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February 15, 2015 Editor, Atmospheric Chemistry and Physics Dr. Yafang Cheng

Dear Dr. Cheng,

Please find in next pages our point-to-point responses to the comments on our manuscript offered by the two reviewers. The new version of the manuscript was significantly revised according to the reviewers' comments. Particularly, we significantly expanded the discussions on the uncertainties for the quantification of K^+ and the validation of PMF factors in section 2.3 in the revised manuscript. The reasons for not resolving biomass burning OA factor were also addressed in the main text. In addition, some major comments by reviewer #4 were further addressed in our response to reviewer #5.

We are confident that we have addressed all comments satisfactorily and that our revised manuscript meets the high quality standards of ACP. We 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 are thankful to the reviewer #5 for his/her constructive comments that help improve the manuscript significantly. Following the reviewer's suggestions, we have revised the manuscript accordingly. In particular, we significantly expanded the discussions on the uncertainties for the K⁺ quantification and the validation of PMF solution in the revised manuscript. The major comments by the reviewer #4 were also further addressed in our response to the reviewer #5. Listed below are our point-by-point responses in blue to the reviewers' comments

Response to Reviewers' comments

This paper described the submicron aerosol composition and sources during Chinese Spring Festival in Beijing, with a focus on firework and holiday effect. On-line ACSM was applied. OA source apportionment were done by PMF. The topic of focusing on influences of anthropogenic activity (i.e., firework and population reduction) on changes of aerosol and its sources is interesting. The author find short term effect of firework on PM1 for being a source of potassium, chloride, sulfate and oxygenated OA. By comparing the compositions of aerosol between Spring Festival and Non Spring Festival periods, the author find a larger reduction of primary species than secondary species due to the less anthropogenic activity in Beijing area, then conclude the controlling primary emission have a limited effect on air quality.

These are interesting results, however, some of the ideas in this paper are not so well organized and written. I firstly read the revised paper without reading comments. I found I have lots of the same questions as the other two reviewers do, e.g., the quantification of potassium with ACSM and the uncertainty of PMF source apportionment results. Although the authors answered all the comments in the responses, their responses to the other two reviewers are not well reflected in the revised manuscript. And also, some conclusions in this paper are needed more precise and careful deductions (see comments below). Thus, I recommend a major revision of this paper. The authors should address the questions below and add related necessary contents including the responses to the other reviewers into the paper.

We thank the reviewer's comments. Following the reviewer's suggestions, we significantly revised the manuscript. The uncertainties in quantification of K^+ and the chosen of PMF solution were addressed in detail in the revised manuscript. Some conclusions in the main text were also revised according to the reviewer's comments. In addition, some critical responses to the reviewer #4 were also included in the new version of the manuscript.

Major comments:

1) The points of this paper should be addressed more carefully. The logic and English writing in the responses to the comments are much better than the main text of this paper itself. Following the reviewer's suggestions, we significantly revised the manuscript in the new version by referring to our previous response to the comments of the reviewer #4. 2) About the quantification of potassium. Reviewer #4 already has a good comment on this issue. The authors should clearly address the large uncertainty of potassium measured in this study and let readers be aware of this issue, as well as KCl.

We agree with the reviewer and also the reviewer #4 that quantification of K^+ in this study might have large uncertainties. As the reviewer suggested, we significantly expanded the discussions on the K^+ quantification in section 2.3 in the revised manuscript letting the readers know the uncertainties of K^+ quantification with the ACSM.

