

1 Date: **June 23, 2014**

2 Submitted to: Atmospheric Chemistry and Physics

3 **Changed manuscript (supplement to ACPD comments)**

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6 **Estimation of **the** direct and indirect impacts of fireworks on the**
7 **physicochemical characteristics of atmospheric **PM₁₀ and PM_{2.5}****

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24 **Abstract:**

25 To quantify the total, direct and indirect impacts of fireworks individually,
26 size-resolved PM samples were collected before, during, and after a Chinese
27 folk-custom festival (Chinese New Year) in a megacity in China. Through chemical
28 analysis and morphological characterisation, a strong influence of fireworks on the
29 physicochemical characteristics of PM₁₀ and PM_{2.5} was observed. The concentrations
30 of many species exhibited an increasing trend during the heavy-firework period,
31 especially for K⁺, Mg²⁺ and Cr; and the results of the non-sea-salt ions demonstrated
32 an anthropogenic influence on K⁺ and Mg²⁺. Then, source apportionment was
33 conducted by receptor models and Peak Analysis. The total influence of the fireworks
34 was quantified by PMF, showing that the fireworks contributed higher fractions
35 (23.40% for PM₁₀ and 29.66% for PM_{2.5}) during the heavy-firework period than
36 during the light-firework period (4.28% for PM₁₀ and 7.18% for PM_{2.5}). The profiles
37 of the total fireworks obtained by two independent methods (PMF and peak analysis)
38 were consistent, with higher abundances of K⁺, Al, Si, Ca and OC. Finally, the
39 individual contributions of the direct and indirect impacts of fireworks were
40 quantified by CMB. The percentage contributions of resuspended dust, biomass
41 combustion and direct fireworks were 36.82±8.37%, 14.08±2.82% and
42 44.44±8.26%, respectively, for PM₁₀ and 34.89±4.19%, 16.60±3.05% and
43 52.54±9.69%, respectively, for PM_{2.5}, in terms of the total fireworks. The
44 quantification of the total, direct and indirect impacts of fireworks in the ambient PM

45 gives an original contribution to understand the physicochemical characteristics and
46 mechanisms of such high-intensity anthropogenic activities.

47 *Keywords:* PM; source apportionment; fireworks; PMF; CMB; Chinese New Year

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49

50 **1 Introduction**

51 Atmospheric particulate matter (PM) is recognised globally as a major
52 environmental issue with adverse effects on air quality, regional visibility, global
53 climate change and health effects (Ding et al., 2008; Robichaud and Ménard, 2014).
54 By scattering and absorbing incoming solar radiation and outgoing terrestrial radiation
55 directly, or by acting as cloud-condensation nuclei and thereby influencing the optical
56 properties of clouds indirectly, atmospheric aerosols can influence the radiation
57 balance of the earth's atmosphere (Lin et al., 2013; Shen et al., 2013). Ambient PM is
58 a complex mixture of components from a variety of sources, including natural and
59 anthropogenic sources (Zheng et al., 2005; Zhao et al., 2013a). In recent years,
60 concerns about short-term air quality degradation events and their continuous negative
61 effects to human health has increased, especially for PM pollution caused by
62 high-intensity anthropogenic activities.

63 Firework displays are high-intensity anthropogenic activities that create notable air
64 pollution and obvious short-term air-quality degradation. Firework displays are used
65 to celebrate popular fiestas, a practice common worldwide (e.g., at the New Year).
66 During firework episodes, there is usually a transient and spectacular increase in the
67 PM pollution. Fireworks contain a variety of metal salts, such as chlorates and
68 perchlorates (Wang et al., 2007; Crespo et al., 2012), leading to extremely high
69 ambient concentrations of these species during the celebration. These heavy metals
70 and perchlorates are all highly toxic (Shi et al., 2011) and are on average fine enough
71 to be easily inhaled and show a health risk to susceptible individuals. Both the

72 long-term and short-term hazardous impacts of fireworks on human health have been
73 paid significant attention by researchers (Wang et al., 2007; Vecchi et al., 2008;
74 Crespo et al., 2012; Cheng et al., 2013).

75 **Studies have** demonstrated that **the** displacement of fireworks could be an
76 important source category **for** atmospheric PM (Vecchi et al., 2008). Fireworks **can**
77 influence the PM directly **by** emitting firework-related species (such as **certain** heavy
78 metals). **Additionally**, the accessory effects, which were indirectly caused by **the**
79 activities of firework displays, should be taken into consideration in the firework
80 events. For example, pyrotechnic device explosions can lead to resuspension of
81 materials already deposited on the ground; and biomass combustion (fireworks **are**
82 made **from** paper and **a** cracker) occurs when the fireworks are displayed and
83 incinerated after display. Although firework-related pollution episodes are transient in
84 nature, they are highly concentrated, and the influence is continuous. Both **the** direct
85 and indirect influences of fireworks might significantly contribute to PM **and the** total
86 annual metal emissions. However, the quantification of firework contributions,
87 especially for its direct and indirect impacts, is very limited.

88 Studies on fireworks **have** mainly applied the following methods: burning fireworks
89 in specific laboratories or fields to characterise their chemical properties (Tsai et al.,
90 2012); **and** investigating **the** environmental impacts through ambient sampling
91 champion during firework periods (Sarkar et al., 2010; Crespo et al., 2012). The
92 former **cannot** reflect the actual ambient condition and indirect impacts. For the latter,
93 celebrations by fireworks **are** usually continued for only a few days. Thus, it **has been**

94 very difficult to quantify **the** contributions of fireworks to ambient PM, especially for
95 **the** quantification of indirect impacts. Selecting an appropriate period and site for this
96 subject is key. Fireworks **are** more prevalent in **certain** places than others. China is the
97 **country** that produces **the most** fireworks in the world (Shi et al., 2011). **In addition**,
98 setting off fireworks is a traditional way to celebrate the Chinese New Year (CNY,
99 Spring Festival) and **is** justifiably welcomed all over the country. CNY is the most
100 important folk-custom holiday in China. Celebrations during CNY season tend to spill
101 over **into** the preceding and succeeding days (usually **until the** Lantern Festival,
102 another important festival in China), along with sporadic fireworks. During the
103 firework episodes, the anthropogenic emission patterns **are** greatly changed (Huang et
104 al., 2012). Many ordinary activities **are** decreased, such as **certain** industries and
105 traffic (Feng et al., 2012), **whereas** degradation of **the** air quality might occur due to
106 displays of fireworks. This provides a unique opportunity to study the drastic source
107 changes and allows **the** quantification of the direct and indirect contributions of
108 fireworks under significantly different emission patterns.

109 Therefore, the purpose of this work is to individually quantify the total, direct and
110 indirect contributions of fireworks to size-resolved PM. A sampling campaign of PM₁₀
111 and PM_{2.5} was **performed** before, during, and after the CNY (as shown in Table S1) in
112 a megacity in China. **The** physicochemical characteristics of **the** PM during this
113 folk-custom festival were investigated, **and** the influence of fireworks on the
114 physicochemical changes was also studied. The tracer species of fireworks were
115 discussed and then used for the source identification. The total, direct and indirect

116 contributions of fireworks to PM were modelled using the Positive Matrix
117 Factorization (PMF), Peak Analysis (PA) and Chemical Mass Balance (CMB) models.
118 The quantitative assessment of the fireworks' direct and indirect impacts on the
119 ambient PM gives an original contribution to understand the physicochemical
120 characteristics and mechanisms during firework displays. The findings will aid in
121 studies on similar high-intensity anthropogenic activities.

122

123 **2 Methodology**

124 **2.1 Sampling**

125 The PM_{10} and $PM_{2.5}$ samples were collected in Tianjin (a megacity in China).
126 Tianjin, the largest harbour of Northern China, is a fast-growing and economically
127 developed city that has a population of more than 12 million and has more than 1.5
128 million automobiles. The air quality of Tianjin has declined with rapid urbanisation
129 and industrialisation. The sampling site is sited on the rooftop of a six-story building
130 that is located in a mixed residential and commercial area in Tianjin. Usually,
131 substantial degradation would occur during the firework displays in such a mixed area.
132 The map of the sampling site was indicated in Fig. S1.

133 The sampling campaign of PM_{10} and $PM_{2.5}$ was performed from 30 January 2013 to
134 24 February 2013, including periods before, during, and after the CNY (until the
135 Lantern Festival). The sampling periods and the corresponding Chinese lunar calendar
136 were listed in Table S1. During the sampling periods, firework displays occurred for
137 the celebration of the CNY holiday. For the period from CNY's Eve to the Lantern

138 Festival, fireworks are allowed in China, and numerous fireworks were consumed;
139 thus, this period is defined as a heavy-firework period. For the period before CNY's
140 Eve, sporadic fireworks might be set off, so it is defined as a light-firework period.

141 Based on our previous works and other related studies (Shi et al., 2009; Xue et al.,
142 2010; Harrison et al., 2012; Tian et al., 2013a; Zhao et al., 2013 a, b), the PM_{2.5} and
143 PM₁₀ were simultaneously collected on quartz-fibre filters and polypropylene-fibre
144 filters using medium-volume air samplers (TH-150) at a flow rate of 100 L/min. The
145 detailed information for the sampling and quality assurance/quality control (QA/QC)
146 are available in the Supplementary Materials.

147

148 **2.2 Chemical Analysis**

149 The elemental compositions (Al, Si, Ca, V, Cr, Mn, Fe, Co, Cu, Zn, As and Pb) of
150 the samples collected on the polypropylene-fibre filters were determined by
151 inductively coupled plasma-mass spectrometry (ICP-AES) (IRIS Intrepid II, Thermo
152 Electron). Ion chromatography (DX-120, DIONEX) was used to analyse the
153 water-soluble ions (NO₃⁻, SO₄²⁻, Na⁺, K⁺ and Mg²⁺) collected on the quartz-fibre
154 filters. The organic carbon (OC) and elemental carbon (EC) concentrations of the
155 samples on the quartz-fibre filters were determined by DRI/OGC carbon analysis, a
156 technique based on the IMPROVE thermal/optical reflectance (TOR) protocol.

157 The background contamination was routinely monitored by blank tests. Enough
158 blank tests were conducted and used to valid and correct the corresponding data.

159 Certified reference materials (CRM, produced by National Research Center for

160 Certified Reference Materials, China) were used for quality assurance and quality
161 control. Blanks and duplicate sample analyses were performed for nearly 10% of the
162 samples. The pretreatment procedure, chemical analysis and QA/QC are described in
163 detail in the Supplementary Materials and refer to our previous works and other
164 related studies (Bi et al., 2007; Shi et al., 2009; Wu et al., 2009; Kong et al., 2010;
165 Xue et al., 2010; Zhao et al., 2013 a, b).

166 In addition, scanning electron microscopy (SEM) determinations were performed
167 by a JEOL JSM-7500F equipped with an X-ray energy dispersive spectrometer (EDS)
168 to perform a morphological characterisation and chemical analysis of the individual
169 particles.

170

171 **2.3 Modelling approaches**

172 **2.3.1 Receptor models**

173 Two widely used receptor models, Positive Matrix Factorisation (PMF) and
174 Chemical Mass Balance (CMB), were applied to quantify the total (sum of the direct
175 and indirect contributions), direct and indirect contributions of fireworks.

