



中国科学院大气物理研究所

INSTITUTE OF ATMOSPHERIC PHYSICS, CHINESE ACADEMY OF SCIENCES

State Key Laboratory of Atmospheric Boundary Layer and
Atmospheric Chemistry (LAPC)
Institute of Atmospheric Physics, CAS
Beijing 100029, P.R.China
Tel: 86-10-82021255
Fax: 86-10-62021393

April 17, 2015

Editor, Atmospheric Chemistry and Physics

Dr. Yafang Cheng

Dear Dr. Cheng,

Please find in next pages our point-to-point responses to the further comments on our manuscript offered by the reviewer. Although there are still uncertainties for our PMF solution, particularly the differentiation of CCOA and HOA, it is the best solution we can get based on current available data. We thank all the reviewers for helping improve the manuscript and look forward to hearing back from you.

A marked-up manuscript version is also enclosed.

Thank you for your consideration.

Sincerely,

Yele Sun

Ph.D., Professor

State Key Laboratory of Atmospheric Boundary Layer and Atmospheric Chemistry (LAPC)

Institute of Atmospheric Physics, Chinese Academy of Sciences

Beijing 100029, China

Phone: +86-10-82021255 (Office)

Email: sunyele@mail.iap.ac.cn

We appreciate the reviewer's further comments. We have checked carefully the several points suggested by the reviewer, and addressed them in the revised manuscript. Listed below are our point-by-point responses in blue to the reviewers' comments

Response to Reviewers' comments

1) It is good that the readers can know some details about the PMF factor determination in this paper. However, I am still not totally convinced about the HOA and CCOA recognition. HOA spectrum in this study shows clear PAHs contributions, which peak around m/z 77, 91, 105, 115 (Fig S6). The PAHs are usually more abundant in the coal combustion OA (Zhang et al., 2008). The authors argued that "Wang et al. (2013) showed a large decrease of these several m/z 's when the coal was burned with a high oxygen to coal ratio." However, the f_{44} ($=m_{44}/OA$) in coal combustion OA spectrum of this paper is around 0.05, indicating this CCOA is a very fresh primary OA factor.

One explanation is fragmentation (the cleavage of C–C bonds) might be a more important mechanism than functionalization (the addition of polar, oxygen-containing functional groups) when coal is burned under conditions with higher oxygen to carbon ratios. Indeed, Wang et al. (2013) also observed low f_{44} in OA spectrum when coal was burned with higher oxygen to carbon ratios.

The author said "Note that CCOA showed a small peak at ~8:00 pm which agrees well with that of chloride. This also supports the identified CCOA factor." It is questionable that if the similar small peaks at 8:00 pm in the diurnal variation of CCOA and chloride are caused by the enhanced concentrations of both species in firework period during Lantern Festival. Diurnal variations excluding three FW periods should be examined. And also it seems that the time series of CCOA and HOA are very similar.

We further checked the diurnal cycles of CCOA and chloride by excluding FW events (Figure R1). The reviewer is right that the FW events had a considerable impact on diurnal cycle of chloride between 18:00 – 24:00. The chloride peak at 20:00 is much smaller after excluding FW events. Therefore, our previous explanation without strong evidence was deleted. The time series of CCOA and HOA was similar. Similar time series of HOA and CCOA was also observed in our previous study (Sun et al., 2013).

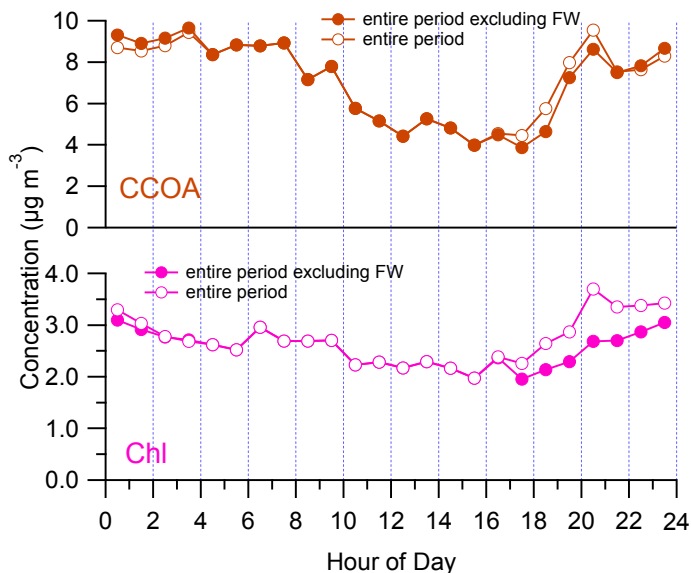


Figure R1. Diurnal cycles of CCOA and chloride for the entire period with and without FW events

If the PMF solution presented in this paper now is the best one that the authors can get, it is fine. However, please consider to address the questions above and avoid confusing.

We agree with the reviewer that there are still uncertainties in our PMF solution. However, it is the best solution we can get based on current available data.

2) In summary, the conclusion that the authors said in line 297 (“Considering above, the contributions estimated in this work would represent the upper limits of FW.”) is invalid and vague. The regional transport is not a reason that makes the estimation of FW contribution an upper limit, or it depends on the upwind air masses. The role of regional transport contribution to the absolute mass conc. and fraction of FW aerosol to the total OA in different FW periods should be fully considered.

We thank the reviewer for careful evaluating the conclusion and we agree with the reviewer. Because of the absence of data for accurate quantification of regional transport, we deleted this sentence in the revised manuscript to avoid misleading readers.

3) Line 319: “Therefore, our results might suggest that a large fraction of aerosol particles from the burning of fireworks was emitted in the size range of 1 – 2.5 µm.” Have the authors considered coagulations and VOCs condensation on the aerosol phase during FW period? It’s a good point. There’s possibility that the particles in the size range of 1 – 2.5 µm were partly from the coagulations and VOCs condensation. Because the particle number size distributions and VOCs data were not available in this study, it is difficult to evaluate the role of coagulation and condensation in the formation of large particles (1 – 2.5 µm).

4) Line 390 in revised paper: “contribution of POA increased as a function of organic loadings which varied from ~35% to 63% when organics was above 80 µg m⁻³ (Fig. 7c).” The Fig. 7c only show data below 60 µg m⁻³, No data above 80 µg m⁻³ are shown.

We thank the reviewer's carefulness. This sentence should be "when organics reached $80 \mu\text{g m}^{-3}$ (Fig. 7c)". It was corrected in the revised manuscript.

5) One suggestion about holiday periods vs non-holiday periods in Fig.9. Have the authors calculated if the O₃ increase in the holiday periods can be fully explained by NO₂ deduction. If not, then the extra O₃ enhancement in the holiday period may indicate the reduction of VOCs from anthropogenic sources in holiday periods as well.

This is a good suggestion. The average mixing ratio of NO during HD and NHD is 19.2 and 42.0 ppb, respectively, and that of NO₂ is 33.3 and 45.2 ppb, respectively. The reduction of 22.8 ppb and 11.9 ppb for NO and NO₂ are much higher than 5.9 ppb of O₃ that was increased during HD. This result suggests that the increase of O₃ during HD can be fully explained by the deduction of NO.

References:

Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.: Aerosol composition, sources and processes during wintertime in Beijing, China, *Atmos. Chem. Phys.*, 13, 4577-4592, 10.5194/acp-13-4577-2013, 2013.

1 Aerosol Composition and Sources during the Chinese Spring
2 Festival: Fireworks, Secondary Aerosol, and Holiday Effects

3
4 Qi Jiang^{1,3}, Yele Sun^{1,2*}, Zifa Wang¹, Yan Yin^{2,3}

5
6 ¹State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric
7 Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing
8 100029, China

9 ²Collaborative Innovation Center on Forecast and Evaluation of Meteorological
10 Disasters, Nanjing University of Information Science & Technology, Nanjing, 210044,
11 China

12 ³Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administra
13 tion, Nanjing University of Information Science & Technology, Nanjing 210044, China

14
15
16
17 Correspondence to: sunyele@mail.iap.ac.cn

18 **Abstract**

19 Aerosol particles were characterized by an Aerodyne Aerosol Chemical Speciation
20 Monitor (ACSM) along with various collocated instruments in Beijing, China to
21 investigate the roles of fireworks (FW) and secondary aerosol in particulate pollution
22 during the Chinese Spring Festival of 2013. Three fireworks events exerting significant
23 and short-term impacts on fine particles ($PM_{2.5}$) were observed on the days of Lunar
24 New Year, Lunar Fifth Day, and Lantern Festival. The FW showed large impacts on
25 non-refractory potassium, chloride, sulfate, and organics in submicron aerosol (PM_1),
26 of which the FW organics appeared to be emitted mainly in secondary with its mass
27 spectrum resembling to that of secondary organic aerosol (SOA). Pollution events (PEs)
28 and clean periods (CPs) alternated routinely throughout the study. Secondary
29 particulate matter ($SPM = SOA + \text{sulfate} + \text{nitrate} + \text{ammonium}$) dominated the total
30 PM_1 mass on average accounting for 63-82% during nine PEs in this study. The
31 elevated contributions of secondary species during PEs resulted in a higher mass
32 extinction efficiency of PM_1 ($6.4 \text{ m}^2 \text{ g}^{-1}$) than that during CPs ($4.4 \text{ m}^2 \text{ g}^{-1}$). The Chinese
33 Spring Festival also provides a unique opportunity to study the impacts of reduced
34 anthropogenic emissions on aerosol chemistry in the city. The primary species showed
35 ubiquitous reductions during the holiday period with the largest reduction for cooking
36 OA (69%), nitrogen monoxide (54%), and coal combustion OA (28%). The secondary
37 sulfate, however, remained minor change, and the SOA and the total $PM_{2.5}$ even slightly
38 increased. Our results have significant implications that controlling local primary
39 source emissions during PEs, e.g., cooking and traffic activities, might have limited
40 effects on improving air quality in megacity Beijing due to the dominance of SPM from
41 regional transport in aerosol particle composition.

42 1 Introduction

43 Air pollution caused by fine particles (PM_{2.5}) is of great concern in densely
44 populated megacities because of its adverse effects on human health and regional air
45 quality (Molina and Molina, 2004; Chan and Yao, 2008). The health risk of air
46 pollution is greater than expected leading to around 7 million people's death in 2012
47 according to the latest report by World Health Organization
48 (<http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/>). The Beijing
49 metropolitan area is one of the most populous megacities in the world with the
50 population reaching 20.69 million by the end of 2012 (Beijing Municipal Bureau of
51 Statistics). According to Beijing Municipal Environmental Protection Bureau, the
52 annual average concentration of PM_{2.5} was 89.5 µg m⁻³ in 2013, which is about 2.5
53 times the National Ambient Air Quality Standards of China (35 µg m⁻³ for annual
54 average). This suggests severe fine particle pollution in Beijing. Extensive studies
55 have been made recently to investigate the chemical composition and sources of PM_{2.5}.
56 The results showed that secondary inorganic aerosol (SIA = sulfate + nitrate +
57 ammonium), coal combustion, traffic emissions (gasoline and diesel), biomass
58 burning, cooking emissions and dust are the major sources of PM_{2.5} (Zheng et al.,
59 2005; Song et al., 2006; Zhang et al., 2013). However, the source contributions varied
60 significantly among different seasons, therefore improving air quality in Beijing
61 remains a great challenge due to the very complex sources and dynamic evolution
62 processes of aerosol particles.