"Quantification of K⁺ is challenging for ACSM because of a large interference of organic C₃H₃⁺ at m/z 39 and also uncertainties caused by surface ionization (Slowik et al., 2010). In this work, we found that m/z39 was tightly correlated with m/z 43 that is completely organics during non-fireworks (NFW) periods (r² = 0.87, slope = 0.45, Fig. S1). However, higher ratios of m/z 39/43 during FW periods were observed due to the elevated K^{+} signal from burning of fireworks. Assuming that m/z 39 was primarily contributed by organics during NFW periods, the excess m/z 39 signal, i.e., K⁺, can then be estimated as m/z 39 – m/z 43 × 0.45. The 41 K⁺ at m/z 41 was calculated using an isotopic ratio of 0.0722, i.e., 41 K⁺ =0.0722 × K⁺. The K⁺ signal was converted to mass concentration with a RIE of 2.9 that was reported by Drewnick et al. (2006). It should be noted that the quantification of K^{+} in this study might have a large uncertainty because of the unknown RIE of K^+ (RIE_k). The RIE_k can vary a lot depending on the tuning of the spectrometer and the temperature of the vaporizer. For example, Slowik et al. (2010) reported a RIE_{K} = 10 based on the calibration of pure KNO₃ particles using a ToF-AMS, which is much higher than the RIE_K = 2.9 obtained from the comparisons of K/S from fireworks and AMS measurements (Drewnick et al., 2006). In addition, the stability of surface ionization (SI) and electron impact (EI) also affects RIE_{κ} . We then checked the variations of the ratio of m/z 39/m/z 23 (two m/z's with similar surface ionization issues). The average ratio of m/z 39/23 during LFD and LF is 8.7 and 11.1, respectively, which is close to 9.0 during the NFW periods. The results suggest that the SI/EI ratio was relatively stable throughout the study. Because we didn't have collocated K measurement, RIE_K = 2.9 that was estimated from fireworks was used in this study. The quantified K^+ during LFD and LF on average contributed 4.5% and 4.7% of PM₁, respectively, which is close to ~5% (PM_{2.5}) reported by Cheng et al. (2014). Also, the large contribution of K^{+} to PM₁ (20.5%) during LNY, likely due to the intensified firework emissions (mainly firecrackers), is consistent with that (17.3%) observed during LNY 2014 in megacity Tianjin (Tian et al., 2014). Using RIE_{κ} =10 will decrease the K^+ concentration by a factor of more than 3, which appears to underestimate K^+ a lot. Therefore, RIE_K = 2.9 for the quantification of K⁺ in our study appear to be reasonable."

If it is possible, it will also be good that the authors can try KNO3/KCl calibration and validate the RIE for their own ACSM instrument. Even the tuning will not be exactly the same as it was in field campaign period. Based on the big differences of RIE of K+ reported in the literature, 2.9 (Drewnick et al., 2006) or 10 (Slowik et al., 2010), the calibration results on their own instruments should be at least more accurate than the literature values from different instruments with different fireworks. The reviewer gave us a good suggestion to address the uncertainties in quantification of K⁺. Unfortunately, we currently are unable to do such experiments for some reasons. RIE_K = 2.9 that was estimated from fireworks (Drewnick et al., 2006) was then used in this study. The quantified K⁺ during LFD and LF contributed 4.5% and 4.7% of PM₁, respectively, which is close to ~5% (PM_{2.5}) reported by Cheng et al. (2014). Also, the large contribution of K^+ to PM_1 (20.5%) during LNY is consistent with that (17.3%) observed during LNY 2014 in megacity Tianjin (Tian et al., 2014). Using the RIE_K=10 will decrease the K^+ concentration by a factor of more than 3, which appears to underestimate K^+ a lot. These results suggest that RIE_K = 2.9 appears to be more reasonable for our study.

3) I think review #4 is right, HOA and CCOA factors on current solution are either mixed with each other or at least should be switched. It looks like that the current spectrum of HOA in Fig. S5 shows significantly higher m/z 91, m/z 77, m/z 105 and m/z 105 (Please check), which is very similar to the CCOA spectrum in Hu et al. (2013) cited by the authors in the responses. Those ions are supposed to be contributed by PAH species in the coal combustion plumes (Hu et al., 2013). The diurnal variation of the current CCOA in paper shows similar trend to HOA in the morning but peaks during night. Huang et al. (2010) also shows a night peak in HOA factor in the summer of Beijing. Assuming the main sources of HOA that are vehicle emissions do not change very much among different seasons and years in Beijing, the current CCOA in this paper is more like HOA.

We agree with the two reviewers that there's possibility that HOA and CCOA were mixed each other in this study. Compared to other versions of AMS, ACSM using the commercial-grade spectrometer only scanned from m/z 10 – 150 (Ng et al., 2011) which missed the important information of large m/z's particularly the PAH-related m/z's. In addition, the m/z's above 50 have higher uncertainties than small m/z's because of the sharp decrease of ion transmission efficiency as a function of m/z. As a result, PMF of ACSM OA spectra might not separate HOA from CCOA as well as that of high resolution mass spectra. In addition, we don't have more external tracers to further separate them like what has been done in Sandradewi et al. (2008)

We defined HOA and CCOA for the reasons below:

