could be regarded as proxy of organic aerosol during the biomass burning events, which indirectly linked itself to the particle formation.” This reviewer still has concern on this statement. Please consider to revise.

Yes, we revised this sentence to be “Although there was no OC data of high time resolution available due to restrictions of instruments in this study, the exclusive correlation between CO and PM<sub>2.5</sub> during the biomass burning events mirrored the dominance of organic aerosol in the particle formation. CO could be regarded as the proxy of organic aerosol when biomass burning dominated”.

Page 21738-21741, This reviewer cannot understand how Section 4 links to the main theme. Please clarify.

Yes, after we consider both of two reviewers’ opinions on this section, we decided to remove this section out in the new version of this paper and make the whole paper more linked to the main theme. Thanks a lot for the reviewer’s good suggestion.

#### References:

- Fu, Q. Y., Zhuang, G. S., Li, J. A., Huang, K., Wang, Q. Z., Zhang, R., Fu, J., Lu, T., Chen, M., Wang, Q. A., Chen, Y., Xu, C., and Hou, B.: Source, long-range transport, and characteristics of a heavy dust pollution event in Shanghai, *J. Geophys. Res.*, 115, D00K29, doi:10.1029/2009JD013208, 2010.
- Zhang, X. Y., Zhang, P., Zhang, Y., Li, X. J., and Qiu, H.: The trend, seasonal cycle, and sources of

tropospheric NO<sub>2</sub> over China during 1997-2006 based on satellite measurement, *Sci. China. Ser. D*, 50, 12, 1877-1884, 2007.