63 Fine particles from various sources can be either primary from direct emissions,
64 e.g., fossil fuel combustion and biomass burning, or secondary from atmospheric
65 oxidation of gas-phase species. The fireworks (FW) is one of the most important

66 primary sources that can exert significant and short-time impacts on air quality. The
67 fireworks burning emits a large amount of gaseous pollutants, e.g., sulfur dioxide
68 (SO_2) and nitrogen oxide (NO_x) (Vecchi et al., 2008;Huang et al., 2012), and also fine
69 particles comprising organic/elemental carbon, sulfate, potassium, chloride and
70 various metals, e.g., copper (Cu), barium (Ba), strontium (Sr) and magnesium (Mg)
71 (Moreno et al., 2007;Wang et al., 2007;Li et al., 2013). The enhanced short-term air
72 pollution by fireworks can substantially increase health risk levels (Godri et al.,
73 2010;Yang et al., 2014) and reduce visibility for hours (Vecchi et al., 2008). Previous
74 studies on chemical characterization of fireworks in China were mostly based on filter
75 measurements with a time resolution of 12 h or 24 h (Wang et al., 2007;Zhang et al.,
76 2010;Feng et al., 2012;Huang et al., 2012;Cheng et al., 2014;Zhao et al., 2014).
77 Considering that the fireworks events usually last less than 12 hours, the filter analysis
78 may introduce large uncertainties in accurate quantification of chemical composition
79 of FW particles due to either the interferences of non-FW (NFW) background
80 aerosols or the difficulties to account for meteorological variations. Drewnick et al.
81 (2006) first conducted real-time size-resolved chemical composition measurements
82 during the New Year's period in Mainz, Germany using an Aerodyne Time-of-Flight
83 Aerosol Mass Spectrometer (ToF-AMS). To our knowledge, there are no such
84 real-time measurements of chemical composition of aerosol particles during fireworks
85 events in China yet, which limits our understanding on the rapid formation and
86 evolution of fireworks events, and also their impacts on particulate matter (PM)
87 pollution.

88 Secondary aerosol is of more concern compared to primary aerosol because it is
89 formed over regional scales and exerts impacts on air quality over wider areas (Matsui

90 et al., 2009;DeCarlo et al., 2010). Therefore, extensive studies have been conducted in
91 recent years to characterize the sources and formation mechanisms of secondary
92 aerosol (Yao et al., 2002;Duan et al., 2006;Sun et al., 2006;Wang et al., 2006;Guo et
93 al., 2010;Yang et al., 2011;Sun et al., 2013b;Zhang et al., 2013;Zhao et al., 2013).
94 SIA was observed to contribute a large fraction of PM_{2.5} and played an enhanced role
95 during haze episodes due to the faster heterogeneous reactions associated with higher
96 humidity (Liu et al., 2013;Sun et al., 2013a;Zhao et al., 2013;Sun et al., 2014;Wang et
97 al., 2014). While SIA was relatively well characterized, secondary organic aerosol
98 (SOA) is not well understood (Huang et al., 2014). The recent deployments of
99 Aerodyne Aerosol Mass Spectrometers (AMS) greatly improved our understanding
100 on sources and evolution processes of organic aerosol (OA) in China, and also the
101 different roles of primary organic aerosol (POA) and SOA in PM pollution (Huang et
102 al., 2010;Sun et al., 2010;He et al., 2011;Sun et al., 2012;Sun et al., 2013b;Zhang et
103 al., 2014). While SOA is more significant in summer (Huang et al., 2010;Sun et al.,
104 2010;Sun et al., 2012), POA generally plays a more important role during wintertime
105 (Sun et al., 2013b). Recently, the role of SOA in fine particle pollution during
106 wintertime – a season with frequent occurrences of pollution episodes in Beijing was
107 extensively investigated and the results highlighted the similar importance of SOA to
108 SIA (Sun et al., 2013b;Sun et al., 2014;Zhang et al., 2014). However, the role of SOA
109 in particulate pollution during periods with largely reduced anthropogenic activities is
110 not well known yet (Huang et al., 2012). This study happened to take place in a month
111 with the most important holiday in China, i.e., the Spring Festival. The source
112 emissions (e.g., traffic and cooking) have significant changes due to a large reduction
113 of population and anthropogenic activities in the city. This provides a unique
114 opportunity to investigate how source changes affect aerosol chemistry including

115 primary emissions and secondary formation in Beijing. Although Huang et al. (2012)
116 investigated such a holiday effect on aerosol composition and optical properties in
117 Shanghai, the data analyses were limited by daily average composition measurements
118 and also the significantly different meteorological conditions between holiday and
119 non-holiday periods.

120 In this study, an Aerosol Chemical Speciation Monitor (ACSM) along with
121 various collocated instruments was deployed in Beijing during February 2013. The
122 chemical composition of submicron aerosol (PM_{10}) from fireworks is quantified based
123 on the highly time – resolved measurements of non-refractory submicron aerosol
124 (NR- PM_{10}) species (organics, sulfate, nitrate, ammonium, chloride, and potassium) and
125 black carbon. The impacts of fireworks on PM pollution during Chinese Lunar New
126 Year (LNY), Lunar Fifth Day (LFD), and Lantern Festival (LF) are investigated, and
127 the roles of secondary formation in PM pollution are elucidated. Further, the effects of
128 reduced anthropogenic emissions on primary and secondary aerosols in the city are
129 illustrated, which has significant implications for making air pollution control
130 strategies in Beijing.

131 **2 Experimental**

132 **2.1 Sampling site**

133 The measurements in this study were conducted at the Institute of Atmospheric
134 Physics (IAP), Chinese Academy of Sciences ($39^{\circ}58'28''N$, $116^{\circ}22'16''E$), an urban
135 site located between the north third and fourth ring road in Beijing (Sun et al., 2012).
136 Aerosol measurements were performed from 1 February to 1 March 2013 when three
137 episodes with significant influences of fireworks, i.e., Lunar New Year (LNY), Lunar
138 Fifth Day (LFD), and Lantern Festival (LF), were observed (Fig. 1). The

139 meteorological conditions during the measurement period are reported in Fig. 1.
140 Winds at the ground surface were generally below 2 m s^{-1} and temperature averaged
141 $0.6 \text{ }^\circ\text{C}$. Relative humidity (RH) varied periodically with higher values generally
142 associated with higher PM pollution.

143 **2.2 Aerosol sampling**

144 The chemical composition of NR-PM₁ including organics, sulfate, nitrate,
145 ammonium, and chloride were measured *in situ* by the ACSM at an approximate
146 15-min time intervals (Ng et al., 2011b). The ACSM has been widely used for
147 long-term and routine aerosol particle composition measurements due to its
148 robustness (Sun et al., 2012; Budisulistiorini et al., 2014; Petit et al., 2014) despite its
149 lower sensitivity and mass resolution compared to previous versions of research-grade
150 AMS (Jayne et al., 2000; DeCarlo et al., 2006). In this study, the ambient air was
151 drawn inside the sampling room at a flow rate of 3 L min^{-1} , of which $\sim 0.1 \text{ L min}^{-1}$ was
152 sub-sampled into the ACSM and 0.85 L min^{-1} into a Cavity Attenuated Phase Shift
153 Spectrometer (CAPS) particle extinction monitor (Massoli et al., 2010). A PM_{2.5}
154 cyclone (Model: URG-2000-30ED) was supplied in front of the sampling line to
155 remove coarse particles with aerodynamic diameters larger than $2.5 \text{ }\mu\text{m}$. The aerosol
156 particles were dried by a silica gel dryer (RH < 40%) before entering the ACSM and
157 the CAPS. The ACSM was operated at a scan rate of 500 ms amu^{-1} for the mass
158 spectrometer from m/z 10 – 150. Because ACSM cannot detect refractory components,
159 e.g., BC and mineral dust, a two-wavelength Aethalometer (Model AE22, Magee
160 Scientific Corp.) was therefore used to measure refractory BC in PM_{2.5}. The light
161 extinction of dry fine particles (b_{ext} , 630 nm) was measured at 1 s time resolution with
162 a precision (3σ) of 1 M m^{-1} by the CAPS monitor. In addition, the mass concentration

163 of PM_{2.5} was determined by a heated Tapered Element Oscillating Microbalance,
164 TEOM, and the collocated gaseous species (including CO, SO₂, NO, NO_x and O₃)
165 were measured by various gas analyzers (Thermo Scientific) at 1 min time resolution.
166 A more detailed descriptions of aerosol and gas measurements were given in Sun et al.
167 (2013b).

168 **2.3 ACSM data analysis**

169 The ACSM data were analyzed for the mass concentrations and chemical
170 composition of NR-PM₁ using standard ACSM software (v 1.5.3.2) written within
171 Igor Pro (WaveMetrics, Inc., Oregon USA). A composition-dependent collection
172 efficiency (CE) recommended by Middlebrook et al.(2012), $CE = \max(0.45, 0.0833$
173 $+ 0.9167 \times \text{ANMF})$, was used to account for the incomplete detection due to the
174 particle bouncing effects (Matthew et al., 2008) and the influences caused by high
175 mass fraction of ammonium nitrate (ANMF). Because aerosol particles were overall
176 neutralized ($\text{NH}_4^+_{\text{measured}}/\text{NH}_4^+_{\text{predicted}} = 1.01$, $r^2 = 0.99$) and also dried before entering
177 the ACSM, the effects of particle acidity and RH would be minor (Matthew et al.,
178 2008;Middlebrook et al., 2012). The default relative ionization efficiencies (RIEs)
179 except ammonium (RIE = 6.5) that was determined from the IE calibration were used
180 in this study..Quantification of K^+ is challenging for ACSM because of a large
181 interference of organic C_3H_3^+ at m/z 39 and also uncertainties caused by surface
182 ionization (Slowik et al., 2010). In this work, we found that m/z 39 was tightly
183 correlated with m/z 43 that is completely organics during non-fireworks (NFW)
184 periods ($r^2 = 0.87$, slope = 0.45, Fig. S1). However, higher ratios of m/z 39/43 during
185 FW periods were observed due to the elevated K^+ signal from burning of fireworks.
186 Assuming that m/z 39 was primarily contributed by organics during NFW periods, the

187 excess m/z 39 signal, i.e., K^+ , can then be estimated as m/z 39 – m/z 43 \times 0.45. The
188 $^{41}K^+$ at m/z 41 was calculated using an isotopic ratio of 0.0722, i.e., $^{41}K^+ = 0.0722 \times$
189 K^+ . The K^+ signal was converted to mass concentration with a RIE of 2.9 that was
190 reported by Drewnick et al. (2006). It should be noted that the quantification of K^+ in
191 this study might have a large uncertainty because of the unknown RIE of K^+ (RIE_K).
192 The RIE_K can vary a lot depending on the tuning of the spectrometer and the
193 temperature of the vaporizer. For example, Slowik et al. (2010) reported a $RIE_K = 10$
194 based on the calibration of pure KNO_3 particles using a ToF-AMS, which is much
195 higher than the $RIE_K = 2.9$ obtained from the comparisons of K/S from fireworks and
196 AMS measurements (Drewnick et al., 2006). In addition, the stability of surface
197 ionization (SI) and electron impact (EI) also affects RIE_K . We then checked the
198 variations of the ratio of m/z 39/ m/z 23 (two m/z 's with similar surface ionization
199 issues). The average ratio of m/z 39/23 during LFD and LF is 8.7 and 11.1,
200 respectively, which is close to 9.0 during the NFW periods. The results suggest that
201 the SI/EI ratio was relatively stable throughout the study. Because we didn't have
202 collocated K measurement, $RIE_K = 2.9$ that was estimated from fireworks was used in
203 this study. The quantified K^+ during LFD and LF on average contributed 4.5% and
204 4.7% of PM_{10} , respectively, which is close to ~5% ($PM_{2.5}$) reported by Cheng et al.
205 (2014). Also, the large contribution of K^+ to PM_{10} (20.5%) during LNY, likely due to
206 the intensified firework emissions (mainly firecrackers), is consistent with that (17.3%)
207 observed during LNY 2014 in megacity Tianjin (Tian et al., 2014). Using $RIE_K=10$
208 will decrease the K^+ concentration by a factor of more than 3, which appears to
209 underestimate K^+ a lot. Therefore, $RIE_K = 2.9$ for the quantification of K^+ in our study
210 appear to be reasonable. The KCl^+ (m/z 74) and $^{41}KCl^+/K^{37}Cl^+$ (m/z 76) were
211 estimated by the differences between the measured and PMF modeled m/z 74 (Fig.

212 S2). Not surprisingly, the quantified KCl^+ highly correlates with K^+ ($r^2 = 0.82$, Fig.
213 S2c). The chloride concentration was also biased at m/z 35 during some periods (e.g.,
214 LNY, Fig. S3), which is likely due to the interferences of NaCl from fireworks.
215 Therefore, Cl^+ (m/z 35) was recalculated based on its correlation with m/z 36 (mainly
216 HCl^+ with negligible C_3^+ and ^{36}Ar), i.e., m/z 35 = $0.15 \times m/z$ 36. The $^{37}\text{Cl}^+$ was
217 calculated using an isotopic ratio of 0.323, i.e., $^{37}\text{Cl}^+ = 0.323 \times ^{35}\text{Cl}^+$. The comparison
218 of the reconstructed chloride from the default values is shown in Fig. S3b.

