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Comment

***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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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.

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Comment: A field campaign was conducted before, during and after the Chinese New Year so as to estimate the influence of firework display on ambient air quality. PM₁₀, PM_{2.5} and chemical compositions were analyzed. Source apportionment models including PMF, PA and CMB models were used to address the contribution of firework display on ambient PM mass concentration. The study developed an interesting and also very useful approach to identify the direct and indirect impacts of firework display on air quality. The future application of the developed model could be expected, and the extracted firework profile is also valuable and expected to be used in other studies. Acceptance of the manuscript, after some revisions listed below, is recommended.

Response: Thank you very much for the acceptance of this work. All the professional and helpful comments were addressed as follows.

Comment (1): 11076, line 8, what did "them" stand for? To be specific.

Response: Thank you. The "them" stand for "K⁺ and Mg²⁺" and had been specified in the revised manuscript. (Line 32 in the supplement to this comment)

Comment (2): 11076, line 16, the mass percents here are arithmetic means or else, and what were the ranges? The sum of resuspended dust, biomass burning and direct-fireworks was >100% for PM_{2.5}. This could be related to the model bias and uncertainties. Thus, the uncertainties in model development and extracted results should be analyzed and added into the present discussion.

Response: Thanks very much for this professional suggestion. It's the arithmetic means, which was outputted by CMB model directly. For CMB model, the results could be acceptable when sum of estimated percentage source contributions (%) is in the range of 80-120%. The performance indices of CMB in this work were summarized in Table S3. The values of the performance indices met the requirement, indicating that results of CMB might be reliable. And the uncertainties in model development and extracted results had been analyzed and added into the discussion. (Line 41-43, 489-500, Table S3 in the supplement to this comment)

Comment (3): 11077, line 12, a ref is needed.

Response: Thank you. The refs had been added. (Line 68 in the supplement to this comment)

Comment (4): 11079, line 9, "size-resolved PM" here means "PM10 and PM2.5" or else? Please clarify.

Response: Thank you very much for the helpful comment. The "size-resolved PM" here means "PM10 and PM2.5" and the sentence had been revised as "The PM10 and PM2.5 samples were collected in Tianjin". (Line 125 in the supplement to this comment)

Comment (5): 11084, lines 6-10, more detailed analysis on the PM2.5/PM10 fraction is required. Did the ratio differ between the light- and heavy-firework period, and did it vary significant during the high PM level episode.

Response: Thank you very much for the helpful comment. The ratio of PM2.5/PM10 during the light- and heavy-firework periods had been provided and compared using T-test. (Line 245-249 in the supplement to this comment)

Comment (6): 11084, lines 17, also a deeper discussion on the OC EC concentrations and OC/EC ratio may be helpful to look into the difference between the light- and heavy-firework periods.

Response: Thank you very much. The discussion about OC, EC concentrations and OC/EC ratio had been added in the revised manuscript according to the helpful and professional suggestion. (Line 269-276 in the supplement to this comment)

Comment (7): 11084, in section 3.1, it is suggested to compare the levels of PM and its chemical compositions during the firework display period (light-, and heavy-firework periods in the present study) to those in other periods, if the authors had some previous studies in the non-firework period.

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Response: Thank you very much. The levels of PM and its chemical compositions during the light- and heavy-firework periods in the present study had been compared with those in winter of Tianjin in previous studies. (Line 254-260 and 278-280 in the supplement to this comment)

Comment (8): 11087, line 20, what are the concentration of nss-SO₄ during the two periods? and is the difference statistically insignificant?

Response: Thanks very much for the helpful comment. The concentrations of nss-SO₄ was 39.46 (in PM₁₀ during light-firework period), 40.19 (in PM₁₀ during heavy-firework period), 29.04 (in PM_{2.5} during light-firework period) and 31.30 (in PM_{2.5} during heavy-firework period) $\mu\text{g}/\text{m}^3$. T-test were used to analyze the difference and statistically insignificant difference were obtained with $p > 0.05$ in both PM. (Line 369-371 in the supplement to this comment)

Comment (9): 11089, line 35, delete "o" after "to"

Response: Thank you. The "o" had been deleted. (Line 432 in the supplement to this comment)

Comment (10): 11091, line 14-15, was the firework profile adopted from the cited reference comparable to that you extracted from the aforementioned PMF and PA?

