Atmos. Chem. Phys. Discuss., 14, C3980–C3986, 2014
www.atmos-chem-phys-discuss.net/14/C3980/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of direct and indirect impacts of fireworks on the physicochemical characteristics of atmospheric fine and coarse particles" *by* Y.-Z. Tian et al.

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Received and published: 23 June 2014

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 will address all the comments and show how we changed the paper accordingly. The changed manuscript will be attached in the supplement to this comment, where we highlighted the changes in the text. Furthermore, the grammar had been edited by a native English language editing service.

Replay to Referee #3

Comment: This is a nice paper studying the direct and indirect impacts of fireworks on particulate matter in a megacity in China. The study is well designed and covered the period before and after the firework episode. It provided a comprehensive analysis through a combination of chemical characterization, microscopic analysis and receptor model. The method used to determine the indirect firework is very interesting and elucidates the fact that certain PMF factor profile is a combination of co-emitted sources. Overall, the paper adds nicely to the current knowledge of impact of firework on particulate matter and merits publication. There are, though, some specific questions need to be clarified (listed below).

Response: Thank you very much for your acceptance for publication and professional comments. The comments had been addressed point by point, as shown in following.

Comment (1): The grammar needs to be edited. The title should be modified. The paper analyzed the firework impact on PM2.5 and PM10, but not "coarse particles", as shown in the title.

Response: Thanks very much. The grammar had been edited by language editing. The title had been modified as "Estimation of direct and indirect impacts of fireworks on the physicochemical characteristics of atmospheric PM10 and PM2.5". (Line 6-7 in the supplement to this comment)

Comment (2): page 11077, line 27-28. Please explain why the "inïňĆuence is continuous" given the fact of "firework-related pollution episodes are transient in nature".

Response: Thank you very much. Fireworks display is one of high-intensive anthropogenic activities that create notable air pollution and obvious air quality degradation. During firework episodes, there is usually a transient and spectacular increase of PM pollution. Fireworks display can produce a considerable quantity of pollutants (Shi et al., 2011, Wang et al., 2007; Crespo et al., 2012) and have negative effects on human health subsequently (Vecchi et al., 2008; Crespo et al., 2012). Additionally, the extremely high pollution wouldn't be eliminated transiently and its influence might be

continuous. Reference: Wang, Y., Zhuang, G.S., Xu, C., and An, Z.S.: The air pollution caused by the burning of fireworks during the lantern festival in Beijing, Atmos. Environ., 41 , 417-431, 2007. Crespo, J., Yubero, E., Nicolás, J.F., Lucarelli, F., Nava, S., Chiari, M., Calzolai, G.: High-time resolution and size-segregated elemental composition in high-intensity pyrotechnic exposures, J. Hazard. Mater., 241-242, 82-91, 2012. Shi, Y.L., Zhang, N., Gao, J.M., Li, X., Cai, Y.Q.: Effect of fireworks display on perchlorate in air aerosols during the Spring Festival, Atmos. Environ., 45, 1323-1327, 2011. Vecchi, R., Bernardoni, V., Cricchio, D., D'Alessandro, A., Fermo, P., Lucarelli, F., Nava, S., Piazzalunga, A., and Valli, G.: The impact of fireworks on airborne particles, Atmos. Environ., 42, 1121-1132, 2008.

Comment (3): page 11080, line 2-4. Only figure S2 is related to the QA/QC, and there is no "detailed information" available in the supplement. Please revise the sentence or add more information.

Response: Thank you for the helpful comment. The more information of QA/QC had been provided and highlighted in the Supplementary material of this work, which were provided in the supplement to this comment. (Line 751-792 in the supplement to this comment)

Comment (4): page 11081, line 23. As the obtained profiles of PM2.5 and PM10 were similar, is it necessary to combine them in PMF? Would it be better to combine chemical composition of PM2.5 and coarse mode (difference between PM2.5 and PM10, instead of PM10) into PMF?

Response: Thank you very much for the professional comment. As discussed in our manuscript, PM data from different sizes (PM2.5 and PM10) were combined and inputted into PMF, as done in related works (Amato et al., 2009; Aldabe et al., 2011), to enhance the number of samples into PMF. The combined data showed the satisfactory results; and further analysis demonstrated that the profiles of PM2.5 and PM10 were similar in this work, which implied that it was reasonable to combine the datasets. In

addition, in this work, the PM2.5 and PM10 were sampled synchronously and independently. In many related works, sampling champion and modeling was carried out in this way.

Comment (5): page 11082, line 6-18. Please add the aim of CMB analysis here. It is until the very end of the manuscript before I understand why and how CMB was used.

Response: Thank you very much. The aim of CMB analysis had been added in this section, as provided in supplement to this comment. (Line 199-200 in the supplement to this comment)

Comment (6): page 11088, line 9. How many samples were included in PMF? Is the number of samples sufficient comparing with the number of species?