219 The positive matrix factorization (PMF) with the algorithm of PMF2.exe in robust
220 mode (Paatero and Tapper, 1994) was performed on organic aerosol (OA) mass
221 spectra (m/z 12 – 120) to resolve distinct OA components from different sources. The
222 PMF results were evaluated with an Igor Pro-based PMF Evaluation Tool (PET, v
223 2.04) (Ulbrich et al., 2009) following the procedures detailed in Zhang et al. (2011).
224 After a careful evaluation of the spectral profiles, diurnal variations and correlations
225 with external tracers, a 6-factor solution ($Q/Q_{\text{exp}} = 4.3$) was chosen, yielding a
226 hydrocarbon-like OA (HOA), a cooking OA (COA), a coal combustion OA (CCOA),
227 and three oxygenated OA (OOA) components. Because of the absence of collocated
228 measurements to validate the different OOA components, the three OOA components
229 were recombined into one OOA component. The contributions of four OA factors
230 were relatively stable across different f_{peak} values (average $\pm 1\sigma$; min – max, Fig. S4):
231 HOA ($14 \pm 1.6\%$; 12 – 16%), COA ($14 \pm 2.8\%$; 11 – 17%), CCOA ($19 \pm 2.7\%$; 15 –
232 22%), and OOA ($51 \pm 1.7\%$; 49 – 55%). However, considering the mass spectra of OA
233 factors at $f_{\text{peak}} = -1$ presented the best correlation with those identified in winter
234 2011-2012 ($r^2 = 0.86 - 0.99$, Fig. S5) (Sun et al., 2013b), the four OA factors with
235 $f_{\text{peak}} = -1$ was chosen in this study. The HOA spectrum resembles to that identified

236 by PMF analysis of high resolution OA mass spectra in Beijing in January 2013
237 (Zhang et al., 2014) which are both characterized pronounced m/z 91 and 115.
238 Although the CCOA spectrum doesn't present similar pronounced m/z 's (e.g., 77, 91,
239 105, and 115) as that resolved at a rural site in Central Eastern China (Hu et al., 2013),
240 it shows more similarity to that resolved in Beijing (Zhang et al., 2014). Also, CCOA
241 correlates better with chloride with an importance source from coal combustion
242 (Zhang et al., 2012) than HOA ($r^2 = 0.41$ vs. 0.24), and also correlates well with m/z
243 60 ($r^2 = 0.77$, Fig. S6) a tracer m/z for biomass burning (Cubison et al., 2011). Note
244 that better correlations between HOA+CCOA and BC ($r^2 = 0.88$), NO_x ($r^2 = 0.77$),
245 and CO ($r^2 = 0.63$) than HOA ($r^2 = 0.36 - 0.47$) were observed in this study, which
246 might suggest that coal combustion emissions are also important sources of CO, BC
247 and NO_x during wintertime (Tian et al., 2008; Zhi et al., 2008).. Although COA didn't
248 have external tracers to validate, it is very distinct as suggested by its unique diurnal
249 patterns (two peaks corresponding to meal time) and high m/z 55/57 ratio. Similar to
250 our previous study (Sun et al., 2013b), the OOA shows a tight correlation with NO_3
251 ($r^2 = 0.90$) and also a good correlation with $\text{SO}_4^{2-} + \text{NO}_3^-$ ($r^2 = 0.87$). The mass spectral
252 profiles and time series of four OA factors are shown in Fig. S6.

253 No biomass burning OA (BBOA) was resolved in this study. One of the reasons is
254 that BBOA was likely not an important component of OA (e.g., < 5%), which is
255 unlikely to be resolved accurately by PMF (Ulbrich et al., 2009). Indeed, we didn't
256 observe strong biomass burning influences throughout the study by checking the
257 scatter plot of f_{60} vs. f_{44} (Fig. S7). We found that f_{60} vs. f_{44} in Fig. S7 is outside of the
258 typical biomass burning region (Cubison et al., 2011) for most of the time during this
259 study. Although the average f_{60} (0.42%) is slightly higher than the typical value of f_{60}

260 (~0.3%) in the absence of biomass burning impact (DeCarlo et al., 2008;Ulbrich et al.,
261 2009), it is also likely due to the short range of m/z (12 – 120) used for the calculation
262 of f_{60} . A summary of other key diagnostic plots of the PMF solution are given in Fig.
263 S8 and Fig. S9.

264 **3 Results and discussion**

265 **3.1 Identification and quantification of fireworks events**

266 Burning of fireworks has been found to emit a large amount of K^+ , which can be
267 used to identify the FW events (Drewnick et al., 2006;Wang et al., 2007). As shown
268 in Fig. 1 and Fig. 2, three FW events with significantly elevated K^+ were observed on
269 the days of Lunar New Year (LNY, 9-10 February), Lunar Fifth Day (LFD, 14
270 February), and Lantern Festival (LF, 24 February), respectively. All three FW events
271 started approximately at 18:00 and ended at midnight except LNY with a continuous
272 FW impact until 4:00 on the second day (Fig. 2). Fig. 1 shows that the relative
273 humidity was generally below 30% during LNY and LFD. While the wind speed at
274 the ground surface remained consistently below 2 m s^{-1} , it was increased to $\sim 4 \text{ m s}^{-1}$
275 at the height of 100 m. Also note that there was a wind direction change in the middle
276 of the two events. The meteorological conditions during LF were stagnant with wind
277 speed generally below 2 m s^{-1} across different heights. The relative humidity was $\sim 50\%$
278 and the temperature averaged 3.5°C .

279 To estimate the contributions of fireworks, we first assume that the background
280 concentration of each species has a linear variation during FW period. A linear fit was
281 then performed on the 6 h data before and after FW events. The difference between
282 the measured and the fitted value is assumed as the contribution from FW. The typical
283 examples for estimating FW contributions are shown in Fig. S10. It should be noted

284 that this approach might significantly overestimate the FW contributions of primary
285 species (e.g., HOA, COA, CCOA, and BC) that were largely enhanced during the
286 typical FW periods (18:00 – 24:00) due to the increased local emissions (see Fig. S11
287 for diurnal variations of aerosol species). However, it should have a minor impact on
288 secondary species (e.g., SO₄, NO₃, and OOA) because of their relatively stable
289 variations between 18:00-24:00. As shown in Fig.2, all aerosol species showed
290 substantial increases from 15:00 to 21:00 on the day of LNY which coincidentally
291 corresponded to a gradual change of wind direction. Therefore, regional transport
292 might have played dominant roles for the evolution of chemical species during this
293 period. For these reasons, only the FW contributions between 23:30, 9 February and
294 3:30, 10 February when the meteorological conditions were stable were estimated.
295 The FW contributions during LFD might also be overestimated due to the influences
296 of regional transport as suggested by the wind direction change in the middle.

297 ~~Considering above, the contributions estimated in this work would represent the upper~~
298 ~~limits of FW.~~

299 **3.2 Mass concentration and chemical composition of FW aerosols**

300 Figure 1 shows the time series of mass concentrations of PM₁, PM_{2.5}, and
301 submicron aerosol species from 1 February to 1 March 2013. Because ACSM cannot
302 measure the metals (e.g., Sr, Ba, Mg, etc.) that were significantly enhanced during
303 FW periods (Wang et al., 2007; Vecchi et al., 2008), the PM₁ in this study refer to
304 NR-PM₁ (= Org + SO₄ + NO₃ + NH₄ + Chl + K + KCl) + BC. The PM_{2.5} showed three
305 prominent FW peaks with the maximum concentration occurring at ~00:30 during
306 LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM_{2.5}
307 during LNY (775 μg m⁻³) is more than 10 times higher than the China National

308 Ambient Air Quality Standard ($75 \mu\text{g m}^{-3}$, 24 h average). The average FW- $\text{PM}_{2.5}$ mass
309 concentrations during three FW events all exceeded $100 \mu\text{g m}^{-3}$. These results suggest
310 that fireworks have large impacts on fine particle pollution, yet generally less than
311 half day (approximately 10 h for LNY, and 6 h for LFD and LF). The PM_1 also
312 showed increases during the FW periods, yet not as significant as $\text{PM}_{2.5}$. In fact the
313 correlation of PM_1 versus $\text{PM}_{2.5}$ shows much lower $\text{PM}_1/\text{PM}_{2.5}$ (0.08 – 0.19) ratios
314 during three FW events than that observed during NFW periods (0.90) (Fig. 3). One
315 of the reasons is likely due to the mineral dust component and metals from fireworks
316 that ACSM did not measure. However, the ACSM un-detected metals (e.g., Mg, Sr,
317 and Ba) that are largely enhanced during FW periods generally contribute a small
318 fraction of PM (<2%) (Wang et al., 2007; Vecchi et al., 2008; Kong et al., 2014).
319 Therefore, our results might suggest that a large fraction of aerosol particles from the
320 burning of fireworks was emitted in the size range of 1 – 2.5 μm . Consistently,
321 Vecchi et al. (2008) found the best correlation between the fireworks tracer, Sr, and
322 the particles between 700-800 nm (mobility diameter, D_m) which is approximately
323 equivalent to 1.9 – 2.2 μm in D_{va} (vacuum aerodynamic diameter, D_{va}) with a density
324 of 2.7 g cm^{-3} (Zhang et al., 2010).

325 Figure 4 shows the average chemical composition of PM_1 and OA from
326 fireworks and also the background composition during LNY, LFD and LF. The
327 background PM_1 during LNY and LFD showed typical characteristics of clean
328 periods with high fraction of organics (> ~50%) (Sun et al., 2012; Sun et al., 2013b),
329 whereas that during LF was dominated by SIA (52%). As a comparison, organics
330 constituted the major fraction of FW- PM_1 , contributing 44 – 55% on average. During
331 LNY, FW exerted large impacts on potassium and chloride whose contributions were

332 elevated to 21% and 15% of PM₁, respectively, from less than 7% (Chl) in the
333 background aerosols. The large increases of potassium and chloride were also
334 observed during LFD and LF, and previous studies in Beijing (Wang et al.,
335 2007; Cheng et al., 2014). As shown in Fig. 4, FW also emitted a considerable amount
336 of sulfate, accounting for 7% - 14% of PM₁. Sulfate correlated strongly with SO₂
337 during all three FW events ($r^2 = 0.49 - 0.92$). Given that the relative humidity was low,
338 < 30% during LNY and LFD, and ~ 50% during LF, aqueous-phase oxidation of SO₂
339 depending on liquid water content could not play significant roles for the sulfate
340 formation (Sun et al., 2013a). Therefore, sulfate in FW-PM₁ was mainly from the
341 direction emissions of FW. Compared to sulfate, FW appeared to show minor impacts
342 on nitrate, for example, 4% and 2% during LNY and LF, respectively. Although
343 nitrate contributed 12% of FW-PM₁ during LFD, most of it was likely from regional
344 transport as supported by synchronous increases of all aerosol species associated with
345 a wind direction change in the middle (Fig. 2).

346 The OOA contributed dominantly to OA during LNY, which is 79% on average
347 (Fig. 4a). As shown in Fig. 5, the mass spectrum of FW-organics is highly similar to
348 that of low-volatility OOA (LV-OOA, $r^2 = 0.94$; $r^2 = 0.89$ by excluding m/z 18 and
349 m/z 44) (Ng et al., 2011a) indicating that the FW-organics is likely emitted in
350 secondary. Consistently, Drewnick et al. (2006) also found large enhancements of the
351 OOA-related m/z 's (e.g., m/z 44) during New Year's fireworks, but the HOA-related
352 m/z 's (e.g., m/z 57) are not significant contributors to FW organics. OOA accounted
353 for a much smaller fraction of OA during LF (28%) due to the large contributions of
354 POA components (72%). Although the OOA contributions varied during three FW
355 events, their absolute concentrations were relatively close ranging from 5.8 to 7.9 μg

356 m^{-3} . It should be noted that our approach might overestimate the POA components in
357 FW-OA because of the influences of NFW sources, in particular during the FW
358 period of LF when the local HOA, COA, and CCOA happened to have large increases.
359 By excluding the POA components in FW-OA, FW on average contributed 15 – 19
360 $\mu\text{g m}^{-3}$ PM_1 during three FW events.