- 1) The mass spectra of HOA and CCOA in this study are very similar to those resolved during wintertime 2011 2012 (Sun et al., 2013b)
- 2) The mass spectra of HOA in this study is very similar to that identified by PMF analysis of high resolution OA mass spectra in Beijing in winter 2013 (Zhang et al., 2014), which also showed pronounced m/z 91 and 115 peaks (not significant in CCOA)
- CCOA shows better correlation with chloride than HOA (r² = 0.41 vs. 0.24). Coal combustion is an important source of chloride during wintertime (Zhang et al., 2012).
- 4) CCOA shows much better correlation with m/z 60 than HOA ($r^2 = 0.77$ vs. 0.37)

The CCOA spectrum resolved by Hu et al. (2013) showed pronounced m/z's 77, 91, 105, and 115. At the same time, the HOA spectrum also showed pronounced m/z 77 and 91 peaks. The CCOA spectrum could be largely different depending on ageing extent, coal types, and coal burning conditions. For example, 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. The CCOA spectrum resolved by a HR-ToF-AMS in Beijing in January 2013 also didn't show pronounced 77, 91, 105, and 115 peaks as those in Hu et al. (2013). Therefore we cannot tell that HOA was switched from CCOA just based on the comparisons of these four pronounced m/z's.

The HOA is mainly from the emissions of diesel trucks and heavy duty vehicles that are not allowed inside the city between 6:00 am – 10:00 pm. This is consistent with the diurnal variation of HOA which showed a continuous increase from 10:00 pm to 5:00 – 6:00 am (Fig. S11). However, the diurnal variation of CCOA during this period was relatively constant. Note that CCOA showed a small peak at ~8:00 pm which agrees well with that of chloride. This also supports the indentified CCOA factor. The HOA in summer might be very different from that during wintertime which is suggested by their largely different spectra between Huang et al. (2010) and Zhang et al. (2014). In addition, the diurnal variation of HOA also depends on the location of the sampling site. Considering above, we believe that the current HOA and CCOA is reasonable. Following the reviewer's comments, we added more discussions on the judgment of HOA and CCOA in the main text.

About the BBOA influences, the authors should not only consider average f60. Because when OA is more aged, f60 will decrease. And also the influences of biomass burning may not exist for the whole campaign. Thus, please check the scatter plot of f44 vs f60 in this study, then compare it to biomass burning area in the scatter plot reported by Cubison et al. (2011). It is should be clear to see if there is biomass burning influences during the campaign.

We thank the reviewer's good suggestions. The scatter plot of f_{44} vs f_{60} for the entire study is shown in Fig. R1. Most of the data points are outside of the typical biomass burning region in f_{44} vs f_{60} (Cubison et al., 2011). Therefore no strong biomass burning influences were observed in this study. The average f_{60} for the entire study is ~0.42%, which is slightly higher than ~0.3% in the absence of biomass burning impact (DeCarlo et al., 2008;Ulbrich et al., 2009;Cubison et al., 2011). The slightly higher f_{60} is also likely due to: 1) the influence of coal combustion emissions, 2) the ACSM f_{60} was calculated as the fraction of m/z 60 in total OA signal (m/z 12 – 120). Therefore, BBOA could not be an important component of OA in this study, while the true components with <5% of the mass are unlikely to be retrieved accurately (Ulbrich et al., 2009).



Fig. R1. Scatter plot of f_{44} (ratio of m/z 44 to total OA signal) versus f_{60} (ratio of m/z 60 to total OA signal). Red circles represent 60-min average data. The two arrow lines represent a typical region influenced by biomass burning (Cubison et al., 2011), and the vertical dash line shows the typical f_{60} value (~0.3%) free from biomass burning.

4) Line 23: The authors said the contributions estimated in this work would represent the upper limits of firework. However, dilution effects may also impact the aerosols during the firework period, which will reduce the firework contributions to total aerosols. Fig.S9(a) shows clearly that when wind direction changed from NW to NE, all mass concentrations of main components in aerosols dropped substantially. It seems that particles were diluted or scavenged after 21:00 including firework period. This assumption is supported by the continuous decrease of the background OA concentrations from 21:00 on 2/9/13 to 12:00 2/10/13 in Fig. 9(a).

We concluded this mainly based on the uncertainties of our approach for the estimation of pure fireworks aerosols. Our approach will overestimate fireworks aerosols when there are strong local sources and/or regional transport during the fireworks periods. Therefore the fireworks quantified in this study are claimed to represent an upper limit.