176 PMF is a useful factorisation methodology that can identify potential source
177 categories and source contributions when the source profiles are not known. It
178 identifies the source profile matrix F and quantifies the source contribution matrix G
179 based on observations at the receptor site (X). Following Paatro and Tapper (1994),
180 the PMF model can be represented in matrix form as:

$$181 \quad X = GF + E, \quad (1)$$

182 The elements of the source contribution matrix G and source profile matrix F are
183 constrained to non-negative values for PMF. PMF uses the residual matrix elements
184 (e_{ik}) and uncertainty estimates (σ_{ij}) to calculate a minimum Q value by using a
185 weighed least square method, which is defined as:

$$186 \quad Q(E) = \sum_{i=1}^m \sum_{j=1}^n (e_{ij} / \sigma_{ij})^2, \quad (2)$$

187 where σ_{ij} is the uncertainty of the j^{th} species in the i^{th} sample, which is used to weight
188 the observations that include sampling errors, detection limits, missing data, and
189 outliers (Paatero,2007). The goal of PMF is to minimise this function.

190 PM data from the different sizes (PM_{2.5} and PM₁₀) were combined and inputted into
191 PMF, as has been done in related works (Amato et al., 2009; Aldabe et al., 2011). The
192 combined data showed satisfactory results, and further analysis demonstrated that the
193 profiles of PM_{2.5} and PM₁₀ were similar in this work. Additionally, different numbers
194 of factors and different Fpeak values were considered and tested when running PMF.
195 The calculations were allowed to repeat ten times from ten pseudorandom starting
196 points for each computation, to test if a global minimum point was reached. The error
197 model code (EM=-14) and the uncertainties required for PMF were chosen according
198 to the user's guide (Paatero, 2007).

199 Then, the CMB model was applied to quantify the individual contributions from the
200 direct and indirect impacts of fireworks. CMB is also a widely used receptor model,
201 for cases in which the number and profiles of sources are available (Watson et al.,
202 1984; Chen et al., 2012). Similar to PMF, the CMB can be described as:

$$203 \quad x_{ij} = \sum_{p=1}^p g_{ip} f_{pj} + e_{ij} \quad (3)$$

204 where x_{ij} is the j^{th} species concentration measured in the i^{th} sample; f_{pj} is the j^{th} species
 205 mass fraction in the p^{th} source; g_{ip} is the contribution of the p^{th} source to the i^{th} sample;
 206 and e_{ij} is the residual (Hopke,2003). **In contrast to** PMF, except for x_{ij} , f_{pj} should also
 207 be available for **the** CMB model. USEPA CMB8.2 (USEPA, 2004) was applied in this
 208 work. The main performance indices of CMB are **the** reduced chi square (χ^2), percent
 209 mass (PM) and R square (R^2). Understanding the information of sources is important
 210 for the CMB **modelling**. In this work, **a** field survey of sources was **performed** before
 211 applying **the** CMB model, to determine the source categories.

212 **2.3.2 Peak Analysis**

213 In the present study, Peak Analysis was used to quantify the species abundances of
 214 the fireworks based on the observations of the PM and chemical species. This method
 215 was successfully applied to determine the profiles of the vehicle emissions (Ke et al.,
 216 2013). The highest and lowest PM or species concentrations were used to represent
 217 the peak and background observations, respectively. The peak period had the strongest
 218 fireworks density, **whereas the** background values corresponded to the lowest
 219 fireworks density. Then, the species abundances were obtained by **normalising** their
 220 concentrations with **the** corresponding PM concentrations, as follows (Ke et al, 2013):

$$221 \quad F_j = \frac{C_{p,j} - C_{b,j}}{C_{p,PM} - C_{b,PM}}, \quad (4)$$

222 where F_j is the abundance (g per g of PM) for the j^{th} species, $C_{p,j}$ and $C_{b,j}$ are the j^{th}
 223 species concentrations ($\mu\text{g}/\text{m}^3$) in the peak observation and the background

224 observation, respectively; and $C_{p, PM}$ and $C_{b, PM}$ are the PM concentrations ($\mu\text{g}/\text{m}^3$) in
225 the peak and the background observations, respectively.

226 The uncertainty (σ_{F_j}) of the j^{th} species abundance (F_j) was defined as follows (Ke
227 et al, 2013):

$$228 \quad \sigma_{F_j} = F_j \sqrt{\left(\frac{\sigma_{p,j}}{c_{p,j}}\right)^2 + \left(\frac{\sigma_{b,j}}{c_{b,j}}\right)^2}, \quad (5)$$

229 where $\sigma_{p,j}$ and $\sigma_{b,j}$ are the measurement uncertainties ($\mu\text{g}/\text{m}^3$) of the j^{th} species in
230 the peak observation and the background observation, respectively. The results of the
231 Peak Analysis method were employed to describe the profiles of the total fireworks.

232

233 **3 Results and discussion**

234 **3.1 Physicochemical characteristics of PM₁₀ and PM_{2.5}**

235 The PM samples were acquired on two filters for each sampling day, so consistency
236 tests play an important role in the QA/QC process. The comparisons between the
237 concentrations measured on the polypropylene-fibre filters and those on the
238 quartz-fibre filters were shown in Fig. S2. A satisfactory consistency (slopes close to
239 unity and high correlations) were observed, indicating good quality assurance.
240 Because the quartz-fibre filters tend to absorb water and be shredded during sampling
241 handling (Cheng et al., 2011), concentrations on the polypropylene-fibre filters were
242 used in the following discussion.

243 The concentrations of the PM₁₀ and PM_{2.5} in Tianjin during the sampling periods
244 are summarised in Fig. S3. The average concentration of PM₁₀ was $212.95 \mu\text{g}/\text{m}^3$, and
245 that of PM_{2.5} was $140.59 \mu\text{g}/\text{m}^3$, with an average ratio of PM_{2.5}/ PM₁₀ of 0.66. The

246 $PM_{2.5}/PM_{10}$ values were 0.65 and 0.66 during the light-firework and heavy-firework
247 periods, respectively. The T-test was used to analyse the difference between the
248 $PM_{2.5}/PM_{10}$ values in the two periods, and the results indicated an insignificant
249 difference with $p > 0.05$. The PM_{10} and $PM_{2.5}$ concentrations were $148.74 \mu\text{g}/\text{m}^3$ and
250 $96.80 \mu\text{g}/\text{m}^3$, respectively, during the light-firework period and $249.08 \mu\text{g}/\text{m}^3$ and
251 $165.23 \mu\text{g}/\text{m}^3$, respectively, during the heavy-firework period. The highest
252 concentrations were observed on CNY's Eve, when massive firework displays usually
253 occur all over the country (Feng et al., 2012), indicating the huge influence of
254 fireworks on the PM concentrations. According to the previous studies, the ambient
255 PM_{10} concentrations of Tianjin in the winter of 2000 was $177.78 \text{ug}/\text{m}^3$ on average,
256 which was higher than $148.74 \mu\text{g}/\text{m}^3$ (during light--firework periods), but lower than
257 $249.08 \mu\text{g}/\text{m}^3$ (during heavy-firework periods) (Tian et al., 2013b). The ambient $PM_{2.5}$
258 concentration of Tianjin in winter of 2008 was $227.2 \text{ug}/\text{m}^3$ on average (Kong et al.,
259 2010), which was higher than $PM_{2.5}$ levels during both light- and heavy-firework
260 periods in the present work.

261 The study of the chemical composition is critical for understanding the
262 physicochemical characteristics of pollution during the folk-custom festival. The
263 average concentrations of the chemical species in PM_{10} and $PM_{2.5}$ during the
264 light-firework period and the heavy-firework period were exhibited in Fig. 1, and the
265 abundance of the species (fractions of the species in PM) in PM_{10} and $PM_{2.5}$ were
266 summarised in Fig. S4. According to Fig. 1 and Fig. S4, the crustal elements (Al, Si,
267 Fe, and Ca), carbonaceous species (OC and EC) and several water-soluble ions (Cl,

268 NO_3^- and SO_4^{2-}) were important species in PM_{10} and $\text{PM}_{2.5}$ during the sampling
269 periods. The concentrations of OC were 9.84 and 15.58 $\mu\text{g}/\text{m}^3$ in PM_{10} and were 7.61
270 and 11.32 $\mu\text{g}/\text{m}^3$ in $\text{PM}_{2.5}$ during the light- and heavy-firework periods, respectively.
271 The concentrations of EC were 5.59 (in PM_{10} during the light-firework period), 6.79
272 (in PM_{10} during the heavy-firework period), 4.20 (in $\text{PM}_{2.5}$ during the light-firework
273 period) and 4.55 (in $\text{PM}_{2.5}$ during the heavy-firework period) $\mu\text{g}/\text{m}^3$. The ratio of
274 OC/EC was higher during the heavy-firework period for both PM_{10} and $\text{PM}_{2.5}$, with
275 values of 1.76 (light-firework) and 2.32 (heavy-firework) for PM_{10} and 1.81
276 (light-firework) and 2.49 (heavy-firework) for $\text{PM}_{2.5}$. Additionally, it was interesting
277 to find that K^+ played an important role during the heavy-firework period, in contrast
278 to its effect during the light-firework period. Compared with prior studies (Kong et al.,
279 2010; Tian et al., 2013b), several differences can be observed in chemical
280 compositions.

281 **(Fig. 1)**

282 In addition, for further characterisation, images and micrographs of the quartz-fibre
283 filters were exhibited. Fig. S5 shows photos of the quartz filters with the PM_{10} and
284 $\text{PM}_{2.5}$ samples for two cases: a normal day in the light-firework period and CNY's
285 Eve in the heavy-firework period. A difference can be observed between the filters.
286 Furthermore, Fig. S6 shows micrographs of $\text{PM}_{2.5}$ for the same days. There were
287 many more particles for the samples from CNY's Eve than for those from the normal
288 day, demonstrating the much higher concentration levels on CNY's Eve.

289

290 3.2 Influence of fireworks on **the** physicochemical characteristics of PM pollution

291 Comparing the mass concentrations and abundances of species during the
292 heavy-firework period with those during the light-firework period could help
293 researchers learn more **about** the influence of this intensive human activity. As shown
294 in Fig. 1, concentrations of most species (**such as** crustal elements, heavy metal
295 species, carbonaceous species, Cl^- , Na^+ , K^+ , and Mg^{2+}) exhibited increasing trends
296 during the heavy-firework period. **It is believed that** Al, Ca, Cr, Cu, Pb, Cl^- , Na^+ , K^+ ,
297 and Mg^{2+} might represent firework-related species. Potassium is one of the major
298 components of fireworks because potassium compounds in black powder (commonly
299 in the form perchlorate or chlorate) act as the main oxidisers during burning, with the
300 corresponding chemical equations being $2\text{KClO}_3=2\text{KCl}+3\text{O}_2$ and $\text{KClO}_4=\text{KCl}+2\text{O}_2$.
301 Ca compounds (such as **calcium** chloride and sulphate) and Cu compounds (such as
302 **copper** chloride and oxide) give rise to orange and blue colourations, respectively. A
303 Cr compound (CuCr_2O_4) is used as a catalyst for **propellants**. Cu, K, and Cr are used
304 to provide silvery and glitter effects as well. Mg is a useful metallic fuel and is also
305 used to produce sparks and crackling stars (in the form of the 50:50 Mg:Al alloy
306 magnalium). Al also can be used alone as a common constituent for fuel, sparks and
307 glitter effects. Pb can help to achieve steady and reproducible burning rates. Many
308 components are in the form of perchlorate or chlorate, leading to high concentrations
309 of Cl^- . The abruptly high emissions of these elements due to firework combustion can
310 explain **the** high concentrations of these firework-related species in atmospheric PM
311 during **the** heavy-firework period. It is noteworthy that **certain** firework-related heavy

312 metals (Cr, Pb, Cu, etc.) are dangerous elements because of their toxicity and are
313 forbidden by law in many countries. Such high concentrations in a short time,
314 especially in a place where a considerable number of people are gathered, might be of
315 concern.