361 **3.3 Secondary aerosol and PM pollution**

362 The PM_1 (NR- PM_1 + BC) varied largely across the entire study with daily average
363 mass concentration ranging from 9.1 to 169 $\mu\text{g m}^{-3}$. The average PM_1 mass
364 concentration was 80 (± 68) $\mu\text{g m}^{-3}$, which is approximately 20% higher than that
365 observed during winter 2011-2012 (Sun et al., 2013b). Organics composed the major
366 fraction of PM_1 accounting for 43%, followed by nitrate (22%), sulfate (14%),
367 ammonium (13%), BC (5%) and chloride (3%). The OA composition was dominated
368 by OOA (53%) with the rest being POA. Compared to winter 2011-2012 (Sun et al.,
369 2013b), this study showed significantly enhanced OOA (53% vs. 31%) and secondary
370 nitrate (22% vs. 16%), indicating that secondary formation have played important
371 roles in the formation of pollution episodes.

372 Figure 1d shows that submicron aerosol species alternated routinely between
373 pollution events (PEs) and clean periods (CPs) throughout the entire study. The PEs
374 generally lasted ~1 – 2 days except the one on 23 – 28 February that lasted more than
375 5 days, whereas the CPs were shorter, generally less than 1 day. In total, 9 PEs and 9
376 CPs were identified in this study (Fig. 1). A statistics of the mass concentrations and
377 mass fractions of aerosol species during 9 PEs is presented in Fig. 6. The average PM_1
378 mass concentration ranged 68 – 179 $\mu\text{g m}^{-3}$ during PEs with the total secondary
379 particulate matter ($\text{SPM} = \text{OOA} + \text{SO}_4 + \text{NO}_3 + \text{NH}_4$) accounting for 63 – 82%. The

380 average mass concentration of SPM for the 9 PEs was $86 (\pm 32) \mu\text{g m}^{-3}$, which is
381 nearly 3 times that of primary PM (PPM = HOA + COA + CCOA + BC + Chl) (30
382 $\pm 9.5 \mu\text{g m}^{-3}$). SPM consistently dominated PM_{10} across different PM levels (69 – 75%),
383 but generally with higher contributions (up to 81%) during daytime (Fig. 7b). The
384 diurnal cycle of SPM presented a gradual increase from 50 to $70 \mu\text{g m}^{-3}$ between
385 10:00 – 20:00, indicating evident photochemical production of secondary species
386 during daytime. It should be also noted that all secondary species showed
387 ubiquitously higher mass concentrations than those of primary species (Fig. 6a).

388 The SOA generally contributed more than 50% to OA with an average of 55%
389 during PEs except the episode on 3 February (35%). It's interesting to note that the
390 contribution of POA increased as a function of organic loadings which varied from
391 $\sim 35\%$ to 63% when organics ~~was above~~reached $80 \mu\text{g m}^{-3}$ (Fig. 7c). Such behavior is
392 mainly caused by the enhanced CCOA at high organic mass loadings, which was also
393 observed during winter 2011 – 2012 (Sun et al., 2013b). These results suggest that
394 POA played more important roles than SOA in PM pollution during periods with high
395 organic mass loadings (e.g., $> 60 \mu\text{g m}^{-3}$). In fact, POA showed even higher mass
396 concentration than OOA at nighttime (0:00 – 8:00) due to the intensified local
397 emissions, e.g., coal combustion for heating. Despite this, the role of POA in PM
398 pollution was compensated by the elevated secondary inorganic species as a function
399 of PM loadings (Fig. 7a) leading to the consistently dominant SPM across different
400 pollution levels. Figure 8a shows an evidently lower contribution of organics to PM_{10}
401 during PEs than CPs. The elevated secondary inorganic species during PEs were
402 closely related to the increase of RH (Fig. 1). For example, during the pollution
403 episode on 3 February, the sulfate concentration increased rapidly and became the

404 major inorganic species when RH was increased from ~60% to > 90%. The gaseous
405 SO₂ showed a corresponding decrease indicating aqueous-phase processing of SO₂ to
406 form sulfate, consistent with our previous conclusion that aqueous-phase processing
407 could contribute more than 50% of sulfate production during winter 2011-2012 (Sun
408 et al., 2013a).

409 The compositional differences between PEs and CPs also led to different mass
410 extinction efficiency (MEE, 630 nm) of PM₁ (Fig. 8b). The higher MEE (6.4 m² g⁻¹)
411 during PEs than CPs (4.4 m² g⁻¹) is primarily due to the enhanced secondary species,
412 and also likely the increases of aerosol particle sizes although we don't have size data
413 to support it. Similar increases of mass scattering efficiency from clean periods to
414 relatively polluted conditions were also observed previously in Beijing and Shanghai
415 (Jung et al., 2009;Huang et al., 2013). It should be noted that the MEE of PM₁ in this
416 study refers to PM_{2.5} *b*_{ext}/PM₁. Considering that PM₁ on average contributes ~60-70%
417 of PM_{2.5} in Beijing (Sun et al., 2012;Sun et al., 2013b), the real MEE of PM₁ during
418 PEs and CPs would be ~3.8 - 4.5 and ~2.6 - 3.1 m² g⁻¹, respectively.

419 **3.4 Holiday Effects on PM Pollution**

420 Figure 9 shows a comparison of aerosol species, gaseous species, and
421 meteorological parameters between holiday (HD) and non-holiday (NHD) periods.
422 The official holiday for the Spring Festival was 9 – 15 February. However, we noted a
423 large decrease of cooking aerosols from 7 February until 19 February (Fig. S6b),
424 whose emissions were expected to be stable under similar meteorological conditions.
425 The decrease of COA was likely due to the reduction of the number of population in
426 Beijing, which agreed with the fact that most migrants from outside Beijing were
427 leaving for hometown before the official holiday. Therefore, 7 – 19 February was

428 used as a longer holiday for a comparison. It was estimated that approximately half of
429 population (9 million) left Beijing before the Spring Festival
430 (http://news.xinhuanet.com/yzyd/local/20130208/c_114658765.htm). Such a great
431 reduction in human activities would exert a large impact on aerosol composition and
432 sources in the city during holidays. To better investigate the HD effects on PM
433 pollution, the data shown in Fig. 9 excluded the CPs marked in Fig. 1. The data with
434 the CPs included are presented in Fig. S12.

435 The differences between HD and NHD for primary species varied largely among
436 different species. COA showed the largest reduction (69%) among aerosol species
437 with the average concentration decreasing from $5.8 \mu\text{g m}^{-3}$ during NHD to $1.8 \mu\text{g m}^{-3}$
438 during HD. The contribution of COA to OA showed a corresponding decrease from
439 12% to 4%. Given the similar meteorological conditions between HD and NHD, e.g.,
440 RH (46% vs. 52%) and wind speed (1.3 m s^{-1} vs. 1.2 m s^{-1}), the reduction of COA
441 clearly indicated a large decrease of population and the number of restaurants opened
442 during HD. The CCOA showed approximately 30% reduction during HD, and its
443 contribution to OA decreased from 23% to 18%. Not surprisingly, chloride showed a
444 similar reduction as CCOA because it was primarily from coal combustion emissions
445 during wintertime (Sun et al., 2013b). Figure 9 also shows a significant reduction
446 (54%) for NO indicating much less traffic emissions in the city during HD. The HOA,
447 however, even showed a slight increase during HD, which appeared to contradict with
448 the reduction of two combustion-related tracers, BC and CO (~20%). This can be
449 explained by the fact that coal combustion is a large source of BC and CO during
450 heating season (Tian et al., 2008; Zhi et al., 2008). Consistently, BC and CO showed
451 relatively similar reductions to CCOA. Therefore, the minor variation of HOA might

452 indicate that the number of heavy-duty vehicles and diesel trucks that dominated
453 HOA emissions (Massoli et al., 2012; Hayes et al., 2013) remained little change during
454 HD period although that of gasoline vehicles was largely decreased. It should be
455 noted that HOA showed a large peak on 9 February – the first day of the official
456 holiday (Fig. S6b) when more traffic emissions were expected due to many people
457 leaving for hometown. After that, HOA showed slightly lower concentration during
458 11 – 17 February than other periods. In fact, the average HOA showed a slight
459 reduction (~5%) during the long holiday period (7 – 19 February) suggesting a small
460 holiday effect on HOA reduction. Together, the total primary aerosol species (PPM)
461 showed an average reduction of 22% because of holiday effects.

462 Nitrate showed the largest reduction among secondary species by 22% during HD,
463 primarily due to a reduction of its precursors NO and NO₂. The results suggest that
464 reducing traffic emissions would help mitigate the nitrate pollution in the city.
465 Compared to nitrate, sulfate showed minor changes (2%) between HD and NHD, and
466 OOA even showed a slight increase (6%) during HD. One of the reasons is that
467 secondary sulfate and OOA were mainly formed over regional scale and less affected
468 by local production, consistent with their relatively flat diurnal cycles (Fig. S11).
469 Ammonium showed a reduction between nitrate and sulfate because ammonium
470 mainly existed in the form of (NH₄)₂SO₄ and NH₄NO₃. Overall, secondary species
471 showed generally lower reductions than primary species with the total secondary
472 species (SPM) showing an average reduction of 9% during HD. The joint reductions
473 of PPM and SPM led to an average reduction of 13% for PM₁ during HD. However,
474 these reductions did not help alleviate the fine particle pollution during HD. The
475 PM_{2.5} excluding FW impacts even showed 27% increase from 96 μg m⁻³ during NHD

476 to $122 \mu\text{g m}^{-3}$ during HD. One possible reason is likely due to the increases of aerosol
477 species in the size range of $1 - 2.5 \mu\text{m}$ during HD period. The longer holiday (LHD, 7
478 – 19 February) showed similar influences on both primary and secondary species as
479 the official holiday (9 – 15 February). COA, CCOA, and NO are the three species
480 with the largest reductions during LHD ($> 50\%$). However, HOA, SO_4 , OOA, and
481 $\text{PM}_{2.5}$ showed rather small changes ($< \pm 7\%$). Therefore, results in this study suggest
482 that controlling the primary source emissions, e.g., cooking and traffic emissions in
483 the city can reduce the primary particles largely, yet has limited effects on secondary
484 species and the total fine particle mass. One of the reasons is because the severe PM
485 pollution in Beijing is predominantly contributed by secondary species (see
486 discussions in section 3.3) that are formed over regional scales. Reducing the primary
487 source emissions in local areas would have limited impacts on mitigation of air
488 pollution in the city. Similarly, Guo et al. (2013) reported a large reduction of primary
489 organic carbon (OC) from traffic emissions and coal combustion during the 2008
490 Olympic Summer Games when traffic restrictions and temporary closure of factories
491 were implemented. However, secondary OC was not statistically different between
492 controlled and non-controlled periods. Our results highlight the importance of
493 implementing joint efforts over regional scales for air pollution control in north
494 China.

495 **4 Conclusions**

496 We have characterized the aerosol particle composition and sources during the
497 Chinese Spring Festival in 2013. The average PM_1 mass concentration was $80 (\pm 68)$
498 $\mu\text{g m}^{-3}$ for the entire study with organics being the major fraction (43%). Nine
499 pollution events and nine clean periods with substantial compositional differences

500 were observed. The secondary particulate matter (= SOA+ sulfate + nitrate +
501 ammonium) played a dominant role for the PM pollution during nine PEs. The
502 contributions of SPM to PM₁ varied from 63% to 82% with SOA on average
503 accounting for ~55% of OA. As a result, the average mass extinction efficiency of
504 PM₁ during PEs (6.4 m² g⁻¹) was higher than that during CPs (4.4 m² g⁻¹). Three FW
505 events, i.e., LNY, LFD, and LF, were identified, which showed significant and
506 short-term impacts on fine particles, and non-refractory potassium, chloride, and
507 sulfate in PM₁. The FW also exerted a large impact on organics that presented mainly
508 in secondary as indicated by its similar mass spectrum to that of oxygenated OA. The
509 holiday effects on aerosol composition and sources were also investigated by
510 comparing the differences between holiday and non-holiday periods. The changes of
511 anthropogenic source emissions during the holiday showed large impacts on reduction
512 of cooking OA (69%), nitrogen monoxide (54%), and coal combustion OA (28%) in
513 the city, yet presented much smaller influences on secondary species. The average
514 SOA and the total PM_{2.5} even increased slightly during the holiday period. Results
515 here have significant implications that controlling the local primary source emissions,
516 e.g., cooking and traffic activities, might have limited effects on improving air quality
517 during polluted days when SPM from regional transport dominated aerosol
518 composition for most of time. Our results also highlight the importance of
519 implementing joint measures over regional scales for mitigation of air pollution in
520 megacity Beijing.