We agree with the reviewer that the dilution effects will affect the absolute concentrations of background and fireworks aerosols, and hence the contribution of fireworks to the total particulate matter pollution during fireworks periods. In fact, such an effect can be demonstrated by the comparison of LNY and LF with largely different background aerosols. Although the FW-PM₁ concentration (31.9 μ g m⁻³) during LF is higher than that (17.5 μ g m⁻³) during LNY, the contribution of FW-PM₁ to the total PM₁ is lower because of higher background aerosols.

Fig. S9(a) shows a wind direction change during the early period of LNY which is associated with an increase of aerosol species particularly organics. Such variations were likely driven by the NFW sources, e.g., regional transport. Our approach will introduce large uncertainties in quantification of fireworks during this period. Therefore, we only quantified the fireworks aerosols between 23:30, 9 February and 3:30, 10 February when the background aerosol particles were cleaned to low levels by northern winds (Section 3.1).

5) Line 251-253: "Metals that were largely enhanced during FW period generally contribute a small fraction of PM (Wang et al., 2007; Vecchi et al., 2008). Please define "a small fraction". And also this statement is conflict to what author saw in this paper. Figure 3 shows that 21% of total PM1 in firework is contributed by K+, let alone the other metal species emitted by the firework.

Thanks the reviewer's carefulness. Metals here were referring to the ACSM un-detected elements including Mg, Sr, and Ba. These three elements have large increases during fireworks events yet contribute a small fraction of particulate mass, generally < 2% (Wang et al., 2007;Vecchi et al., 2008;Kong et al., 2014).

We clarified this point in the revised manuscript. Now it reads: "However, the ACSM un-detected metals (e.g., Mg, Sr, and Ba) that were largely enhanced during FW periods generally contribute a small fraction of PM (<2%) (Wang et al., 2007;Vecchi et al., 2008;Kong et al., 2014)."

6) Line252: "Therefore, our results suggest that burning of fireworks has the most impact on aerosol particles in the size range of $1 - 2.5 \mu m$ ". Very vague statement. What does "impact" mean? Is firework the source of particle with size range of $1 - 2.5 \mu m$, or through other effect?

We revised this sentence in the revised manuscript. Now it reads: "Therefore, our results might suggest that a large fraction of aerosol particles from the burning of fireworks was emitted in the size range of $1 - 2.5 \mu m$.

7) Line 272: The authors stated that it is not impossible for aqueous-phase formation of sulfate during firework, and regarded "sulfate in FW-PM1 was mainly from the direction emissions of FW". Is there any other observation that support sulfate can be emitted directly in firework? One reference in this paper: want et al. (2007) suggested sulfate may come from heterogeneous catalytic transformations of SO2. Fe could be the catalyst. Have the authors considered this probability?

We concluded that aqueous-phase formation of sulfate during fireworks periods would not be significant in this study for two reasons: 1) the relative humidity was low which is < 30% during LNY and LFD, and ~ 50% during LF. The aqueous-phase processing strongly depends on liquid water content that is positively related to relative humidity. The heterogeneous oxidation of SO₂ by catalyst (e.g., Fe and Mn) also depends on liquid water content. In Wang et al. (2007)'s study, the average relative humidity during the fireworks was 88% which is much higher than those in our study. In fact, Wang et al. (2007) defined this event as "fog" event. The heterogeneous oxidation of SO₂ under such a high humidity condition might play an important role; 2) The variations of sulfate are similar to chloride that was dominantly from the burning of fireworks. If the heterogeneous formation of sulfate played the dominant role during fireworks periods, these two species would have much different variations because of their different sources. Drewnick et al. (2006) also observed the very similar variations of sulfate, chloride, and potassium during the fireworks event. These results together suggest that burning of fireworks can directly emit sulfate particles.

8) Line 275-277: "most of it (nitrate) was likely from local sources and/or regional transport as supported by the large contributions of local". If nitrate can be contributed by regional transport during firework period, sulfate and other species also have the probability to be influenced by regional transport. This is conflict with what authors said that sulfate was mainly come from direct emission of firework. One literature reported that "Nitrate was mainly formed through homogeneous gas-phase reactions of NO2" during firework. "Fe could catalyze the formation of nitrate through the reaction of a-Fe2O3 with HNO3". (Wang et al., 2007). Vecchi et al. (2007) and Cheng et al. (2014) also found a rapid increase of NO3 with a factor of ~5 during firework period. In this paper, the authors saw various fraction of NO3 in different firework period. High fraction of nitrate (12%) in LFD in this paper was also reported. Why do authors still think the nitrate mainly come from local sources/or regional transport during firework? Have the authors considered other probability like Want et al. (2007) suggested?