316 Except for the directly firework-related species, an increase was also observed for
317 most of the crustal elements (such as Al, Si and Ca). Although Al and Ca might derive
318 from industrial and direct firework sources, the abrupt increase of crustal elements
319 might also be due to the resuspension of materials already deposited on the ground,
320 caused by pyrotechnic device explosions. Additionally, higher concentrations of K^+
321 and OC, which are good markers of biomass combustion, might imply the
322 contribution of biomass combustion during the heavy-firework period. The higher
323 concentrations of these indirect markers during the heavy-firework period can be
324 ascribed partly to the indirect influence of fireworks. The important contributions of
325 the resuspended dust and biomass combustion must be taken into consideration as
326 sources of PM in these firework events. Moreover, the mass ratios of NO_3^- to SO_4^{2-}
327 (NO_3^-/SO_4^{2-}) during the heavy-firework period were lower than the ratios during the
328 light-firework period. Similarly, lower NO_3^-/SO_4^{2-} during CNY was observed in
329 Beijing (Feng et al., 2012). The change in NO_3^-/SO_4^{2-} might be partly due to differing
330 formation mechanisms. Wang et al. (2007) reported the dominance of metal-catalysed
331 heterogeneous formation of sulphate during the firework period. Goodman et al.
332 (2001) reported a different formation mechanism for nitrate. However, the formation
333 of secondary particles might be influenced by considerable factors (such as

334 meteorological conditions and precursors) and is very complex.

335 Some of the species mentioned here require further study. SEM micrographs and
336 EDS spectra of particles on the normal day and on CNY's Eve were exhibited in Fig.
337 S7. The individual-particle analysis showed a higher K level on CNY's Eve than on
338 the normal day. Furthermore, K^+ , Cr, and Mg^{2+} particularly stand out as species that
339 have far higher concentrations during the heavy-firework period than that during the
340 light-firework period. The H/L values (the ratios of the concentrations in the
341 heavy-firework period to those in the light-firework period) of the K^+ , Cr, and Mg^{2+}
342 concentrations were 6.29, 5.52, 3.97 for PM_{10} and 5.78, 4.63, 6.32 for $PM_{2.5}$ (as shown
343 in Fig. 1), respectively. The mass concentrations may not completely reflect the
344 composition of PM, so a comparison of the species abundances was also performed.
345 As shown in Fig. S4, the abundances of K^+ , Cr, and Mg^{2+} were obviously higher
346 during the heavy-firework period, with H/L values of 3.08, 4.44, 1.78 for PM_{10} and
347 2.68, 2.06 and 2.37 for $PM_{2.5}$, respectively. The high H/L values of the abundances
348 can demonstrate the intensive influence of the fireworks on these species. For further
349 investigation, the daily variations in the concentrations and abundances of the most
350 firework-influenced species (K^+ , Mg^{2+} and Cr) were shown in Fig. S8. Similar
351 temporal patterns were observed for these species. Both the mass concentrations and
352 abundances of K^+ , Mg^{2+} and Cr showed sharp peaks for CNY's Eve for $PM_{2.5}$ and
353 PM_{10} . Obvious increases also occurred on the fifth day of the Chinese lunar calendar
354 and the Lantern Festival, which are important folk-custom days for fireworks. Thus,
355 K^+ , Mg^{2+} and Cr could indicate the influence of fireworks, which is important for

356 further identification of **the source categories of fireworks**.

357 Species in PM **may result** from anthropogenic and natural sources. To focus the
358 characteristics of anthropogenic emissions, the non-sea-salt (nss) ions were calculated.
359 Assuming that all of the Na^+ ions were from sea salt, the concentrations of nss-SO_4^{2-} ,
360 nss-Cl^- , nss-K^+ and nss-Mg^{2+} in PM_{10} and $\text{PM}_{2.5}$ were calculated based on the
361 composition of average seawater. The relative mass concentrations of SO_4^{2-} , Cl^- , K^+
362 and Mg^{2+} to Na^+ are 0.252, 1.8, 0.037 and 0.119, **respectively** (Feng et al., 2012).
363 Nearly all SO_4^{2-} and K^+ (>98%) and most Mg^{2+} (>75%) **ion** in both PM_{10} and $\text{PM}_{2.5}$
364 are from non-sea-salt. It is interesting to find a higher percentage of nss-Cl^- in $\text{PM}_{2.5}$
365 (72%) than in PM_{10} (57%), indicating the stronger influence of anthropogenic
366 emissions on **the** Cl^- in fine PM. This result is reasonable because sea salt contributes
367 **a** higher fraction to the coarse fraction of PM (Keuken et al., 2013). To investigate the
368 anthropogenic impacts of fireworks, a comparison was conducted between nss-ions
369 during the light-firework and heavy-firework periods. **The nss-SO_4^{2-} accounts for**
370 **more than 98% of the SO_4^{2-} in both the $\text{PM}_{2.5}$ and PM_{10} , and there was no obvious**
371 **difference ($p > 0.05$) during the two periods.** The concentrations of nss-Cl^- , nss-K^+
372 and nss-Mg^{2+} as well as their percentages in the total ions were exhibited in **Fig. S9**.
373 For these three ions, much higher concentrations and higher percentages occurred
374 during the heavy-firework period, especially on CNY's Eve, with **more than 88%** of
375 the ions **resulting** from anthropogenic impacts. These results suggest the large
376 anthropogenic influence on Cl^- , K^+ , Mg^{2+} , which may be mainly caused by fireworks
377 during the Chinese folk-custom festival; as a result, they were effective to indicate the

378 **presence of** fireworks. In the present work, the sources to Cl⁻ are complex, including
379 not only anthropogenic but also natural sources (Vassura et al., 2014). Additionally,
380 considering **insufficient** concentrations of Cr in the PM mass, K⁺ and Mg²⁺ may be
381 more powerful as tracers of fireworks for the following source apportionment, **as**
382 **indicated by** related reports (Wang et al., 2007; Cheng et al., 2013).

383

384 **3.3 Sources of PM**

385 To quantitatively evaluate the total, direct and indirect impacts of fireworks on
386 ambient PM, source apportionment of size-resolved PM samples was **modelled** by the
387 PMF, Peak Analysis and CMB models in this section.

388 **3.3.1 Total contributions of fireworks by PMF modelling**

389 PMF was first applied to identify the possible source categories and to quantify
390 their contributions to PM during the sampling periods. By checking the variation in
391 the Q values and the model performance, the five-factor solution and F_{peak}=0.1 were
392 determined for fitting. The fitting plot between the measured and estimated PM
393 concentrations was exhibited in **Fig. S10**. The slope of the regression was 0.96, and
394 the Pearson correlation coefficient was 0.98, suggesting perfect performance of PMF
395 in this run (the estimated PM concentrations for most samples were **similar** to the
396 measured concentrations).

397 The source profiles obtained by PMF are listed in Fig. 2 **and Table S2**. According
398 to Fig. 2, Factor 1 exhibited high loadings for Al, Si, Ca, etc., which are associated
399 with crustal dust (Pant and Harrison et al., 2012). In Factor 2, relatively higher

400 loadings of Al, Si and OC were observed. Previous studies demonstrated that
401 **simultaneously** high Al, Si and OC might indicate **coal combustion** as the source
402 category (Zhang et al., 2011; Pant and Harrison et al., 2012). Factor 3 correlates
403 strongly with SO_4^{2-} and NO_3^- , consistent with source categories related to secondary
404 particles (secondary sulphate and secondary nitrate) (Gao et al., 2011; Tian et al.,
405 2013a). Factor 4 is mainly characterised by OC and EC, which were indicative of
406 vehicular exhaust (Pant and Harrison et al., 2012). The percentage contributions of
407 these source categories were summarised in Fig. 2 as well.

408 **(Fig. 2)**

409 In Factor 5, K^+ presented obviously high weightings. As discussed above, K^+ may
410 be a tracer of fireworks, and the higher loading of Mg^{2+} in this factor might also
411 indicate the impacts of fireworks. Furthermore, it is interesting to find **the** relatively
412 higher weightings of other species, **such as** OC, Al, Si and Ca. Strong links with K^+
413 and OC could demonstrate biomass combustion (Cheng et al., 2013). Factor 5 was
414 also associated with Al, Si and Ca, which are crustal elements. Biomass combustion
415 might be indirectly caused by fireworks, which **can** occur when the fireworks are
416 displayed and incinerated after display. Crustal elements might **result** from the
417 resuspension of materials already deposited on the ground (caused by pyrotechnic
418 device explosions). Therefore, Factor 5 **represented** the total influence of fireworks,
419 which might include **the** direct-firework contribution and indirect impacts (biomass
420 combustion and resuspended dust). PMF extracts source profiles and quantifies
421 contributions based on the temporal variation of **the** chemical species, so source

422 categories in one emission pattern might be identified as one factor. In this work,
423 direct fireworks, resuspended dust and biomass combustion caused by fireworks may
424 have similar emission patterns.

425 As exhibited in Fig. 2, the total influence of fireworks, including the direct-firework
426 and indirect impacts, contributed 19.29% to PM_{10} and 24.09% to $PM_{2.5}$ during the
427 entire sampling periods. The averaged percentage source contributions to PM_{10} and
428 $PM_{2.5}$ during the light-firework and heavy-firework periods, respectively, are
429 calculated and shown in Fig. 3. A large difference can be observed. During the
430 light-firework period, the total influence of fireworks contributed 4.28% to PM_{10} and
431 7.18% to $PM_{2.5}$; while during the heavy-firework period, the total influence of the
432 fireworks increased to rather high fractions (23.40% for PM_{10} and 29.66% for $PM_{2.5}$).
433 The time series of the percentage contributions of the total firework impacts on PM_{10}
434 and $PM_{2.5}$ were exhibited in Fig. 4, which could represent the trend of the total
435 firework impacts (including direct fireworks, resuspended dust and biomass
436 combustion). The most significant peak of the total firework contributions was
437 presented on CNY's Eve, indicating the heavy impacts of the total fireworks on this
438 day, although the contributions may be overestimated due to the uncertainties in the
439 results by PMF. In most cities in China, fireworks are allowed from CNY's Eve to the
440 Lantern Festival (namely, the heavy-firework period in this work). Numerous
441 fireworks were displayed on the night of CNY's Eve, as it is the most important
442 celebration of the year. Another peak of firework contributions was observed on the
443 Lantern Festival. The Lantern Festival is also an important festival in China and is the

444 last day that fireworks are allowed. The variation in the firework contributions was
445 consistent with the Chinese folk celebrations, which demonstrate the good
446 performance of PMF for **modelling the** total firework contributions in this work.