521

522 **Acknowledgements**

523 This work was supported by the National Key Project of Basic Research
524 (2014CB447900), the Strategic Priority Research Program (B) of the Chinese
525 Academy of Sciences (Grant No. XDB05020501), and the National Natural Science
526 Foundation of China (41175108). We thank Huabin Dong, Hongyan Chen, and Zhe
527 Wang's help in data collection, and also the Technical and Service Center, Institute of
528 Atmospheric Physics, Chinese Academy of Sciences for providing meteorological
529 data.

530

531 **References**

- 532 Budisulistiorini, S. H., Canagaratna, M. R., Croteau, P. L., Baumann, K., Edgerton, E. S.,
533 Kollman, M. S., Ng, N. L., Verma, V., Shaw, S. L., Knipping, E. M., Worsnop, D. R.,
534 Jayne, J. T., Weber, R. J., and Surratt, J. D.: Intercomparison of an Aerosol Chemical
535 Speciation Monitor (ACSM) with ambient fine aerosol measurements in downtown
536 Atlanta, Georgia, *Atmos. Meas. Tech.*, 7, 1929-1941, 10.5194/amt-7-1929-2014, 2014.
- 537 Chan, C. K., and Yao, X.: Air pollution in mega cities in China, *Atmos. Environ.*, 42, 1-42,
538 DOI: 10.1016/j.atmosenv.2007.09.003, 2008.
- 539 Cheng, Y., Engling, G., He, K.-b., Duan, F.-k., Du, Z.-y., Ma, Y.-l., Liang, L.-l., Lu, Z.-f., Liu,
540 J.-m., Zheng, M., and Weber, R. J.: The characteristics of Beijing aerosol during two
541 distinct episodes: Impacts of biomass burning and fireworks, *Environ. Pollut.*, 185,
542 149-157, <http://dx.doi.org/10.1016/j.envpol.2013.10.037>, 2014.
- 543 Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., Brune,
544 W. H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J.,
545 Mikoviny, T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J.,
546 Wisthaler, A., and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass
547 burning smoke in aircraft and laboratory studies, *Atmos. Chem. Phys.*, 11, 12049-12064,
548 10.5194/acp-11-12049-2011, 2011.
- 549 DeCarlo, P. F., Kimmel, J. R., Trimborn, A., Northway, M. J., Jayne, J. T., Aiken, A. C.,
550 Gonin, M., Fuhrer, K., Horvath, T., Docherty, K. S., Worsnop, D. R., and Jimenez, J. L.:
551 Field-Deployable, High-Resolution, Time-of-Flight Aerosol Mass Spectrometer, *Anal.*
552 *Chem.*, 78, 8281-8289, 2006.
- 553 DeCarlo, P. F., Dunlea, E. J., Kimmel, J. R., Aiken, A. C., Sueper, D., Crounse, J., Wennberg,
554 P. O., Emmons, L., Shinozuka, Y., Clarke, A., Zhou, J., Tomlinson, J., Collins, D. R.,
555 Knapp, D., Weinheimer, A. J., Montzka, D. D., Campos, T., and Jimenez, J. L.: Fast
556 airborne aerosol size and chemistry measurements above Mexico City and Central
557 Mexico during the MILAGRO campaign, *Atmos. Chem. Phys.*, 8, 4027-4048, 2008.

558 DeCarlo, P. F., Ulbrich, I. M., Crounse, J., de Foy, B., Dunlea, E. J., Aiken, A. C., Knapp, D.,
559 Weinheimer, A. J., Campos, T., Wennberg, P. O., and Jimenez, J. L.: Investigation of the
560 sources and processing of organic aerosol over the Central Mexican Plateau from aircraft
561 measurements during MILAGRO, *Atmos. Chem. Phys.*, 10, 5257-5280,
562 10.5194/acp-10-5257-2010, 2010.

563 Drewnick, F., Hings, S. S., Curtius, J., Eerdekens, G., and Williams, J.: Measurement of fine
564 particulate and gas-phase species during the New Year's fireworks 2005 in Mainz,
565 Germany, *Atmos. Environ.*, 40, 4316-4327, 10.1016/j.atmosenv.2006.03.040, 2006.

566 Duan, F. K., He, K. B., Ma, Y. L., Yang, F. M., Yu, X. C., Cadle, S. H., Chan, T., and
567 Mulawa, P. A.: Concentration and chemical characteristics of PM_{2.5} in Beijing, China:
568 2001–2002, *Sci. Total Environ.*, 355, 264-275, 10.1016/j.scitotenv.2005.03.001, 2006.

569 Feng, J., Sun, P., Hu, X., Zhao, W., Wu, M., and Fu, J.: The chemical composition and
570 sources of PM_{2.5} during the 2009 Chinese New Year's holiday in Shanghai, *Atmospheric*
571 *Research*, 118, 435-444, <http://dx.doi.org/10.1016/j.atmosres.2012.08.012>, 2012.

572 Godri, K. J., Green, D. C., Fuller, G. W., Dall'Osto, M., Beddows, D. C., Kelly, F. J.,
573 Harrison, R. M., and Mudway, I. S.: Particulate oxidative burden associated with firework
574 activity, 21, 8295-8301 pp., 2010.

575 Guo, S., Hu, M., Wang, Z. B., Slanina, J., and Zhao, Y. L.: Size-resolved aerosol
576 water-soluble ionic compositions in the summer of Beijing: implication of regional
577 secondary formation, *Atmos. Chem. Phys.*, 10, 947-959, 10.5194/acp-10-947-2010, 2010.

578 Guo, S., Hu, M., Guo, Q., Zhang, X., Schauer, J. J., and Zhang, R.: Quantitative evaluation of
579 emission controls on primary and secondary organic aerosol sources during Beijing 2008
580 Olympics, *Atmos. Chem. Phys.*, 13, 8303-8314, 10.5194/acp-13-8303-2013, 2013.

581 Hayes, P. L., Ortega, A. M., Cubison, M. J., Froyd, K. D., Zhao, Y., Cliff, S. S., Hu, W. W.,
582 Toohey, D. W., Flynn, J. H., Lefer, B. L., Grossberg, N., Alvarez, S., Rappenglück, B.,
583 Taylor, J. W., Allan, J. D., Holloway, J. S., Gilman, J. B., Kuster, W. C., de Gouw, J. A.,
584 Massoli, P., Zhang, X., Liu, J., Weber, R. J., Corrigan, A. L., Russell, L. M., Isaacman, G.,
585 Worton, D. R., Kreisberg, N. M., Goldstein, A. H., Thalman, R., Waxman, E. M.,
586 Volkamer, R., Lin, Y. H., Surratt, J. D., Kleindienst, T. E., Offenberg, J. H., Dusanter, S.,
587 Griffith, S., Stevens, P. S., Brioude, J., Angevine, W. M., and Jimenez, J. L.: Organic
588 aerosol composition and sources in Pasadena, California during the 2010 CalNex
589 campaign, *Journal of Geophysical Research: Atmospheres*, 118, 9233–9257,
590 10.1002/jgrd.50530, 2013.

591 He, L.-Y., Huang, X.-F., Xue, L., Hu, M., Lin, Y., Zheng, J., Zhang, R., and Zhang, Y.-H.:
592 Submicron aerosol analysis and organic source apportionment in an urban atmosphere in
593 Pearl River Delta of China using high-resolution aerosol mass spectrometry, *J. Geophys.*
594 *Res.*, 116, D12304, 10.1029/2010jd014566, 2011.

595 Hu, W. W., Hu, M., Yuan, B., Jimenez, J. L., Tang, Q., Peng, J. F., Hu, W., Shao, M., Wang,
596 M., Zeng, L. M., Wu, Y. S., Gong, Z. H., Huang, X. F., and He, L. Y.: Insights on organic
597 aerosol aging and the influence of coal combustion at a regional receptor site of central
598 eastern China, *Atmos. Chem. Phys.*, 13, 10095-10112, 10.5194/acp-13-10095-2013,
599 2013.

600 Huang, K., Zhuang, G., Lin, Y., Wang, Q., Fu, J. S., Zhang, R., Li, J., Deng, C., and Fu, Q.:
601 Impact of anthropogenic emission on air quality over a megacity – revealed from an
602 intensive atmospheric campaign during the Chinese Spring Festival, *Atmos. Chem. Phys.*,
603 12, 11631-11645, 10.5194/acp-12-11631-2012, 2012.

604 Huang, R.-J., Zhang, Y., Bozzetti, C., Ho, K.-F., Cao, J.-J., Han, Y., Daellenbach, K. R.,
605 Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A.,
606 Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbazade, G.,
607 Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., Haddad, I. E.,
608 and Prevot, A. S. H.: High secondary aerosol contribution to particulate pollution during
609 haze events in China, *Nature*, advance online publication, 10.1038/nature13774, 2014.

610 Huang, X. F., He, L. Y., Hu, M., Canagaratna, M. R., Sun, Y., Zhang, Q., Zhu, T., Xue, L.,
611 Zeng, L. W., Liu, X. G., Zhang, Y. H., Jayne, J. T., Ng, N. L., and Worsnop, D. R.:
612 Highly time-resolved chemical characterization of atmospheric submicron particles
613 during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass
614 Spectrometer, *Atmos. Chem. Phys.*, 10, 8933-8945, 10.5194/acp-10-8933-2010, 2010.

615 Huang, Y., Li, L., Li, J., Wang, X., Chen, H., Chen, J., Yang, X., Gross, D. S., Wang, H.,
616 Qiao, L., and Chen, C.: A case study of the highly time-resolved evolution of aerosol
617 chemical and optical properties in urban Shanghai, China, *Atmos. Chem. Phys.*, 13,
618 3931-3944, 10.5194/acp-13-3931-2013, 2013.

619 Jayne, J. T., Leard, D. C., Zhang, X., Davidovits, P., Smith, K. A., Kolb, C. E., and Worsnop,
620 D. R.: Development of an aerosol mass spectrometer for size and composition analysis of
621 submicron particles, *Aerosol Sci. Tech.*, 33, 49-70, 2000.

622 Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Hu, M., and Sugimoto, N.: Optical properties
623 of atmospheric aerosols obtained by in situ and remote measurements during 2006
624 Campaign of Air Quality Research in Beijing (CAREBeijing-2006), *J. Geophys. Res.*,
625 114, D00G02, 10.1029/2008jd010337, 2009.

626 Kong, S., Li, L., Li, X., Yin, Y., Chen, K., Liu, D., Yuan, L., Zhang, Y., Shan, Y., and Ji, Y.:
627 The impacts of fireworks burning at Chinese Spring Festival on air quality and human
628 health: insights of tracers, source evolution and aging processes, *Atmos. Chem. Phys.*
629 *Discuss.*, 14, 28609-28655, 10.5194/acpd-14-28609-2014, 2014.

630 Li, W., Shi, Z., Yan, C., Yang, L., Dong, C., and Wang, W.: Individual metal-bearing
631 particles in a regional haze caused by firecracker and firework emissions, *Sci. Total*
632 *Environ.*, 443, 464-469, <http://dx.doi.org/10.1016/j.scitotenv.2012.10.109>, 2013.

633 Liu, X. G., Li, J., Qu, Y., Han, T., Hou, L., Gu, J., Chen, C., Yang, Y., Liu, X., Yang, T.,
634 Zhang, Y., Tian, H., and Hu, M.: Formation and evolution mechanism of regional haze: a
635 case study in the megacity Beijing, China, *Atmos. Chem. Phys.*, 13, 4501-4514,
636 10.5194/acp-13-4501-2013, 2013.