The reviewer is right that sulfate and other species could be also influenced from regional transport. The contribution of regional transport during the Lunar Fifth Day is difficult to quantify. Because of this, we claimed in section 3.1 that "The FW contributions during LFD might also be overestimated due to the influences of regional transport as suggested by the wind direction change in the middle. Considering above, the contributions estimated in this work would represent the upper limits of FW."

If assuming (1) fireworks does not emit much nitrate and (2) nitrate was dominantly from regional transport during LFD, we can also see an evident fireworks influences on organics, sulfate, and chloride based on a comparison of their ratios to nitrate with a similar event (similar wind direction change) occurring before LNY without firework influences (12:00 - 18:00, 9 February). The ratios are ~4 for Org/NO₃, ~1 for SO₄/NO₃ and Chl/NO₃ during LFD, which are all higher than those (~2 for Org/NO₃, ~0.5 for SO₄/NO₃, and ~0.1 for Chl/NO₃) during the NFW event. The enhancements of organics, sulfate, and chloride were likely from the fireworks which are consistent with the other two episodes.

The homogeneous gas-phase reactions of NO₂ to form nitrate mainly occurs during daytime via OH radical oxidation. Because the fireworks event took place at night, homogeneous reaction would not play a significant role. Similar to sulfate, the heterogeneous reactions to form nitrate during three FW events could not be significant as well. The major path for nighttime nitrate formation is: NO₂+O₃ \rightarrow NO₃ + O₂, NO₂+NO₃ \rightarrow N₂O₅, N₂O₅ + H₂O \rightarrow 2HNO₃. The hydrolysis of N₂O₅ also depends on liquid water content which is positively related to relative humidity.

Wang et al. (2007) reported that the concentration of NO_3^- during the burning night (12 February) is 7 times higher than that observed in normal days. Such a conclusion was drawn mainly based on the comparison to a sample collected during 14 – 15 February (defined as normal days) which is already two days after the Lantern Festival. We would argue that such a comparison might have large uncertainties. In fact, the Fig. 2e in Wang et al. (2007) clearly showed that the nitrate concentration during the burning night was slightly lower than those in two samples collected right after the Lantern Festival. Vecchi et al. (2008) observed an increase factor of 5.2 for NO_3^- during the fireworks event. However, Fig. 5 in Vecchi et al. (2008) also showed an increase of factor of ~5 during the night of 6 – 7 July (free from fireworks) by calculating the similar ratio as the fireworks event. Comparatively, the fireworks tracers including Sr, Mg, Ba and K showed largely different behaviors from NO_3^- with the increase factor during the fireworks much higher than those free from fireworks. These results actually support that fireworks in Cheng et al. (2014) which defined the period of 15 – 30 January, 2012 (15 days) as the firework episode and the rest sampling days as the typical winter period.

All these results suggest that the traditional filter measurements might have large uncertainties in quantification of fireworks composition. The highly-time resolved measurements are needed for a better quantification of fireworks aerosols. Drewnick et al. (2006) conducted aerosol composition measurement using a ToF-AMS during the New Year's 2005 fireworks in Mainz, central Germany. The mass concentration of NO₃⁻ during the fireworks was relatively stable which is consistent with our conclusion that fireworks could not be a major source of NO₃⁻ particles. This also supports that high

concentration of NO₃⁻ during the LFD in this study was from other sources. Based on the investigation of the meteorological conditions and aerosol composition change, we concluded that it was likely from regional transport.

9) Changes of absolute mass concentrations of different species in PM1 between firework period and non-firework period should be considered as well.

Thank the reviewer's suggestions. The average mass concentrations of the total PM₁ are shown on the top of pie charts, which help readers understand the variations of absolute mass concentrations of different species during fireworks and non-fireworks periods.