447 **(Fig. 3)**

448 **(Fig. 4)**

449 **3.3.2 Species abundances of the total firework impacts**

450 **In addition to** PMF, Peak Analysis was also employed to better understand the total
451 firework impacts. As discussed above, the profiles and contributions of the total
452 firework impacts were determined by PMF for PM₁₀ and PM_{2.5}. Furthermore, Peak
453 Analysis (Ke et al., 2013) was employed in this section to investigate the species
454 abundances of **the** total fireworks in terms of the observations. The species
455 abundances of the total firework impacts obtained by the two independent methods
456 (PMF and Peak Analysis) were exhibited in Fig. 5. Considering the complexity
457 mentioned **previously**, the secondary ions were not included in the comparison.
458 Comparing the firework profiles, the abundances of most chemical species by Peak
459 Analysis were similar to the corresponding values by PMF. As shown in Fig. 5, the
460 abundance of K⁺, which is the main marker of direct-fireworks as discussed above,
461 were **consistent** in the three firework profiles, with values of 16.34% by PMF, 15.21%
462 by Peak Analysis for PM₁₀ and 17.33% by Peak Analysis for PM_{2.5}. The Al, Si and Ca
463 (resuspended dust elements) as well as OC (marker of biomass combustion along with
464 K⁺) **levels** were also in agreement. The abundances of Al were 5.87%, 6.72% and
465 7.02%; Si were 9.87%, 10.07% and 11.85%; and OC were 6.32%, 5.87% and 6.60%,

466 estimated by PMF, by Peak Analysis for PM₁₀ and by Peak Analysis for PM_{2.5},
467 respectively. For a further investigation of the similarity among these profiles,
468 regression analyses and correlation coefficients (R) were computed and shown in Fig.
469 5. It is visible from Fig. 5 that all of the correlation coefficients were greater than 0.9,
470 suggesting that the total firework profiles obtained by PMF and by Peak Analysis
471 were concordant. In addition, the firework profiles of PM_{2.5} were similar with those of
472 PM₁₀, implying that it is reasonable to introduce the combined dataset of PM_{2.5} and
473 PM₁₀ into PMF.

474 (Fig. 5)

475 3.3.3 Contributions of the direct and indirect firework impacts

476 As discussed above, the total influence of the fireworks might include indirect
477 impacts (resuspended dust and biomass combustion) and direct-fireworks. Thus, it is
478 necessary to deeply evaluate the individual impacts of fireworks. In this work, the
479 total firework profiles calculated by Peak Analysis were applied as the receptors in
480 the CMB model, and the source profiles of three contributors (resuspended dust,
481 biomass combustion and direct-fireworks) were incorporated into the model, to
482 individually determine the direct and indirect impacts of fireworks. In this work, the
483 source profiles of the resuspended dust were from our prior works in Tianjin (Zhang
484 et al., 2011); the biomass combustion profiles were from speciate 4.0 of the US EPA;
485 and the firework profiles were taken from a reported work (Tsai et al., 2012). The
486 performance indices of CMB in this work were summarised in Table S3. The values
487 of the performance indices met the requirement, indicating that the results of CMB

488 might be reliable.

489 The individual contributions to the total firework impacts (based on Peak Analysis)
490 were exhibited in Fig. 3 and Table S3. According to the estimations, the percentage
491 contributions of resuspended dust, biomass combustion and direct-fireworks were
492 $36.82\pm 8.37\%$, $14.08\pm 2.82\%$ and $44.44\pm 8.26\%$, respectively, for PM_{10} , accounting for
493 the total firework contribution. For $PM_{2.5}$, the percentage contributions were estimated
494 to be $34.89\pm 4.19\%$ from resuspended dust, $16.60\pm 3.05\%$ from biomass combustion,
495 and $52.54\pm 9.69\%$ from direct-fireworks. The sum of resuspended dust, biomass
496 burning and direct-fireworks were in the range of the 80-120%, which met the
497 requirement of the CMB. The results demonstrated that fireworks can lead to a
498 comprehensive influence on the ambient PM. In addition to the direct-firework
499 influence, resuspended dust and biomass combustion indirectly caused by fireworks
500 should also be considered.

501

502 **4 Conclusions**

503 To quantify the total, direct and indirect impacts of fireworks, size-resolved PM
504 samples were collected in a megacity in China. The sampling campaign covered a
505 Chinese folk-custom festival (CNY), which provides a unique opportunity to quantify
506 the contributions of fireworks under significantly different emission patterns. The
507 strong influence of fireworks on the physicochemical characteristics of atmospheric
508 PM_{10} and $PM_{2.5}$ was observed. The highest PM concentrations were observed on
509 CNY's Eve, when massive firework displays usually take place all over the country.

510 The concentrations of most species (such as crustal elements, heavy metal species,
511 carbonaceous species, Cl^- , Na^+ , K^+ , and Mg^{2+}) exhibited increased trends during the
512 heavy-firework period. Among these, K^+ , Mg^{2+} and Cr showed the most obvious
513 increase, and the results of non-sea-salt ions demonstrated the anthropogenic
514 influence on these species. K^+ , Mg^{2+} and Cr can be good tracers for fireworks,
515 especially for K^+ and Mg^{2+} , which had higher concentrations. Subsequently, source
516 apportionment was conducted using receptor models. The total influence of fireworks
517 was quantified by PMF, contributing higher fractions during the heavy-firework
518 period than those during the light-firework period. The profiles of the total fireworks
519 obtained by PMF and Peak Analysis were consistent, with higher abundances of K^+ ,
520 Al, Si, Ca and OC. Finally, the individual contributions of the direct and indirect
521 impacts of fireworks were determined by the CMB model based on profiles from
522 Peak Analysis. The present study demonstrated that fireworks might lead to a
523 comprehensive influence on the ambient PM. Both the direct influence and indirect
524 impacts (resuspended dust and biomass combustion) caused by fireworks should be
525 considered. The present work can be helpful in understanding the physicochemical
526 characteristics and mechanisms of such high-intensity anthropogenic activities.

527

528 **Acknowledgments**

529 This study is supported by the National Natural Science Foundation of China
530 (21207070 and 41375132), Special Funds for Research on Public Welfares of the
531 Ministry of Environmental Protection of China (201409003) and the Combined

532 Laboratory of the Tianjin Meteorological Bureau.

533

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Figure captions

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667

668 **Fig. 1.** The averaged concentrations of the chemical species in PM₁₀ and PM_{2.5} during
669 the light-firework period and the heavy-firework period.

670

671

672 **Fig. 2.** Source profiles ($\mu\text{g}/\text{m}^3$) and percentage source contributions (%) estimated by
673 PMF for PM₁₀ and PM_{2.5}.

674

675

676 **Fig. 3.** The total influence of fireworks on PM₁₀ and PM_{2.5} (%) during the
677 light-firework and heavy-firework periods, estimated by PMF (column chart), and the
678 individual percentage contributions to the total firework impacts estimated by CMB
679 based on Peak Analysis (pie chart).

680

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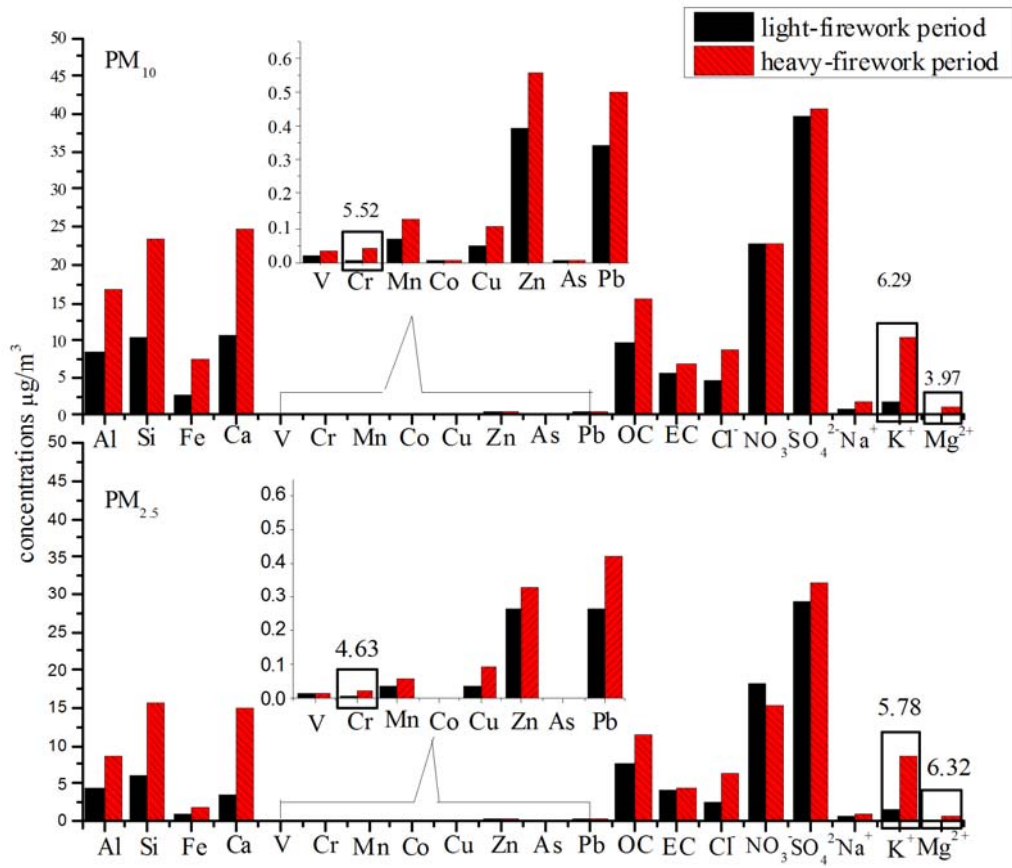
682 **Fig. 4.** The daily percentage contributions of the total firework impacts to PM₁₀ and
683 PM_{2.5}, estimated by PMF.