637 Massoli, P., Keabian, P. L., Onasch, T. B., Hills, F. B., and Freedman, A.: Aerosol Light
638 Extinction Measurements by Cavity Attenuated Phase Shift (CAPS) Spectroscopy:
639 Laboratory Validation and Field Deployment of a Compact Aerosol Particle Extinction
640 Monitor, *Aerosol Sci. Tech.*, 44, 428-435, 10.1080/02786821003716599, 2010.

641 Massoli, P., Fortner, E. C., Canagaratna, M. R., Williams, L. R., Zhang, Q., Sun, Y., Schwab,
642 J. J., Trimborn, A., Onasch, T. B., Demerjian, K. L., Kolb, C. E., Worsnop, D. R., and

643 Jayne, J. T.: Pollution Gradients and Chemical Characterization of Particulate Matter
644 from Vehicular Traffic Near Major Roadways: Results from the 2009 Queens College Air
645 Quality Study in NYC, *Aerosol Sci. Tech.*, 46, 1201-1218,
646 10.1080/02786826.2012.701784, 2012.

647 Matsui, H., Koike, M., Kondo, Y., Takegawa, N., Kita, K., Miyazaki, Y., Hu, M., Chang, S.
648 Y., Blake, D. R., Fast, J. D., Zaveri, R. A., Streets, D. G., Zhang, Q., and Zhu, T.: Spatial
649 and temporal variations of aerosols around Beijing in summer 2006: Model evaluation
650 and source apportionment, *J. Geophys. Res.*, 114, D00G13, 10.1029/2008jd010906, 2009.

651 Matthew, B. M., Middlebrook, A. M., and Onasch, T. B.: Collection Efficiencies in an
652 Aerodyne Aerosol Mass Spectrometer as a Function of Particle Phase for Laboratory
653 Generated Aerosols, *Aerosol Sci. Tech.*, 42, 884 - 898, 2008.

654 Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of
655 Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass
656 Spectrometer using Field Data, *Aerosol Sci. Tech.*, 46, 258-271, 2012.

657 Molina, M. J., and Molina, L. T.: Megacities and atmospheric pollution, *J. Air Waste Manage.*
658 *Assoc.*, 54, 644-680, 2004.

659 Moreno, T., Querol, X., Alastuey, A., Cruz Minguillón, M., Pey, J., Rodriguez, S., Vicente
660 Miró, J., Felis, C., and Gibbons, W.: Recreational atmospheric pollution episodes:
661 Inhalable metalliferous particles from firework displays, *Atmos. Environ.*, 41, 913-922,
662 <http://dx.doi.org/10.1016/j.atmosenv.2006.09.019>, 2007.

663 Ng, N. L., Canagaratna, M. R., Jimenez, J. L., Zhang, Q., Ulbrich, I. M., and Worsnop, D. R.:
664 Real-Time Methods for Estimating Organic Component Mass Concentrations from
665 Aerosol Mass Spectrometer Data, *Environ. Sci. Technol.*, 45, 910-916,
666 10.1021/es102951k, 2011a.

667 Ng, N. L., Herndon, S. C., Trimborn, A., Canagaratna, M. R., Croteau, P. L., Onasch, T. B.,
668 Sueper, D., Worsnop, D. R., Zhang, Q., Sun, Y. L., and Jayne, J. T.: An Aerosol
669 Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and
670 Mass Concentrations of Ambient Aerosol, *Aerosol Sci. Tech.*, 45, 770 - 784, 2011b.

671 Paatero, P., and Tapper, U.: Positive matrix factorization: A non-negative factor model with
672 optimal utilization of error estimates of data values, *Environmetrics*, 5, 111-126, 1994.

673 Petit, J. E., Favez, O., Sciare, J., Canonaco, F., Croteau, P., Močnik, G., Jayne, J., Worsnop,
674 D., and Leoz-Garziandia, E.: Submicron aerosol source apportionment of wintertime
675 pollution in Paris, France by double positive matrix factorization (PMF2) using an aerosol
676 chemical speciation monitor (ACSM) and a multi-wavelength Aethalometer, *Atmos.*
677 *Chem. Phys.*, 14, 13773-13787, 10.5194/acp-14-13773-2014, 2014.

678 Slowik, J. G., Stroud, C., Bottenheim, J. W., Brickell, P. C., Chang, R. Y. W., Liggio, J.,
679 Makar, P. A., Martin, R. V., Moran, M. D., Shantz, N. C., Sjostedt, S. J., van Donkelaar,
680 A., Vlasenko, A., Wiebe, H. A., Xia, A. G., Zhang, J., Leaitch, W. R., and Abbatt, J. P. D.:
681 Characterization of a large biogenic secondary organic aerosol event from eastern
682 Canadian forests, *Atmos. Chem. Phys.*, 10, 2825-2845, 2010.

683 Song, Y., Zhang, Y., Xie, S., Zeng, L., Zheng, M., Salmon, L. G., Shao, M., and Slanina, S.:
684 Source apportionment of PM_{2.5} in Beijing by positive matrix factorization, *Atmos.*
685 *Environ.*, 40, 1526-1537, DOI: 10.1016/j.atmosenv.2005.10.039, 2006.

686 Sun, J., Zhang, Q., Canagaratna, M. R., Zhang, Y., Ng, N. L., Sun, Y., Jayne, J. T., Zhang, X.,
687 Zhang, X., and Worsnop, D. R.: Highly time- and size-resolved characterization of
688 submicron aerosol particles in Beijing using an Aerodyne Aerosol Mass Spectrometer,
689 *Atmos. Environ.*, *44*, 131-140, 2010.

690 Sun, Y., Zhuang, G., Tang, A., Wang, Y., and An, Z.: Chemical Characteristics of PM_{2.5} and
691 PM₁₀ in Haze-Fog Episodes in Beijing, *Environ. Sci. Technol.*, *40*, 3148-3155, 2006.

692 Sun, Y., Jiang, Q., Wang, Z., Fu, P., Li, J., Yang, T., and Yin, Y.: Investigation of the Sources
693 and Evolution Processes of Severe Haze Pollution in Beijing in January 2013, *Journal of*
694 *Geophysical Research: Atmospheres*, *119*, 4380-4398, 10.1002/2014JD021641, 2014.

695 Sun, Y. L., Wang, Z., Dong, H., Yang, T., Li, J., Pan, X., Chen, P., and Jayne, J. T.:
696 Characterization of summer organic and inorganic aerosols in Beijing, China with an
697 Aerosol Chemical Speciation Monitor, *Atmos. Environ.*, *51*, 250-259,
698 10.1016/j.atmosenv.2012.01.013, 2012.

699 Sun, Y. L., Wang, Z., Fu, P., Jiang, Q., Yang, T., Li, J., and Ge, X.: The Impact of Relative
700 Humidity on Aerosol Composition and Evolution Processes during Wintertime in Beijing,
701 China, *Atmos. Environ.*, *77*, 927-934, <http://dx.doi.org/10.1016/j.atmosenv.2013.06.019>,
702 2013a.

703 Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.:
704 Aerosol composition, sources and processes during wintertime in Beijing, China, *Atmos.*
705 *Chem. Phys.*, *13*, 4577-4592, 10.5194/acp-13-4577-2013, 2013b.

706 Tian, L., Lucas, D., Fischer, S. L., Lee, S. C., Hammond, S. K., and Koshland, C. P.: Particle
707 and Gas Emissions from a Simulated Coal-Burning Household Fire Pit, *Environ. Sci.*
708 *Technol.*, *42*, 2503-2508, 10.1021/es0716610, 2008.

709 Tian, Y. Z., Wang, J., Peng, X., Shi, G. L., and Feng, Y. C.: Estimation of the direct and
710 indirect impacts of fireworks on the physicochemical characteristics of atmospheric
711 PM₁₀ and PM_{2.5}, *Atmos. Chem. Phys.*, *14*, 9469-9479, 10.5194/acp-14-9469-2014,
712 2014.

713 Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.:
714 Interpretation of organic components from Positive Matrix Factorization of aerosol mass
715 spectrometric data, *Atmos. Chem. Phys.*, *9*, 2891-2918, 2009.

716 Vecchi, R., Bernardoni, V., Cricchio, D., D'Alessandro, A., Fermo, P., Lucarelli, F., Nava, S.,
717 Piazzalunga, A., and Valli, G.: The impact of fireworks on airborne particles, *Atmos.*
718 *Environ.*, *42*, 1121-1132, 10.1016/j.atmosenv.2007.10.047, 2008.

719 Wang, Y., Zhuang, G., Sun, Y., and An, Z.: The variation of characteristics and formation
720 mechanisms of aerosols in dust, haze, and clear days in Beijing, *Atmos. Environ.*, *40*,
721 6579-6591, 2006.

722 Wang, Y., Zhuang, G., Xu, C., and An, Z.: The air pollution caused by the burning of
723 fireworks during the lantern festival in Beijing, *Atmos. Environ.*, *41*, 417-431,
724 10.1016/j.atmosenv.2006.07.043, 2007.

725 Wang, Y., Yao, L., Wang, L., Liu, Z., Ji, D., Tang, G., Zhang, J., Sun, Y., Hu, B., and Xin, J.:
726 Mechanism for the formation of the January 2013 heavy haze pollution episode over
727 central and eastern China, *Sci. China Earth Sci.*, *57*, 14-25, 10.1007/s11430-013-4773-4,
728 2014.

729 Yang, F., Tan, J., Zhao, Q., Du, Z., He, K., Ma, Y., Duan, F., and Chen, G.: Characteristics of
730 PM_{2.5} speciation in representative megacities and across China, *Atmos. Chem. Phys.*, 11,
731 5207-5219, 10.5194/acp-11-5207-2011, 2011.

732 Yang, L., Gao, X., Wang, X., Nie, W., Wang, J., Gao, R., Xu, P., Shou, Y., Zhang, Q., and
733 Wang, W.: Impacts of firecracker burning on aerosol chemical characteristics and human
734 health risk levels during the Chinese New Year Celebration in Jinan, China, *Sci. Total*
735 *Environ.*, 476-477, 57-64, <http://dx.doi.org/10.1016/j.scitotenv.2013.12.110>, 2014.

736 Yao, X., Chan, C. K., Fang, M., Cadle, S., Chan, T., Mulawa, P., He, K., and Ye, B.: The
737 water-soluble ionic composition of PM_{2.5} in Shanghai and Beijing, China, *Atmos.*
738 *Environ.*, 36, 4223-4234, Doi: 10.1016/s1352-2310(02)00342-4, 2002.

739 Zhang, H., Wang, S., Hao, J., Wan, L., Jiang, J., Zhang, M., Mestl, H. E. S., Alnes, L. W. H.,
740 Aunan, K., and Mellouki, A. W.: Chemical and size characterization of particles emitted
741 from the burning of coal and wood in rural households in Guizhou, China, *Atmos.*
742 *Environ.*, 51, 94-99, 10.1016/j.atmosenv.2012.01.042, 2012.

743 Zhang, J. K., Sun, Y., Liu, Z. R., Ji, D. S., Hu, B., Liu, Q., and Wang, Y. S.: Characterization
744 of submicron aerosols during a month of serious pollution in Beijing, 2013, *Atmos. Chem.*
745 *Phys.*, 14, 2887-2903, 10.5194/acp-14-2887-2014, 2014.

746 Zhang, M., Wang, X., Chen, J., Cheng, T., Wang, T., Yang, X., Gong, Y., Geng, F., and Chen,
747 C.: Physical characterization of aerosol particles during the Chinese New Year's firework
748 events, *Atmos. Environ.*, 44, 5191-5198, 10.1016/j.atmosenv.2010.08.048, 2010.

749 Zhang, Q., Jimenez, J., Canagaratna, M., Ulbrich, I., Ng, N., Worsnop, D., and Sun, Y.:
750 Understanding atmospheric organic aerosols via factor analysis of aerosol mass
751 spectrometry: a review, *Anal. Bioanal. Chem.*, 401, 3045-3067,
752 10.1007/s00216-011-5355-y, 2011.

753 Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z.,
754 Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM_{2.5} in
755 Beijing: seasonal perspective, *Atmos. Chem. Phys.*, 13, 7053-7074,
756 10.5194/acp-13-7053-2013, 2013.

757 Zhao, S., Yu, Y., Yin, D., Liu, N., and He, J.: Ambient particulate pollution during Chinese
758 Spring Festival in urban Lanzhou, Northwestern China, *Atmospheric Pollution Research*,
759 5, 335-343, doi: 10.5094/APR.2014.039, 2014.

