

Dear Editor:

Thank you very much for the time invested in reviewing our manuscript. We highly appreciate the professional and helpful comments for improving our work. We have revised the manuscript accordingly and the point-by-point replies to the comments are as follows.

Thanks a lot!

Sincerely

Guo-liang Shi

Replay to Report #1 (Referee #2)

Recommendation to the Editor

1) Scientific Significance

Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?

Good

2) Scientific Quality

Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?

Good

3) Presentation Quality

Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?

Good

For final publication, the manuscript should be **accepted subject to minor revisions**

(1) Line 41, delete one “%”

Response: Thanks very much for the helpful comment. The “%” had been deleted.

(Line 41)

(2) Line 41-43, three significant digits may be enough

Response: Thanks a lot. The significant digits had been revised. (Line 41-42)

(3) Line 143, please be specific-“24 hour samples”

Response: The “24 hour samples” had been specified. (Line 143)

(4) Line 255-260, “177.78 ug/m³ of PM₁₀ in winter 2000”, “227.2 ug/m³ of PM_{2.5} in 2008 winter”. It is of concern why 2000 (nearly 15 years ago) and 2008

were chosen here, and why different years are selected for PM₁₀ and PM_{2.5} respectively. Are these studies conducted during the whole winter period, for example, three months? Are the sampling in 2008 and 2000, also covered the CNY or not? If so, what's the difference in ambient PM levels during the CNY in different years?

In winter 2008, from the date you present here, why the concentration was much higher than that during the heavy-firework period, and if PM_{2.5} reached 227 ug/m³, the level of PM₁₀ would be more higher that will higher than 249.08 ug/m³ for PM₁₀ during the heavy-firework period in your study.

It is accepted that ambient PM pollution levels changed over the last several years. Therefore, it is better for the authors to compare the results from the near year like 2012-2014 of your sampling period in 2013, if you can access the data. Maybe the office data in Chinese monitoring network can be useful.

Response: Thank you very much for the very helpful suggestion. In the revised version, the concentrations of PM_{2.5} and PM₁₀ which were sampled in Tianjin during non-firework period in 2013 were listed and compared. The texts are “Additionally, the concentrations of PM₁₀ and PM_{2.5} during non-firework period were 133.30 μg/m³ and 83.98 μg/m³, which were sampled from Tianjin in March and April, 2013. Compared with non-firework period, the PM levels during light-firework period were slightly higher and those during heavy-firework period were much higher.” (Line 254-258)

(5) Line 270-275, it appears that in both light- and heavy-firework periods, the mass percents of OC in fine PM_{2.5} to that in total PM₁₀ were slightly higher, may be statistically insignificant, than those for EC. Is there any explanation as EC is often believed to preferably present in fine particle than OC? Can it be due to specific source of OC/EC during the firework burning, or mean a strong influence of secondary formation of OC in fine particle?

In addition, in the extracted profile of fireworks, did the author observe a higher percent of EC in PM_{2.5} to EC in PM₁₀, than OC?

Response: Thank you for the professional comment. In the extracted profile of fireworks, the ratio of EC in PM_{2.5} to EC in PM₁₀ (1.2) was slightly higher than the ratio for OC (1.1), which was inconsistent with that in receptor. This might imply that the strong influence of secondary formation of OC in fine particle. However, the formation of secondary particles is very complex. The reviewer gave us very important suggestion and showed us an interesting issue in the further work.

(6) Line 305, “50:50”-mass ratio?

Response: The 50:50 is the mass ratio.

Replay to Report #2 (Referee #3)

Recommendation to the Editor

1) Scientific Significance

Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?

Good

2) Scientific Quality

Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?

Excellent

3) Presentation Quality

Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?

Good

For final publication, the manuscript should be **accepted subject to minor revisions**

(1) Line 393-396. The authors explained (in Replay to Referee #3, comment 7) that information other than correlation of PM₁₀ were used when "judging the performance of PMF solutions", and revisions should be made also in the manuscript.

Response: Thanks very much for the professional and helpful comment. The texts had been added in the manuscript as "PMF was first applied to identify the possible source categories and to quantify their contributions to PM during the sampling periods. The variation in the Q values, actual condition based on the field survey, the estimated source profiles and source contributions, the correlations between measured and estimated concentrations were taken into consideration when judging the performance of PMF solutions. Finally, the five-factor solution and $F_{peak}=0.1$ were determined for fitting. The fitting plot between the measured and estimated PM concentrations was

exhibited in Fig. S10. The slope of the regression was 0.96, and the Pearson correlation coefficient was 0.98, suggesting perfect performance of PMF in this run (the estimated PM concentrations for most samples were similar to the measured concentrations).” (Line 387-396)

(2) I also suggest to explain the PMF factors using normalized profiles from table S2 as well (by incorporating it into figure 2).

Response: Thanks a lot for the helpful suggestion. The normalized profiles in Table S2 had been discussed when explaining PMF factors. “The source profiles obtained by PMF are listed in Fig. 2 and Table S2. According to Fig. 2, Factor 1 exhibited high loadings for Al, Si, Ca (0.31, 0.35 and 0.63 in normalized source profiles as shown in Table S2), etc., which are associated with crustal dust (Pant and Harrison et al., 2012). In Factor 2, relatively higher loadings of Al, Si and OC were observed. Previous studies demonstrated that simultaneously high Al, Si and OC might indicate coal combustion as the source category (Zhang et al., 2011; Pant and Harrison et al., 2012). Factor 3 correlates strongly with SO_4^{2-} and NO_3^- , consistent with source categories related to secondary particles (secondary sulphate and secondary nitrate) (Gao et al., 2011; Tian et al., 2013a). Factor 4 is mainly characterised by OC and EC (0.48 and 0.50 in normalized source profiles), which were indicative of vehicular exhaust (Pant and Harrison et al., 2012). The percentage contributions of these source categories were summarised in Fig. 2 as well.” and “In Factor 5, K^+ presented obviously high weightings. As discussed above, K^+ may be a tracer of fireworks. And the higher

loading of Mg²⁺ (0.65 in normalized source profiles in Table S2) and Cr (0.71 in normalized source profiles) in this factor might also indicate the impacts of fireworks.” (Line 397-413)