684

685

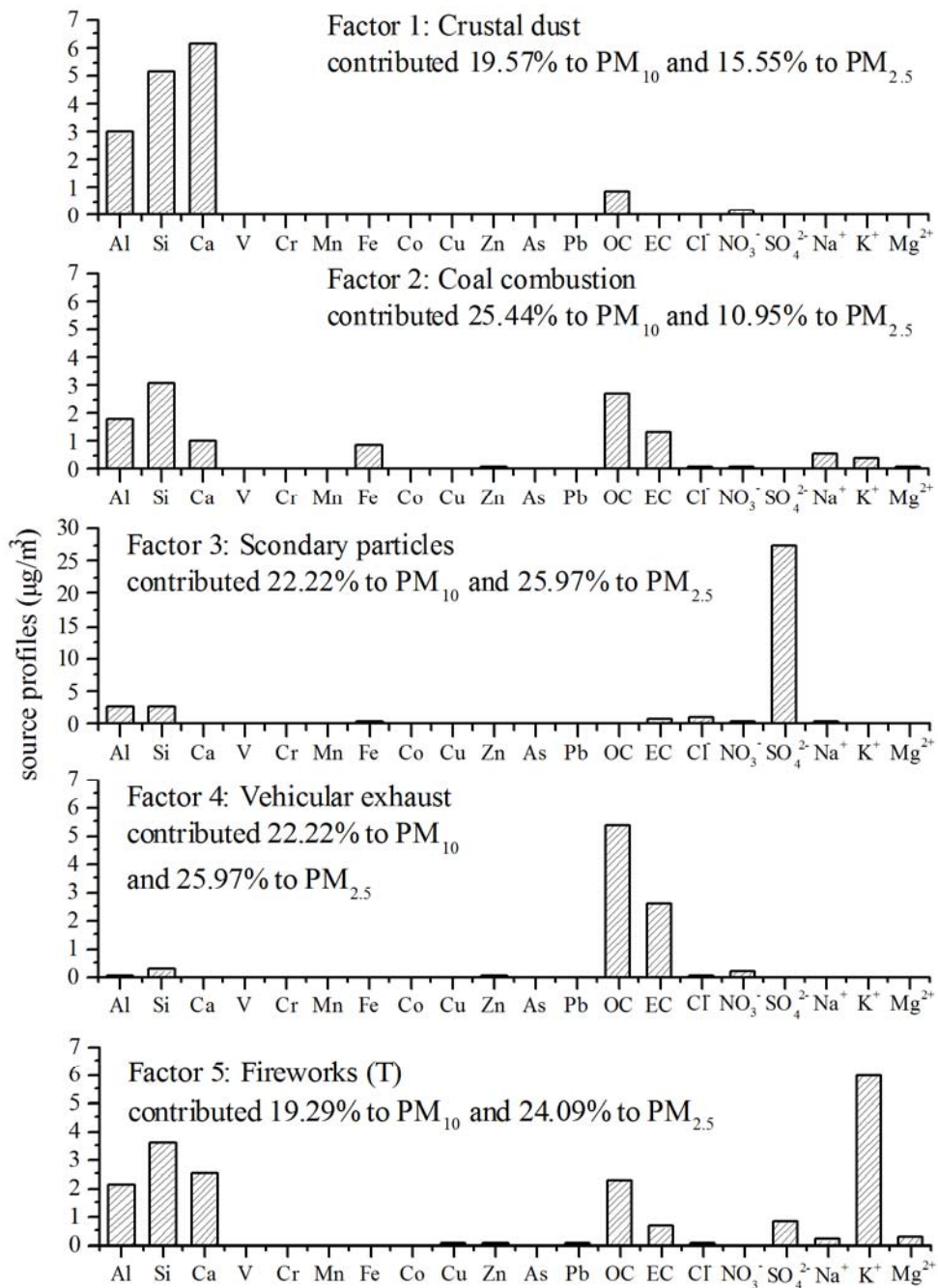
686 **Fig. 5.** Profiles of fireworks (g/g) estimated by PMF for PM₁₀ and PM_{2.5}, Peak
687 Analysis for PM₁₀ and Peak Analysis for PM_{2.5}, and the regression plots between these
688 profiles.

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Fig. 1



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Fig. 2

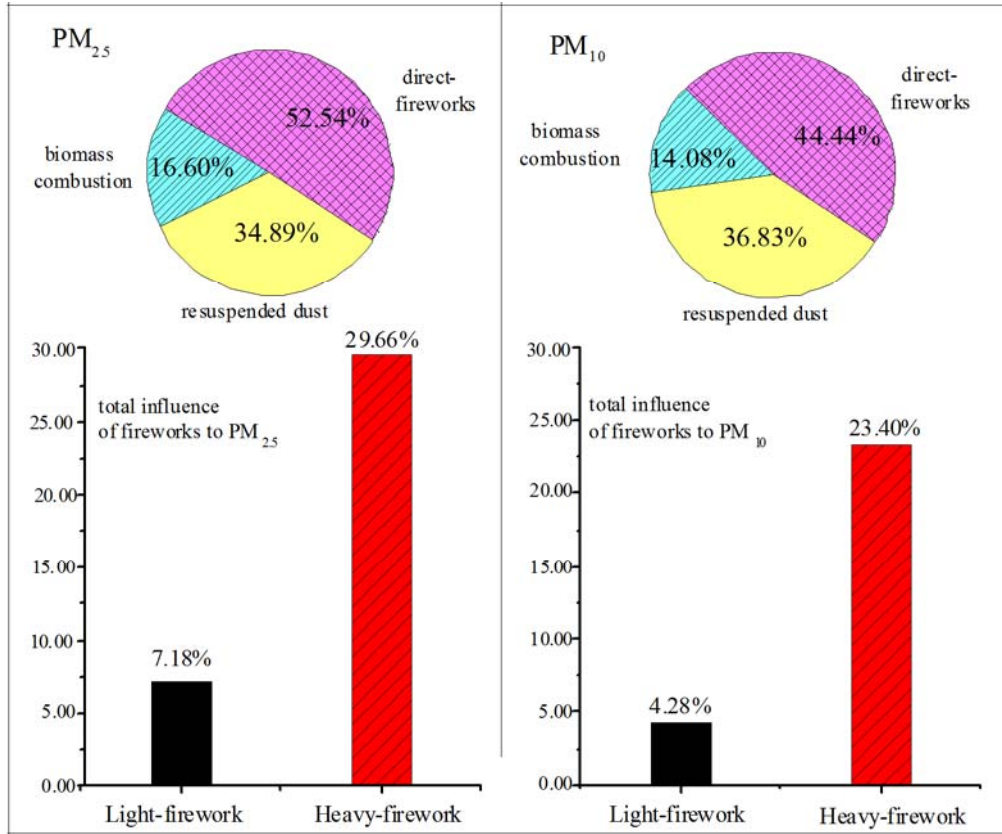
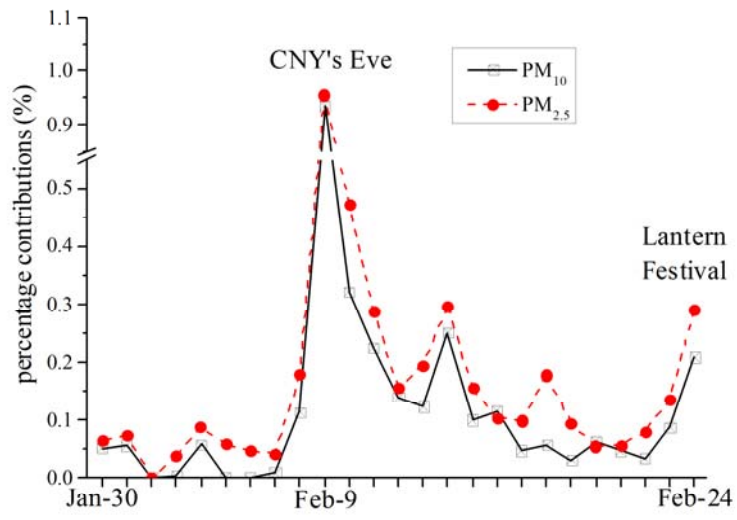


Fig. 3

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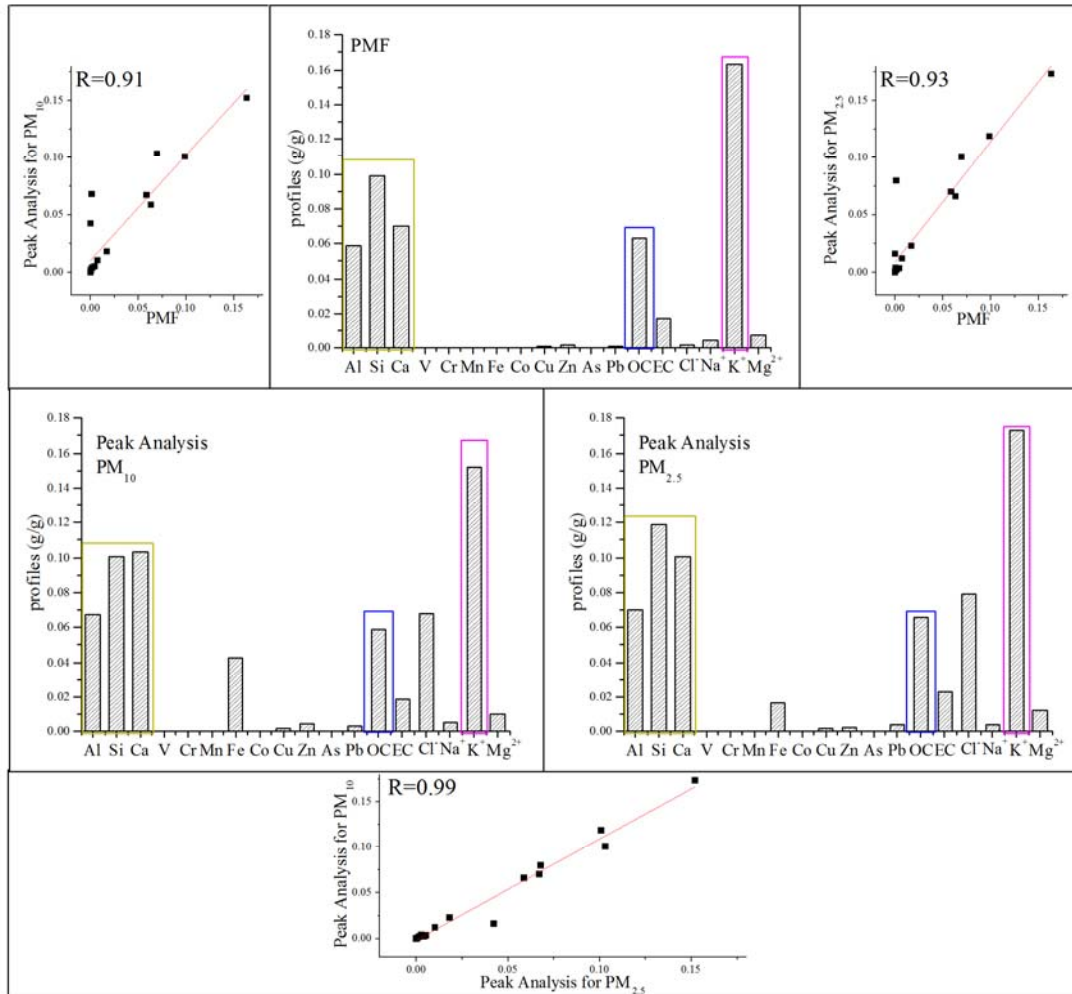


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Fig. 4



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Fig. 5

Supplementary Materials

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710 **Estimation of the direct and indirect impacts of fireworks on the**
711 **physicochemical characteristics of atmospheric PM₁₀ and PM_{2.5}**

712

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727

728 **2 Methodology**

729 **2.1 Sampling**

730 The PM_{10} and $PM_{2.5}$ samples were collected in Tianjin (a megacity in China).
731 Tianjin, the largest harbor of Northern China, is a fast-growing and economically
732 developed city who has a population of over 12 million and more than 1.5 million
733 automobiles. The air quality of Tianjin declined with rapid urbanization and
734 industrialization. The sampling site is sited at the rooftop of a six-story building which
735 is located in a mixed residential and commercial area in Tianjin. Usually, substantial
736 degradation would occur during the firework displays in such a mixed area. The map
737 of the sampling site was indicated in Fig. S1.