Response: Thank you very much for the helpful comment. The firework profiles extracted from the PMF and PA in this work were the profiles for total influence of fireworks, including direct-fireworks emission, resuspended dust and biomass combustion. The firework profile in line 14-15 which was used in the CMB model was the profile of direct-fireworks.

Comment (11): 11091, section 3.3.3 In the CMB analysis, the total firewok profiles from PA were used to investigate the direct and indirect impacts. Did any effort to use the extracted profiles from PMF analysis, although the results from PMF and PA generated comparable profiles, but in fact not the exactly same? And, is there any difference

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between that based on PA and that based on PMF?

Response: Thank you very much. The extracted profiles from PMF for the combined dataset were also analyzed by CMB and the results were summarized as follows (Fig. A1). Very consistent results were obtained. Fig. A1 The individual percentage contributions to the total firework impacts estimated by CMB based on PMF.

Comment (12): 11092, line 13, it stated that K Mg and Cr could be good tracers of firework, but on line 17, the profile of firework was reported with higher abundance of K Al Si Ca and OC. Two concerns arisen here, the first one is it is appropriate to use K⁺ as a tracer for firework since it is widely accepted to be the tracer for the biofuel (crop straw and firewood) burning. A tracer must be unique for a specific source. The second concern is about Mg and Cr, if the level of them is very low (not high abundant species in firework profile), the use of them to distinguish firework and non-firework display periods might result in large uncertainty, Why Mg exhibit higher concentrations, but the major species did not have a higher level?

Response: Thank you very much for the professional comment. As discussed, K⁺, Mg²⁺ and Cr could be good tracers of direct-fireworks. The firework profile reported in this line was the profile for the total influence of fireworks (including direct-fireworks, resuspended dust and biomass combustion), so it was reasonable to see higher abundance of K⁺, Al, Si, Ca and OC. Through the time series of K⁺ concentrations and abundances, obvious peaks were observed at the CNY's Eve, fifth day of Chinese Lunar calendar and Lantern Festival, which were important folk-custom days for fireworks. That is to say, the variation of K⁺ was consistent with the variation of fireworks. Additionally, as shown by reviewer, K⁺ is widely accepted to be the tracer for the biofuel (crop straw and firewood) burning, however, there was few biofuel burning caused by crop straw and firewood during sampling period according to the field survey in this work. Most biomass combustion (firework is made by paper and cracker) in this work occurred when the fireworks were displayed and incinerated after display. Thus, they might be evidences that K⁺ could indicate the fireworks influence, similar with related

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literatures (Wang et al., 2007). Furthermore, as discussed in the manuscript, K^+ , Mg^{2+} and Cr could indicate the fireworks influence; however, considering too lower concentrations of Cr in the PM mass, K^+ and Mg^{2+} might be more powerful to be the tracers of fireworks. As to that Mg^{2+} is a useful metallic fuel and is also used to produce sparks and crackling stars, the concentration of Mg^{2+} is relatively higher in receptor samples, compared with Cr. Additionally, the concentration of Mg^{2+} in this factor was significantly higher than in other factors. In this work, K^+ accompanied with Mg^{2+} was used as markers to indicate the fireworks influence. 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.

Comment (13): Fig 1. in addition to the mass concentration, it is more informative to compare the normalized composition profiles between the light- and heavy-firework periods (maybe a new figure added in SI), so as to clearly indicate the higher abundance species from firework display.

Response: Thanks for the very helpful suggestion. The abundances of species and the comparison between light- and heavy-firework periods were provided in Fig. S4 in the revised supplementary material of the manuscript. (Fig. S4 and Line 345-348 in the supplement to this comment).

