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. Furthermore, the grammar had been edited by a native English language editing service.

Replay to Referee #2

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. PM_{10}, $PM_{2.5}$ 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 Mg^{2+}” and had been specified in the revised manuscript. (Line 32)

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 PM2.5. This could be related to the model bias

and uncertainties. Thus, the uncertainties in model development and extracted results should be analzyed 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)

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

Response: Thank you. The refs had been added. (Line 68)

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 "PM_{10} and $PM_{2.5}$" and the sentence had been revised as "The PM_{10} and $PM_{2.5}$ samples were collected in Tianjin". (Line 125)

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 $PM_{2.5}/PM_{10}$ during the light- and heavy-firework periods had been provided and compared using T-test. (Line 245-249)

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)

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.

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)

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

Response: Thanks very much for the helpful comment. The concentrations of nss-SO_4^{2-} was 39.46 (in PM_{10} during light-firework period), 40.19 (in PM_{10} 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) μg/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)

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

Response: Thank you. The "o" had been deleted. (Line 432)

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 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.

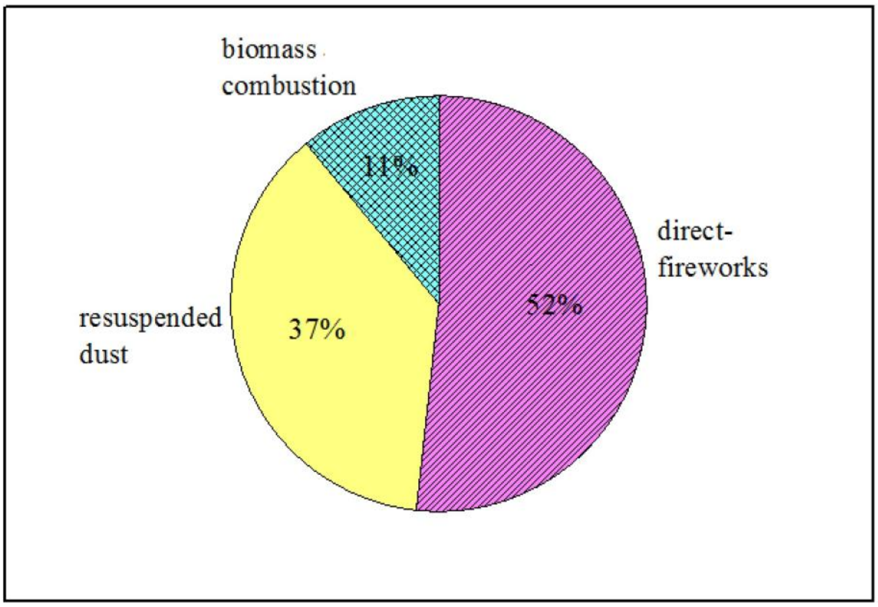

Fig. A1 The individual percentage contributions to the total firework impacts estimated by CMB based on PMF.

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 arosen here, the first one is it is

approciate 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^+, Mg^{2+} 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 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^+ companied 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-fireword 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).

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 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.

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 service. The title had been modified as "Estimation of direct and indirect impacts of fireworks on the physicochemical characteristics of atmospheric PM_{10} and $PM_{2.5}$". (Line 6-7)

Comment (2): page 11077, line 27-28. Please explain why the "influence 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 in the Supplementary material of this work. (Supplementary material)

Comment (4): page 11081, line 23. As the obtained profiles of $PM_{2.5}$ and PM_{10} were similar, is it necessary to combine them in PMF? Would it be better to combine chemical composition of $PM_{2.5}$ and coarse mode (difference between $PM_{2.5}$ and PM_{10}, instead of PM_{10}) into PMF?

Response: Thank you very much for the professional comment. As discussed in our manuscript, PM data from different sizes ($PM_{2.5}$ and PM_{10}) 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 $PM_{2.5}$ and PM_{10} were similar in this work, which implied that it was reasonable to combine the datasets. In addition, in this work, the $PM_{2.5}$ and PM_{10} 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 the revised manuscript. (Line 199-200)

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) × 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)

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 $PM_{2.5}$ 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 ($\mu g/m^3$) of PM_{10} were higher than corresponding $PM_{2.5}$ concentrations, so the percentage contributions to PM_{10} might be lower than those to $PM_{2.5}$. Of course, modeling uncertainties might be another influence factor.