738 The sampling campaign of PM_{10} and $PM_{2.5}$ was carried out from 30 January 2013
739 to 24 February 2013, including periods before, during, and after CNY (till to Lantern
740 Festival). The sampling periods and the corresponding Chinese Lunar calendar were
741 listed in Table S1. During the sampling periods, firework displays took place for
742 celebration of the CNY holiday. For the period from CNY's Eve to Lantern Festival,
743 fireworks are allowed in China and numerous fireworks were consumed, thus, this
744 period is defined as heavy-firework period. For the period before the CNY's Eve,
745 sporadic fireworks might be set off, so light-firework period is defined.

746 Based on our previous works and other related studies (Shi et al., 2009; Xue et al.,
747 2010; Harrison et al., 2012; Tian et al., 2013; Zhao et al., 2013 a, b), the $PM_{2.5}$ and
748 PM_{10} were simultaneously collected on quartz fiber filters (90 mm in diameter,
749 2500QAT-UP, Pall Life Sciences) and polypropylene fiber filters (90 mm in diameter,

750 Beijing Synthetic Fiber Research Institute, China) using medium-volume air samplers
751 (TH-150) at a flow rate of 100 L/min. Before sampling, to remove any organic
752 compounds that may be present on the filters, the quartz and polypropylene fiber
753 filters were baked in the oven at 400-500 °C and 60°C, respectively. Before and after
754 sampling, all the filters were equilibrated for 48 h in desiccators at room temperature.
755 Then each filter was weighted more than 3 times by a sensitive microbalance with
756 balance sensitivity ± 0.010 mg. After weighing, the filters were stored at -4 °C until
757 chemical analysis and the chemical analysis would be conducted in less than a month.

758

759 **2.2 Chemical Analysis**

760 The elemental compositions (Al, Si, Ca, V, Cr, Mn, Fe, Co, Cu, Zn, As and Pb) of
761 the collected samples on polypropylene fiber filters were determined by inductively
762 coupled plasma-mass spectrometry (ICP-AES) (IRIS Intrepid II, Thermo Electron).
763 Before measured by ICP-AES, half of polypropylene filters were cut into fragments
764 and placed into a conical flask. Acid solutions (15 mL of HNO₃ and 5 mL of HClO₄)
765 were added into the flask and the flask was heated by electric stove. The solution was
766 evaporated until about 3 mL residual left. After being cooled and filtered, the solution
767 was decanted into a test tube and diluted to 15 mL with deionized water. Alkali
768 solution was used for measuring the concentrations of silicon. For QA/QC, standard
769 reference materials were pre-treated and analyzed with the same procedure, with the
770 recovered values for all the target elements falling into the range or within 5% of
771 certified values.

772 The ion chromatography (DX-120, DIONEX) was used to analyze the water
773 soluble ions (NO_3^- , SO_4^{2-} , Na^+ , K^+ and Mg^{2+}) collected on quartz-fiber. A 1/8 piece of
774 each quartz fiber filter was put into a glass tube and deionized water was used to
775 extract. The extraction procedure was conducted for at least three times so that the
776 water soluble ions of samples were extracted adequately into the solution. Before
777 detection of ions, standard solutions were prepared and were detected for over three
778 times; and low relative standard deviations were observed.

779 Organic carbon (OC) and elemental carbon (EC) concentrations of the samples was
780 determined by means of DRI/OGC carbon analyzers, a technique based on the
781 IMPROVE thermal/optical reflectance (TOR) protocol. The first sample was analyzed
782 again every ten samples and the precision should be less than 2%. The calibration of
783 the analyzer should be done before and after sample analysis every day.

784 Some chemical residues may be dissolved into the solution during laboratory
785 analysis. Thus, background contamination was routinely monitored through blank
786 tests. Enough blank tests were conducted and used to valid and correct corresponding
787 data. Certified reference materials (CRM, produced by National Research Center for
788 Certified Reference Materials, China) were used to ensure QA/QC. Blanks and
789 duplicate sample analyses were carried out for nearly 10% of samples. The
790 pre-treatment procedure, chemical analysis and QA/QC were referred to our previous
791 works and other related studies (Bi et al., 2007; Shi et al., 2009; Kong et al., 2010;
792 Xue et al., 2010; Zhao et al., 2013 a, b).

793 In addition, scanning electron microscopy (SEM) determinations were performed

794 by a JEOL JSM-7500F equipped with an X-ray energy dispersive spectrometer (EDS),
795 to investigate morphology characterization and chemical analysis of individual
796 particles.

797

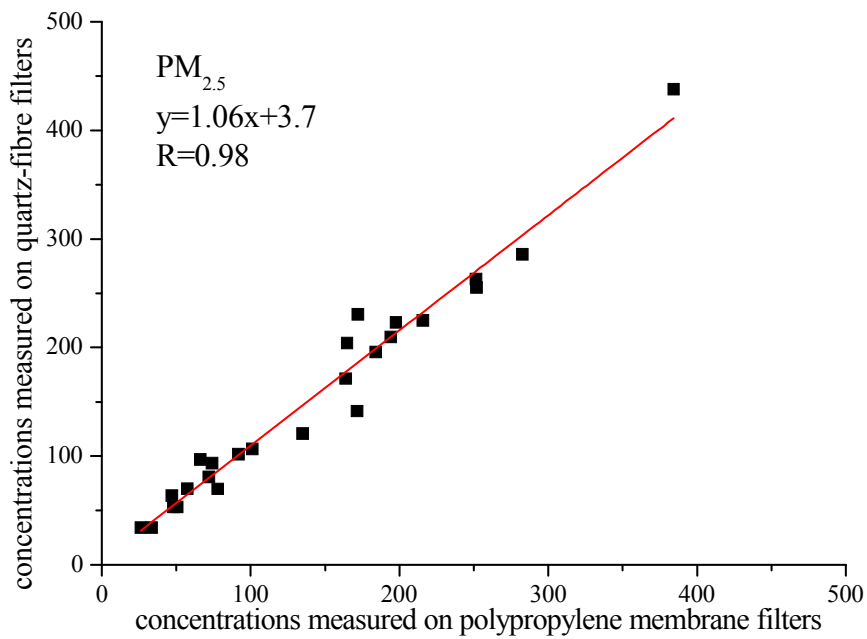
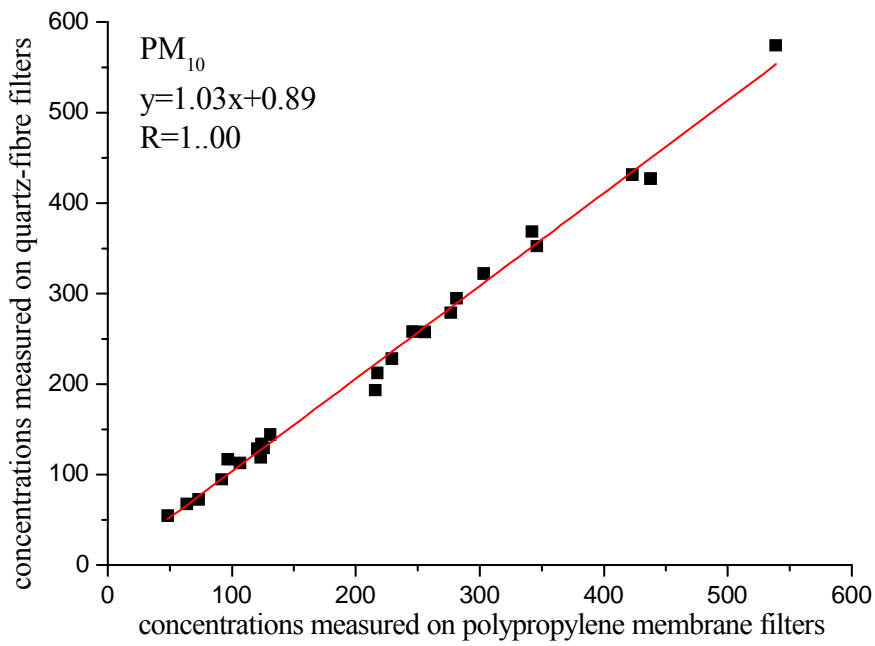


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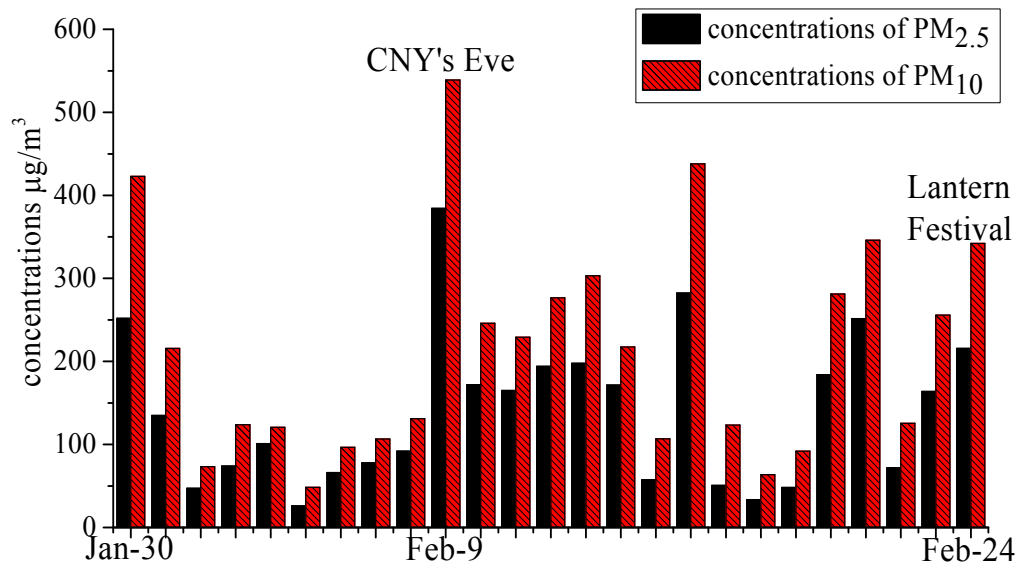
Fig. S1 The map of sampling site, in a megacity in China.

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805

Fig. S2 The comparisons between concentrations measured on polypropylene fibre filters and that on quartz fibre filters.