Comment (14): Fig 3. and also section 3.3.3, are the data in Pie chart the individual percents estimated during the heavy firework period? Is there any estimation of individual contributions during the light-firework period? And, are there any differences in the individual contributions between the light- and heavy-firework display periods.

Response: Thank you very much for the professional and helpful comment. The purpose of this work is to individually quantify the total, direct and indirect contributions of fireworks. The total influence of fireworks was obtained by Peak Analysis and PMF. Peak Analysis was used to quantify the profile of the total fireworks based on the observations of the PM and chemical species. The highest and lowest PM or

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species concentrations were used to represent the peak and background observations, respectively. The peak period had the strongest fireworks density, whereas the background values corresponded to the lowest or no fireworks density. PMF extracted the profile of total fireworks based on the variation of emission during the whole sampling periods. The total influence of the fireworks might include indirect impacts (resuspended dust and biomass combustion) and direct-fireworks. To individually determine the direct and indirect impacts of fireworks, the total firework profiles calculated by Peak Analysis and PMF were applied as the receptors into the CMB model. In summary, it's necessary to obtain the profiles of total fireworks based on the dataset including light- and heavy-firework periods, meaning that the pie charts were the average contributions for the fireworks during the sampling periods.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/14/C3972/2014/acpd-14-C3972-2014-supplement.pdf>

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

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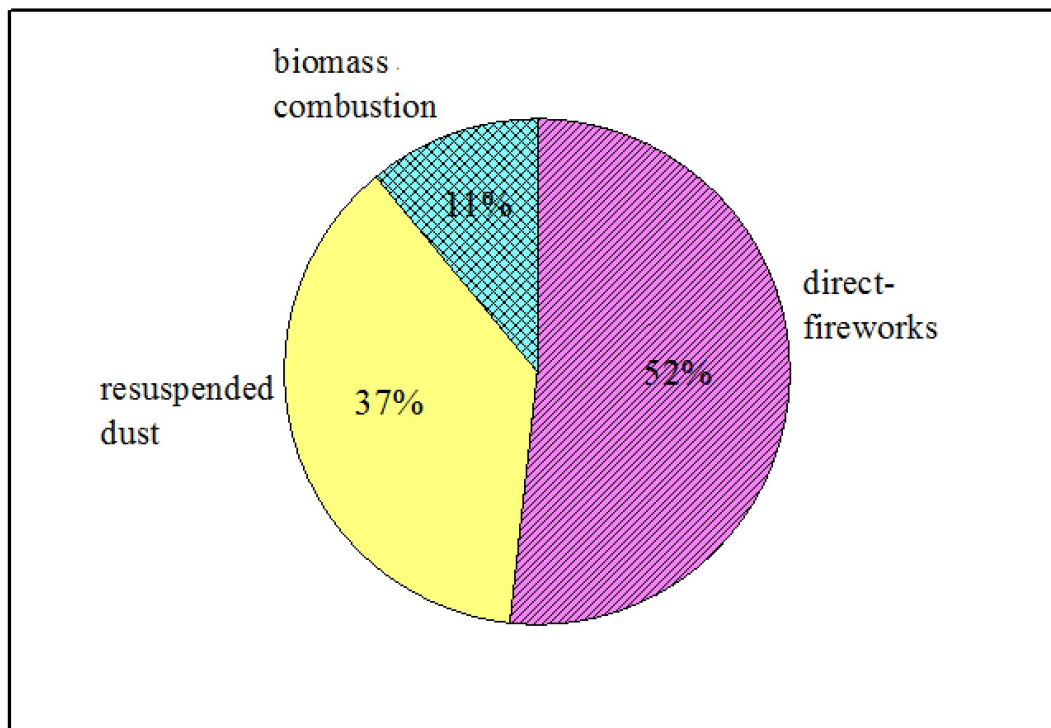
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Fig. 1. Fig. A1 The individual percentage contributions to the total firework impacts estimated by CMB based on PMF.

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