760 Zhao, X. J., Zhao, P. S., Xu, J., Meng, W., Pu, W. W., Dong, F., He, D., and Shi, Q. F.:
761 Analysis of a winter regional haze event and its formation mechanism in the North China
762 Plain, *Atmos. Chem. Phys.*, 13, 5685-5696, 10.5194/acp-13-5685-2013, 2013.

763 Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., and Cass, G. R.:
764 Seasonal trends in PM_{2.5} source contributions in Beijing, China, *Atmos. Environ.*, 39,
765 3967-3976, DOI: 10.1016/j.atmosenv.2005.03.036, 2005.

766 Zhi, G., Chen, Y., Feng, Y., Xiong, S., Li, J., Zhang, G., Sheng, G., and Fu, J.: Emission
767 characteristics of carbonaceous particles from various residential coal-stoves in China,
768 *Environ. Sci. Technol.*, 42, 3310-3315, 2008.

769

770 **Figure Captions:**

771 **Fig. 1.** Time series of meteorological parameters (a) relative humidity (RH) and
772 temperature (T); (b) wind direction (WD) and wind speed (WS) at the height of 8m
773 and 100 m; mass concentrations of (c) PM_{2.5} and NR-PM₁ + BC and (d) submicron
774 aerosol species. The extinction coefficient (b_{ext}) at 630 nm is shown in (c). Three
775 events, i.e., Lunar New Year (LNY), Lunar Fifth Day (LFD) and Lantern Festival (LF)
776 with significant influences of fireworks are marked in (c). In addition, the classified
777 clean periods (CPs) and polluted events (PEs) are marked as shaded light blue and
778 pink areas, respectively.

779 **Fig. 2.** Time series of PM₁ species (Org, SO₄, NO₃, NH₄, Chl, K, KCl, and BC) and
780 meteorological variables (wind direction (100 m) and wind speed (8 m)) during three
781 firework events, i.e., (a) Lunar New Year, (b) Lunar Fifth Day, and (c) Lantern
782 Festival. The two blue arrow lines represent the starting and ending times of fireworks
783 events.

784 **Fig. 3.** Correlation of PM₁ vs. PM_{2.5} with the data segregated into three fireworks
785 events (LNY, LFD, and LF) and non-fireworks periods (NFW). The blank circles
786 represent FW data between 18:00 – 23:30 on 9 February which had large influences
787 from NFW sources.

788 **Fig. 4.** Average chemical composition of PM₁ and OA from fireworks and
789 background during three FW events.

790 **Fig. 5.** (a) Average mass spectra (MS) of OA during the firework period of Lunar
791 New Year (23:30, 9 February – 3:30, 10 February) and the period of background (BG,
792 4:30 – 11:00, 10 February). (b) Comparison of the difference spectrum from (a), i.e.,
793 MS_{FW+BG} – MS_{BG}, with the average LV-OOA spectrum in Ng et al.(2011a). Note that
794 five m/z 's, 37 (³⁷Cl⁺), 58 (NaCl⁺), 60 (Na³⁷Cl⁺), 74 (KCl⁺), and 76 (K³⁷Cl⁺/ ⁴¹KCl⁺)
795 marked in the figure were dominantly from fragmentation of inorganic salts during
796 fireworks.

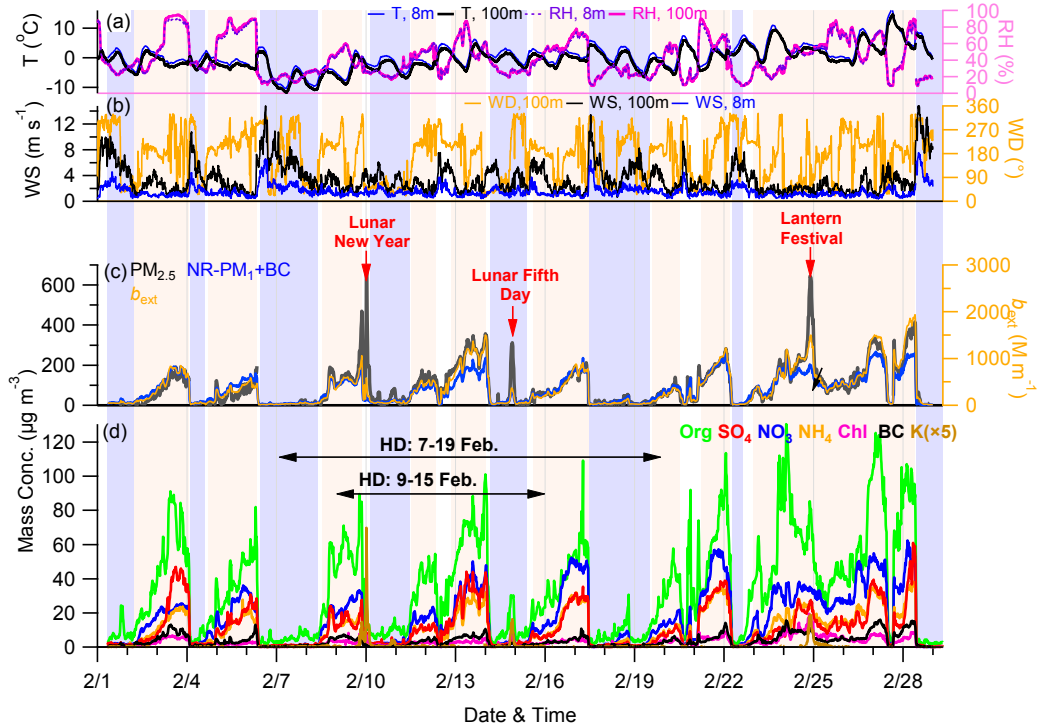
797 **Fig. 6.** Box plots of (a) mass concentrations and (b) mass fractions of aerosol species
798 for 9 pollution events marked in Fig. 1. The mean (cross), median (horizontal line),
799 25th and 75th percentiles (lower and upper box), and 10th and 90th percentiles (lower
800 and upper whiskers) are shown for each box.

801 **Fig. 7.** Left panel: variations of chemical composition of (a) organics, SNA (=sulfate
802 + nitrate + ammonium), and others (the rest species in PM₁); (b) SPM and PPM; and
803 (c) SOA and POA as a function of PM₁ and organics loadings, respectively. The

804 middle and right panels show the diurnal profiles of composition and mass
805 concentrations, respectively.

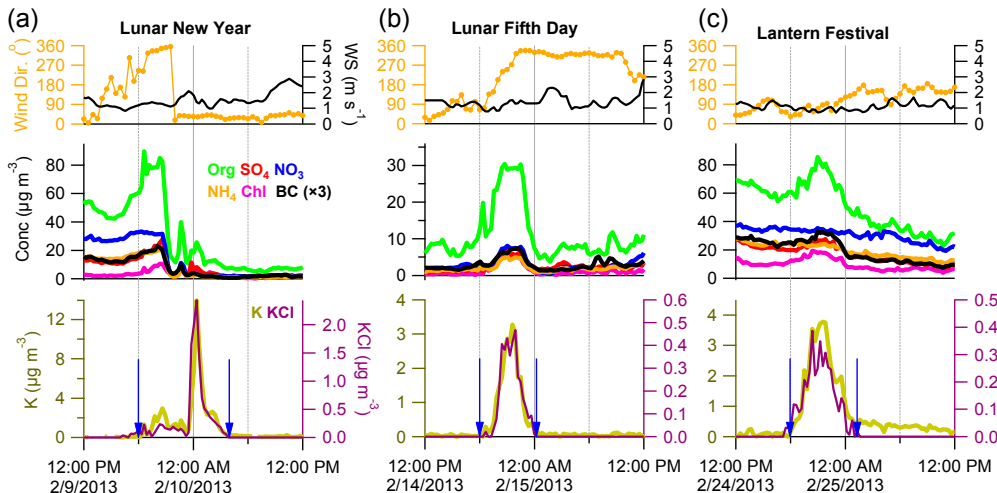
806 **Fig. 8.** (a) Average mass fraction of organics (f_{Org}) as a function of PM_{10} mass, and (b)
807 correlations of extinction coefficients ($\text{PM}_{2.5}$) vs. PM_{10} for 9 pollution events (PEs) and
808 9 clean periods (CPs) marked in Fig. 1. The error bar represents one standard
809 deviations of the average for each event.

810 **Fig. 9.** The average ratios of aerosol species, gaseous species, PM mass
811 concentrations, extinction coefficient, and meteorological parameters between holiday
812 (HD) and non-holiday (NHD) periods. Two different holidays, i.e., the official
813 holiday of 9 – 15 February and the longer holiday of 7 – 20 February were used for
814 averages. Also note that the averages were made by excluding clean periods and
815 firework events during both HD and NHD days. The error bars are the standard errors
816 of the ratios.



817

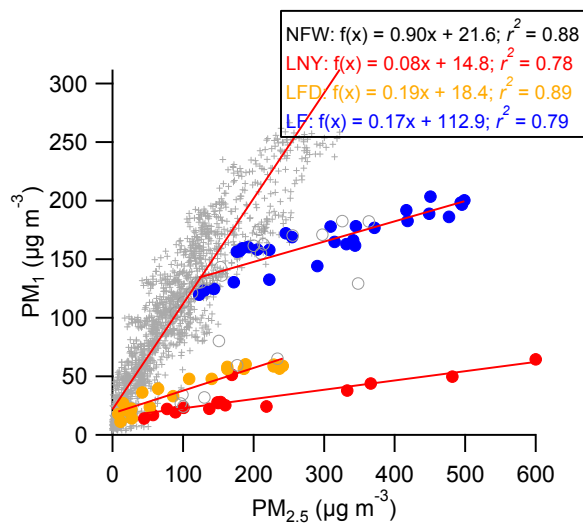
818 **Fig. 1.** Time series of meteorological parameters (a) relative humidity (RH) and
 819 temperature (T); (b) wind direction (WD) and wind speed (WS) at the height of 8m
 820 and 100 m; mass concentrations of (c) $PM_{2.5}$ and $NR-PM_1 + BC$ and (d) submicron
 821 aerosol species. The extinction coefficient (b_{ext}) at 630 nm is shown in (c). Three
 822 events, i.e., Lunar New Year (LNY), Lunar Fifth Day (LFD) and Lantern Festival (LF)
 823 with significant influences of fireworks are marked in (c). In addition, the classified
 824 clean periods (CPs) and polluted events (PEs) are marked as shaded light blue and
 825 pink areas, respectively.



826

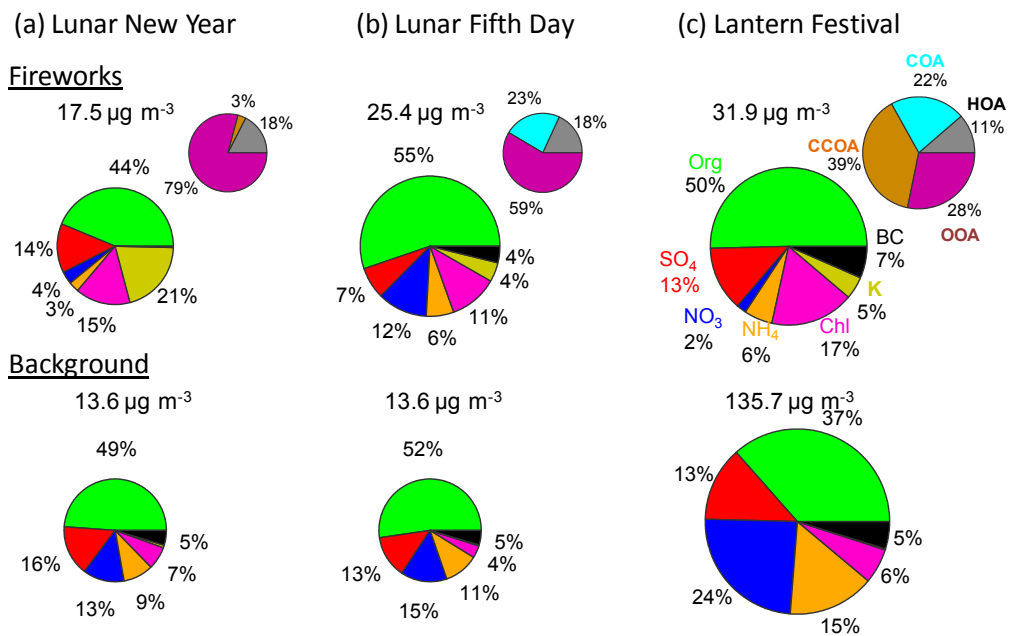
827 **Fig. 2.** Time series of PM₁ species (Org, SO₄, NO₃, NH₄, Chl, K, KCl, and BC) and
828 meteorological variables (wind direction (100 m) and wind speed (8 m)) during three
829 firework events, i.e., (a) Lunar New Year, (b) Lunar Fifth Day, and (c) Lantern
830 Festival. The two blue arrow lines represent the starting and ending times of fireworks
831 events.