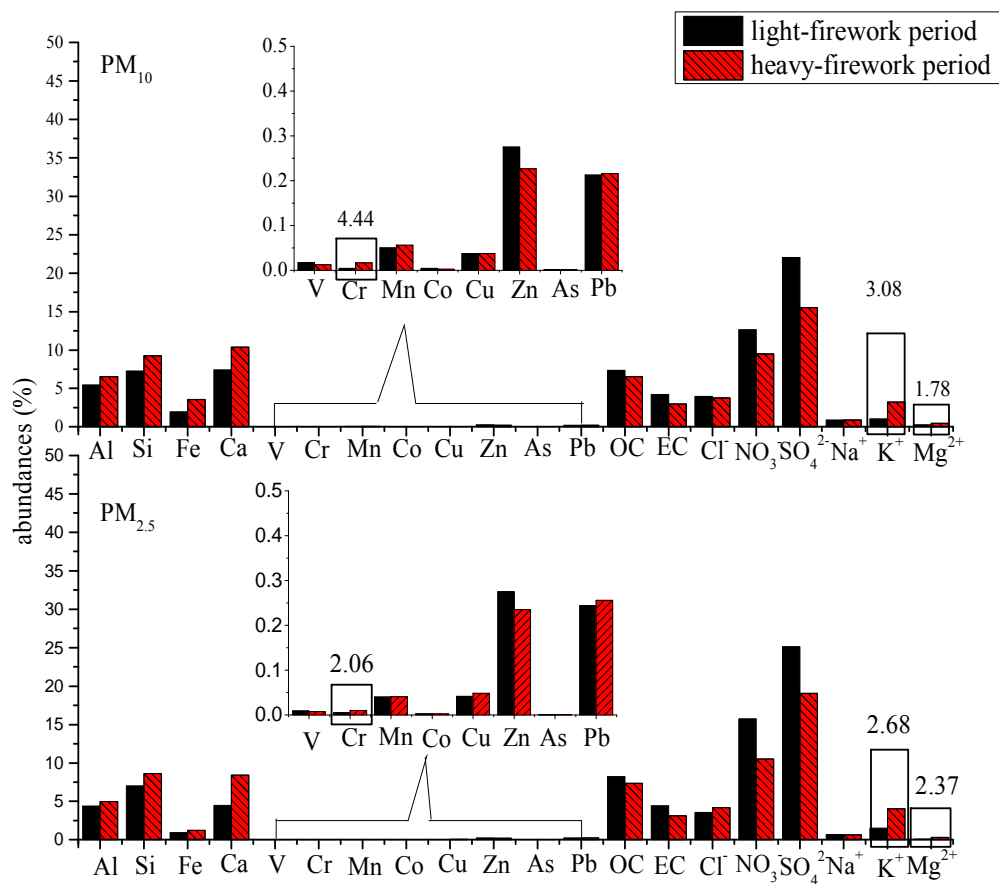


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Fig. S3 The concentrations of the PM₁₀ and PM_{2.5} in Tianjin during sampling periods

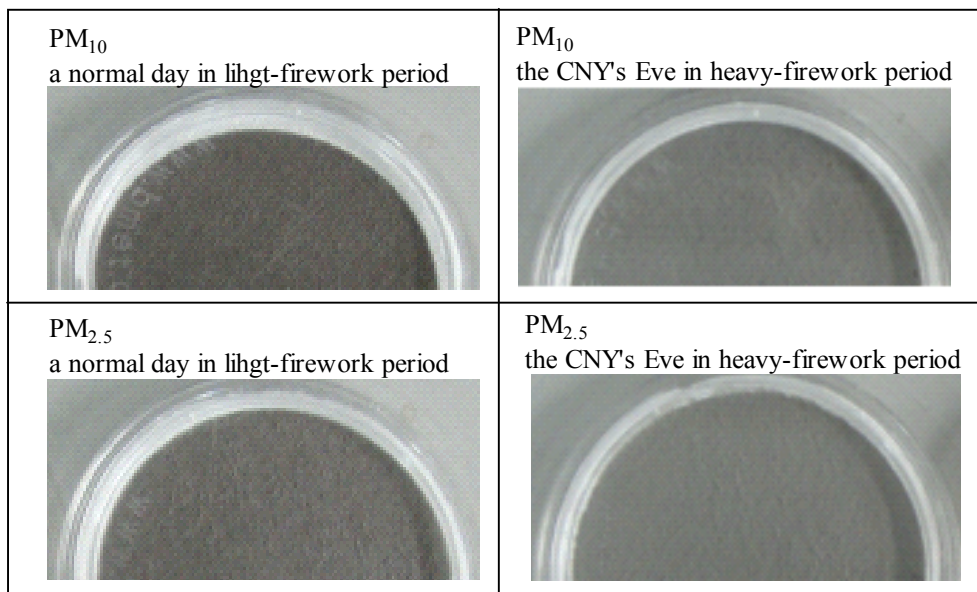


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810 **Fig. S4** The abundances of species (fractions of species in PM) in PM₁₀ and PM_{2.5}
 811 (%).

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813



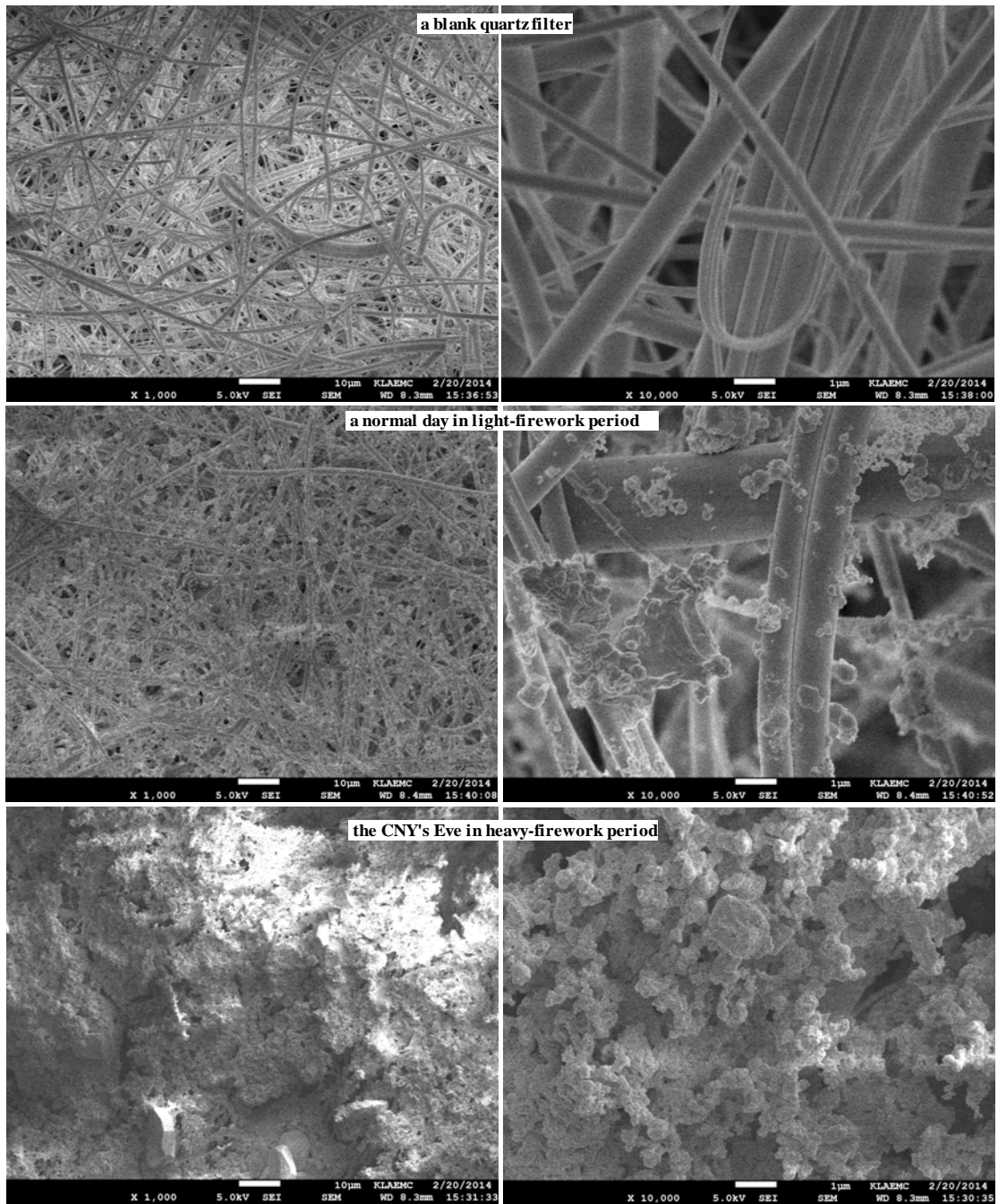
814

815 **Fig. S5** Photos of quartz filters for PM₁₀ and PM_{2.5} samples in two cases: a normal day
 816 in the light-firework period and the CNY's Eve in the heavy-firework period.