Response: Thank you for the very helpful comment. A 50 (number of samples) $\times$ 20 (number of species) matrix was inputted into PMF. The number of samples was higher than the number of species. As discussed in the manuscript, the result of PMF is satisfactory, indicating that the number of samples might be enough. Furthermore, previous study has demonstrated that if the variances among samples are significant, it allows users to obtain physically meaningful PMF (Sun et al., 2011). Reference: Yele Sun, Qi Zhang, Mei Zheng, Xiang Ding, Eric S. Edgerton, and Xinming Wang. Characterization and Source Apportionment of Water-Soluble Organic Matter in Atmospheric Fine Particles (PM2.5) with High-Resolution Aerosol Mass Spectrometry and GC-MS. Environ. Sci. Technol. 2011, 45, 4854 – 4861.

Comment (7): page 11088, line 13. The regression between modeled and observed PM can be used to check the model, but a good correlation does not necessarily suggest "perfect performance of PMF in this run". Besides, what are the correlations for other solutions (6-factor, or 7-factor)?

Response: Thank you very much for the professional comment. The good correlations, Q values, actual condition based on the field survey, the estimated source profiles and

source contributions were all taken into consideration when judging the performance of PMF solutions. Different numbers of factors were all considered and tested to determine the best solution. For p-1 factors, one factor will contain two sources, leading to some source can't be identified; for p+1 factors, they may cause the dissociation of sources and produce more noise. In this work, when running with 6 factors, coal combustion was divided into two factors, although the regression between modeled and observed PM were good as well. For 7-factors solution, the factors were confused to be identified.

Comment (8): page 11088, line 17-25. The interpretation of the profiles rely on only the mass concentrations of a few major species. Trace metals with low concentrations are not visible at all. I suggest to include the percentage of species in one factor compared with the total concentration of that species.

Response: Thanks for the professional and helpful suggestion. The normalized source profiles (percentage of species in one factor compared with the total concentration of that species) estimated by PMF had been included in Table S2. (Table S2 in the supplement to this comment)

Comment (9): PAGE 11090; LINE 20. Potassium is used as the marker of direct fireworks in the paper, however, an "indirect biomass burning" factor was characterized using CMB, which means K is also emitted from biomass burning. The question is how certain is the "indirect biomass burning" associated with firework? Is it possible to characterize a "biomass burning" factor directly from PMF?

Response: Thank you very much for the professional comment. For source apportionment works, field survey to understand the source condition before sampling champion is very important for next sampling and modeling. The sampling period was in winter. In China, coal is used for heating in winter and other biofuel burning (like firewood) is very scarce in the non-harvesting season (Zheng et al., 2005). In this work, according to the field survey, there was few other biofuel burning (like caused by crop straw

and firewood) during the whole sampling periods; and most biomass combustion in this work occurred when the fireworks (fireworks are made by paper and cracker) are displayed and incinerated after display. In addition, PMF extracts source profiles and quantifies contributions based on the temporal variation of chemical species, so source categories in one emission pattern might be identified as one factor. In this work, direct-fireworks, resuspended dust and biomass combustion caused by fireworks might have the similar emission pattern and were extracted as one factor. Different numbers of factors were tried when running PMF, but no "biomass burning" factor was found by PMF directly, which also implies the biomass combustion was "indirect biomass burning" associated with fireworks. Reference: Zheng, M., Salmon, L.G., Schauer, J.J., Zeng, L., Kiang, C.S., Zhang, Y., Cass, G.R., 2005. Seasonal trends in PM2.5 source contributions in Beijing, China. Atmospheric Environment 39, 3967–3976.

Comment (10): Fig. 3. What are the contributions of indirect "biomass burning" from fireworks before February 9? If there were contributions, were they emitted from fireworks or from normal biomass burning sources? I am also wondering whether there were fireworks or not before February 9?

Response: Thank you very much. As mentioned above, in this work, according to the field survey, there was few other biofuel burning (like caused by crop straw and firewood) during the whole sampling periods. The contributions of indirect biomass combustion from fireworks before February 9 were also associated with fireworks. Setting off fireworks is a traditional way to celebrate the Chinese New Year (CNY, Spring Festival). Celebrations during CNY season tend to spill over to the preceding and succeeding days, along with sporadic fireworks. During the sampling periods in this work, firework displays took place for celebration of the CNY season. For the period from CNY's Eve to Lantern Festival, numerous fireworks were consumed, thus, this period is defined as heavy-firework period. For the period before the CNY's Eve (February 9), sporadic fireworks might be set off, so light-firework period is defined.

Comment (11): Fig. 4. The percentage contributions of total firework impacts to PM10

were zero from February 5-7, while the contribution to PM2.5 were between 5% - 10%. What's the reason for this?

Response: Thanks very much for the professional and helpful comment. The contributions showed in Fig. 4 were the percentage contributions (%) accounting for PM. The mass concentrations (μg/m3) of PM10 were higher than corresponding PM2.5 concentrations, so the percentage contributions to PM10 might be lower than those to PM2.5. Of course, modeling uncertainties might be another influence factor.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C3980/2014/acpd-14-C3980-2014-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 11075, 2014.