832



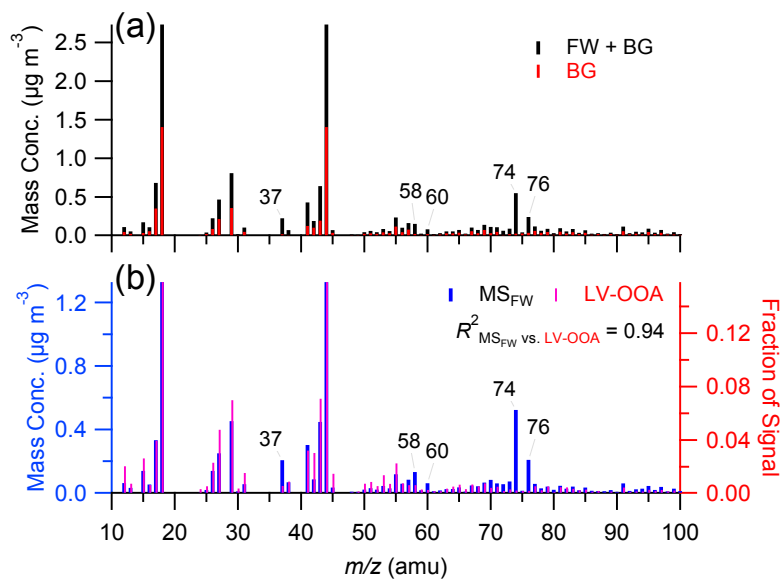
833

834 **Fig. 3.** Correlation of PM₁ vs. PM_{2.5} with the data segregated into three fireworks
835 events (LNY, LFD, and LF) and non-fireworks periods (NFW). The blank circles
836 represent FW data between 18:00 – 23:30 on 9 February which had large influences
837 from NFW sources.



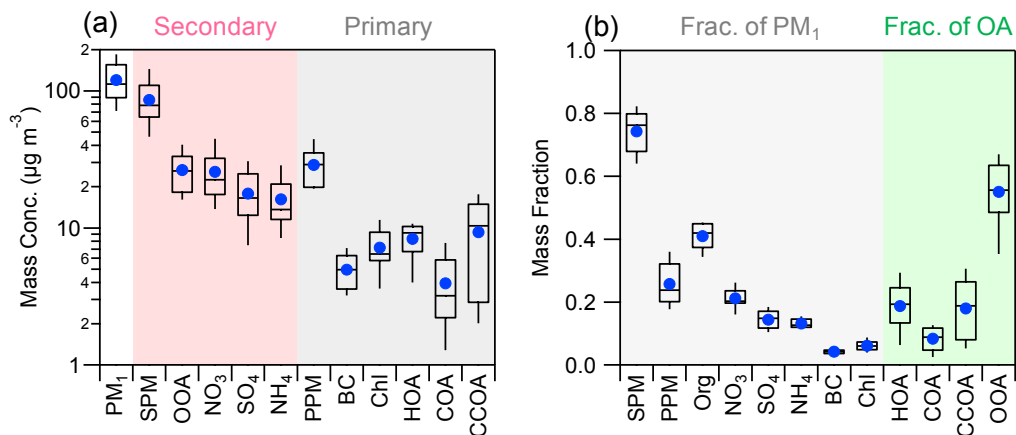
838

839 **Fig. 4.** Average chemical composition of PM_{10} and OA from fireworks and
 840 background during three FW events.



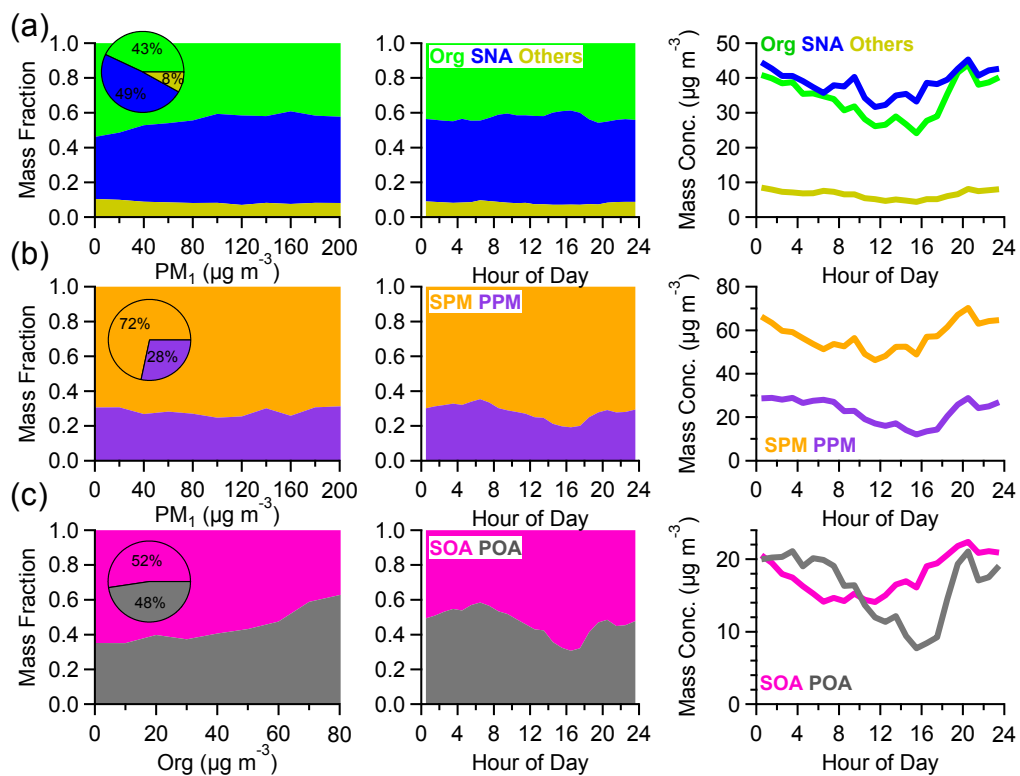
841

842 **Fig. 5.** (a) Average mass spectra (MS) of OA during the firework period of Lunar
 843 New Year (23:30, 9 February – 3:30, 10 February) and the period of background (BG,
 844 4:30 – 11:00, 10 February). (b) Comparison of the difference spectrum from (a), i.e.,
 845 $\text{MS}_{\text{FW+BG}} - \text{MS}_{\text{BG}}$, with the average LV-OOA spectrum in Ng et al.(2011a). Note that
 846 five m/z 's, 37 ($^{37}\text{Cl}^+$), 58 (NaCl^+), 60 ($\text{Na}^{37}\text{Cl}^+$), 74 (KCl^+), and 76 ($\text{K}^{37}\text{Cl}^+ / ^{41}\text{KCl}^+$)
 847 marked in the figure were dominantly from fragmentation of inorganic salts during
 848 fireworks.



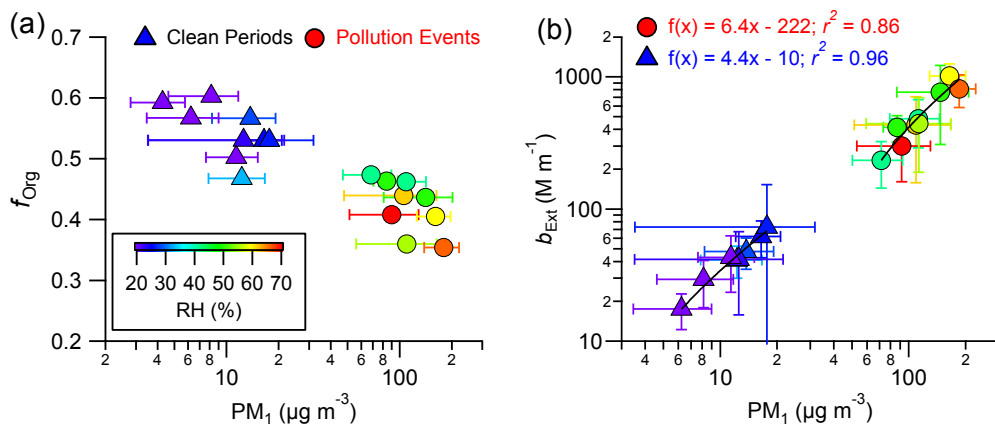
849

850 **Fig. 6.** Box plots of (a) mass concentrations and (b) mass fractions of aerosol species
 851 for 9 pollution events marked in Fig. 1. The mean (cross), median (horizontal line),
 852 25th and 75th percentiles (lower and upper box), and 10th and 90th percentiles (lower
 853 and upper whiskers) are shown for each box.



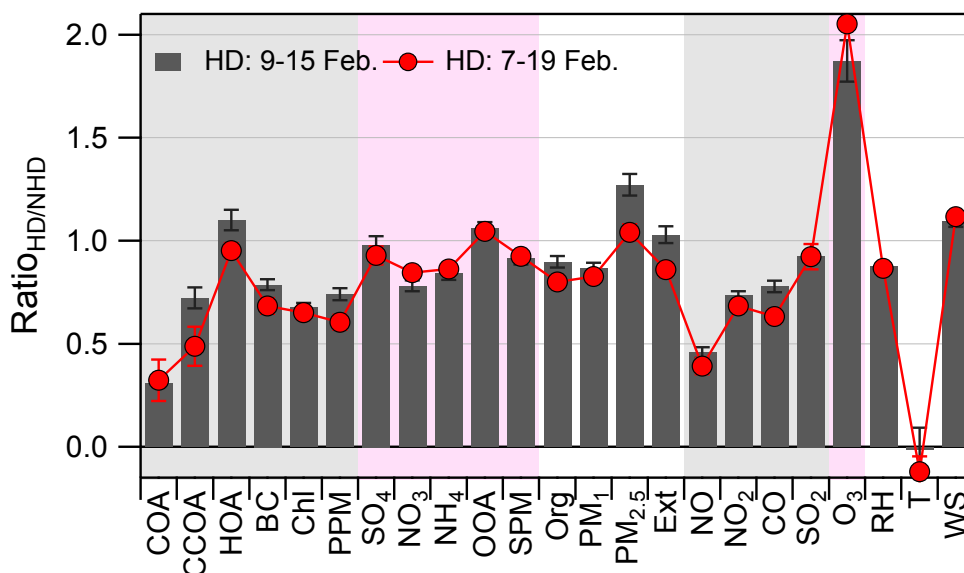
855

856 **Fig. 7.** Left panel: variations of chemical composition of (a) organics, SNA (=sulfate
 857 + nitrate + ammonium), and others (the rest species in PM_1); (b) SPM and PPM; and
 858 (c) SOA and POA as a function of PM_1 and organics loadings, respectively. The
 859 middle and right panels show the diurnal profiles of composition and mass
 860 concentrations, respectively.



861

862 **Fig. 8.** (a) Average mass fraction of organics (f_{Org}) as a function of PM_1 mass, and (b)
 863 correlations of extinction coefficients ($PM_{2.5}$) vs. PM_1 for 9 pollution events (PEs) and
 864 9 clean periods (CPs) marked in Fig. 1. The error bar represents one standard
 865 deviations of the average for each event.



866

867 **Fig. 9.** The average ratios of aerosol species, gaseous species, PM mass
 868 concentrations, extinction coefficient, and meteorological parameters between holiday
 869 (HD) and non-holiday (NHD) periods. Two different holidays, i.e., the official
 870 holiday of 9 – 15 February and the longer holiday of 7 – 20 February were used for
 871 averages. Also note that the averages were made by excluding clean periods and
 872 firework events during both HD and NHD days. The error bars are the standard errors
 873 of the ratios.