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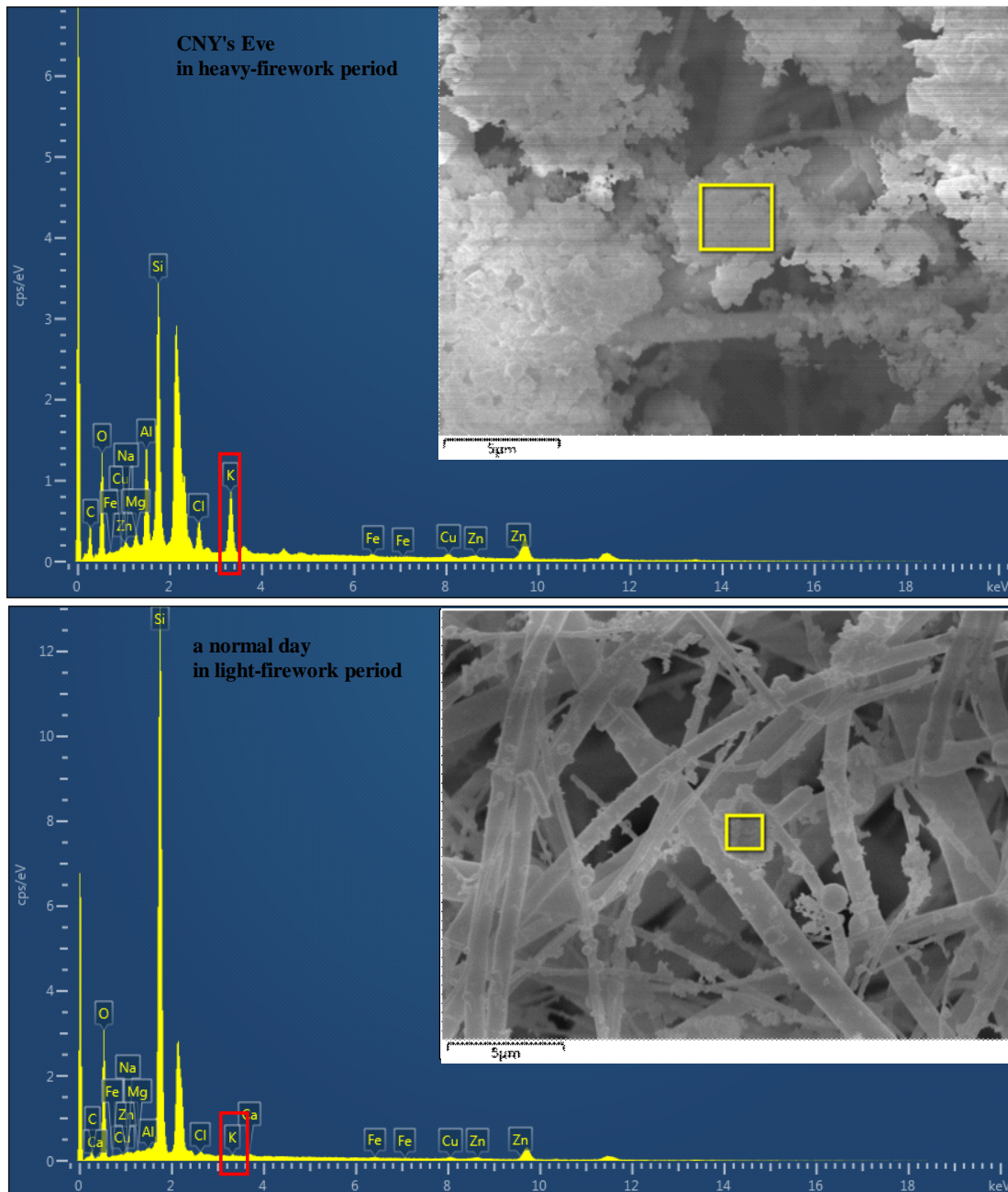
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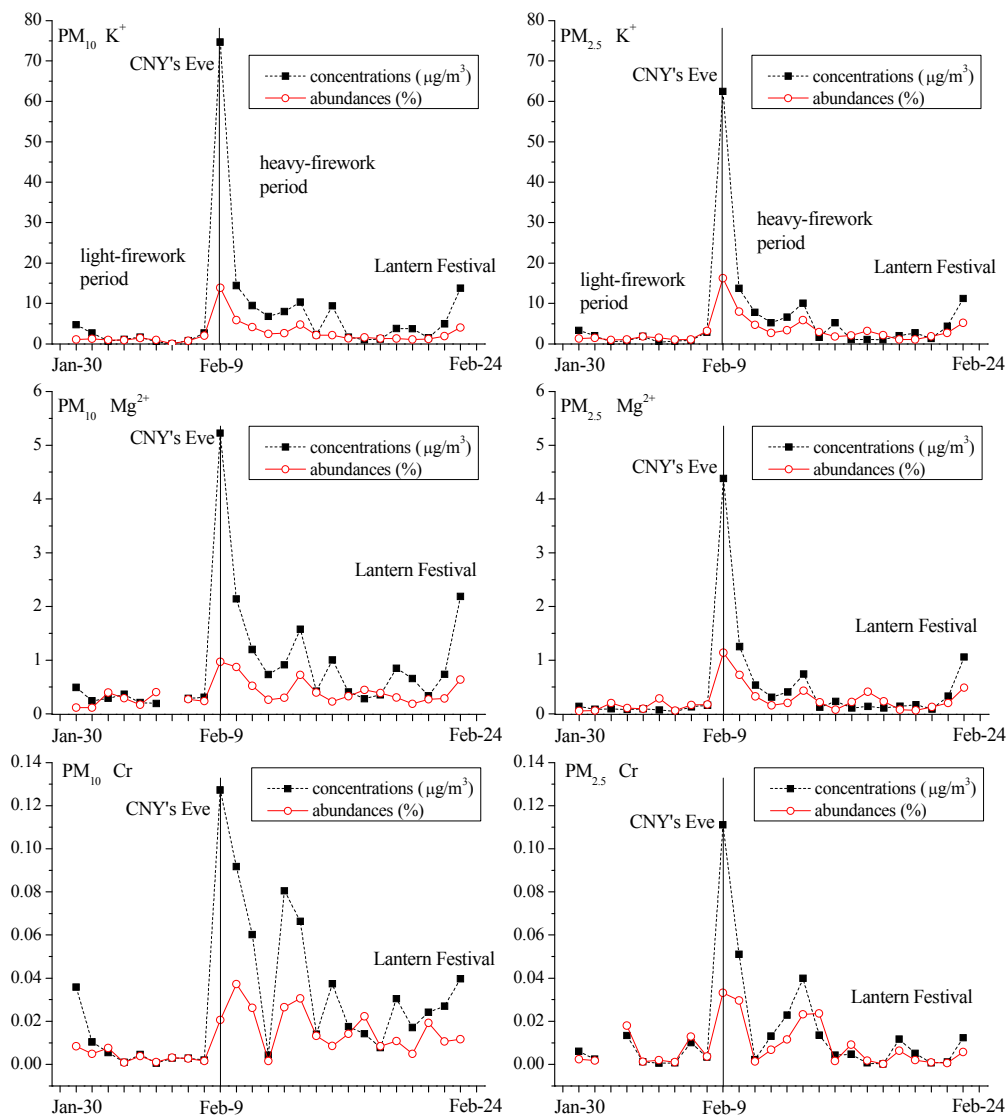
821 **Fig. S6** Micrographs of PM_{2.5} for a blank quartz filter, a normal day in light-firework
 822 period and the CNY's Eve in heavy-firework period.

823



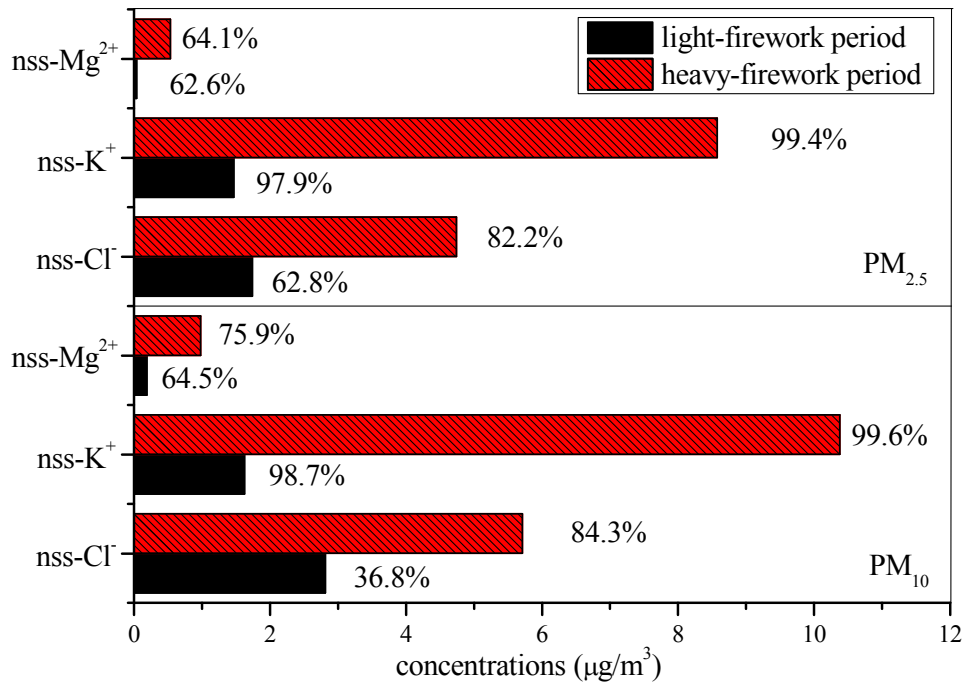
825

826 **Fig. S7** SEM micrographs and EDS spectra of particles in two cases: a normal day in
 827 the light-firework period and the CNY's Eve in the heavy-firework period.



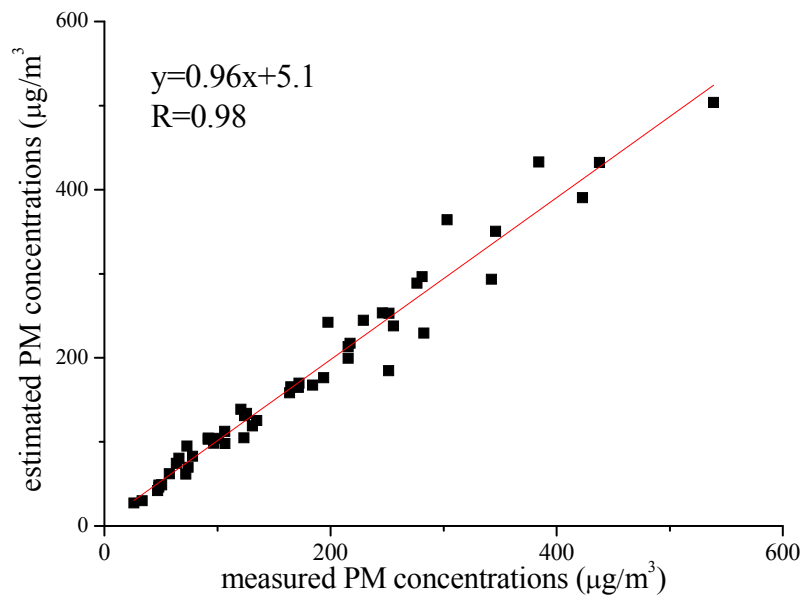
828

829 **Fig. S8** The daily variations of concentrations and abundances of the most
 830 firework-influenced species (K^+ , Mg^{2+} and Cr).



831

832 **Fig. S9** The concentrations of nss-Cl⁻, nss-K⁺ and nss-Mg²⁺ as well as their
 833 percentages in total ions.



834

835 **Fig. S10** The fitting plot between the measured and estimated PM concentrations by
836 PMF.

837 **Table S1** Sampling periods of this work.

light-firework period	Date in 2013	30 Jan	31 Jan	1 Feb	2 Feb	3 Feb	4 Feb	5 Feb	6 Feb	7 Feb	8 Feb
	Chinese Lunar calendar	19	20	21	22	23	24	25	26	27	28
heavy-firework period	Date in 2013	9 Feb	10 Feb	11 Feb	12 Feb	13 Feb	14 Feb	15 Feb	16 Feb	17 Feb	18 Feb
	Chinese Lunar calendar	CNY's Eve	CNY	2	3	4	5	6	7	8	9
	Date in 2013	19 Feb	20 Feb	21 Feb	22 Feb	23 Feb	24 Feb				
	Chinese Lunar calendar	10	11	12	13	14	Lantern Festival				

838

839 **Table S2** Normalized source profiles (the percentage of species in one factor
840 compared with the total concentration of that species) estimated by PMF.

species	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
Al	0.31	0.19	0.28	0.00	0.22
Si	0.35	0.21	0.18	0.02	0.24
Ca	0.63	0.10	0.00	0.00	0.26
V	0.00	0.71	0.00	0.12	0.17
Cr	0.28	0.00	0.00	0.00	0.71
Mn	0.05	0.42	0.23	0.12	0.18
Fe	0.04	0.60	0.36	0.00	0.00
Co	0.00	0.53	0.17	0.21	0.10
Cu	0.00	0.38	0.20	0.03	0.39
Zn	0.00	0.29	0.41	0.10	0.20
As	0.30	0.23	0.24	0.00	0.23
Pb	0.19	0.15	0.58	0.00	0.08
OC	0.08	0.24	0.00	0.48	0.21
EC	0.00	0.26	0.12	0.50	0.12
Cl ⁻	0.00	0.10	0.84	0.02	0.04
NO ₃ ⁻	0.20	0.13	0.42	0.25	0.00
SO ₄ ²⁻	0.00	0.00	0.97	0.00	0.03
Na ⁺	0.00	0.53	0.31	0.00	0.16
K ⁺	0.00	0.07	0.03	0.00	0.90
Mg ²⁺	0.05	0.30	0.00	0.00	0.65

841

842

843 **Table S3 Percentage contributions and Performance indices (R^2 , χ^2 and the percent**
 844 **mass) of CMB results.**

	percentage contributions			performance indices		
	Resuspended dust	Biomass combustion	Direct-firworks	PM	χ^2	R^2
PM ₁₀	36.82±8.37%	14.08±2.82%	44.44±8.26%	95.35	2.65	0.96
PM _{2.5}	34.89±4.19%	16.60±3.05%	52.54±9.69%	104.03	3.37	0.97

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