Minor comments:

1) Experimental part: some important information of instrumentation should be more specifically addressed: What's the RH in the sampling line before ACSM, which will influence CE of ACSM quite a lot (Middlebrook et al., 2011). Did author use a cyclone before ACSM and Aethalometer? What are the detection limits of different species in ACSM? They may be important for clean period in Beijing. Following the reviewer's suggestions, more descriptions on the sampling and operations of the instruments were expanded in section 2.2 and 2.3 in the revised manuscript. In this study, a silica gel dryer in front of the ACSM and the CAPS monitor was used to dry aerosol particles. The relative humidity in the sampling line was generally below 40%. In addition, the measurement was conducted in winter. The temperature difference between outside and the sampling room was often larger than 10°C which also help to reduce the RH in the sampling line. Based on previous studies by Matthew et al. (2008) and Middlebrook et al. (2012), the RH only has a large impact on collection efficiency when it is >~ 80%. Therefore, the RH would have a minor effect on CE in this study.

For the ACSM and CAPS measurements, a URG $PM_{2.5}$ cyclone (Model: URG-2000-30ED) was used to remove coarse particles with aerodynamic diameters larger than 2.5 μ m. A $PM_{2.5}$ cyclone from the manufacturer (Magee Scientific Corp.) was used for the Aethalometer measurement. Such information is now added in the revised manuscript.

The 30-minture detection limits are 0.54, 0.06, 0.07, 0.25, and 0.03 μ g m⁻³ for organics, nitrate, sulfate, ammonium, and chloride, respectively which were reported in our previous study (Sun et al., 2012). The mass concentrations of organics (~2 – 3 μ g m⁻³) and sulfate, nitrate and ammonium (~0.5 – 1 μ g m⁻³) during clean periods were generally well above the detection limits. The chloride during most time of the clean periods is also above the detection limit. Several clean days with the mass concentration of chloride close to the detection limit were also observed, e.g., 28 February.

2) Figure S9 should be added into the main text. Most of the descriptions in 3.1 section are based on Figure S9. It is hard to figure out what the authors said based on Fig.1, e.g., line 223-225: the authors described the wind changed at 15:00-21:00 based on Fig.1. Whereas, it is not impossible to see that on Fig.1. Fig. S9 looks better.

Good point. Fig. S9 is moved to the main text in the revised manuscript.

3) Line 315-325: It is confusing that the authors talked about diurnal variations of several species based on their absolute mass concentrations but did not show them in graphs. These graphs should be at least added into supplementary information.

Thank the reviewer's comment. The diurnal profiles of the mass concentrations of aerosol species are now shown in Fig. 7.



Fig. R2. Left panel: variations of chemical composition of (a) organics, SNA (=sulfate + nitrate + ammonium), and others (the rest species in PM_1); (b) SPM and PPM; and (c) SOA and POA as a function of PM_1 and organics loadings, respectively. The middle and right panels show the diurnal profiles of composition and mass concentrations, respectively.

4) Line 327: "These results suggest that POA played more important roles than SOA in PM pollution during periods with high organic mass loadings.". Please define "high mass loading".

The contribution of POA exceeded that of SOA at the organic mass loading of ~ 60 μ g m⁻³ (Fig. 7c). Following the reviewer's suggestion, we revised this sentence as "These results suggest that POA played more important roles than SOA in PM pollution during periods with high organic mass loadings (e.g., > 60 μ g m⁻³)"

5) Line 59: It is not proper to use "despite this" here. It is not a contrast transition between two sentences. Similar to the usage of "despite this" in line 99. "Despite this" in line 59 was changed to "Therefore", and that in line 99 was changed to "However". 6) Line 61-63: "Fine particles from various source emissions can be either primary from direct emissions...or secondary from atmospheric oxidation of gas-phase species." "Secondary" is not "source emissions", author cannot category secondary aerosol to be emissions.

This sentence was revised as "Fine particles from various sources can be either primary from direct emissions, e.g., fossil fuel combustion and biomass burning, or secondary from atmospheric oxidation of gas-phase species".

7) Line 64: "one of the most important primary source", should be "sources" Changed

8) Line 75-76. "However, previous studies on chemical characterization of fireworks in China were mostly based on filter measurements with a time resolution of 12 h or 24 h...which may have large uncertainties in accurate quantification of chemical composition of FW particles".

Why do filter on firework studies have large uncertainties? The ACSM cannot detect metal and semirefractory species as the other filter-based techniques. It seems that the ACSM have larger uncertainties than filter method.

Because the fireworks events usually last less than 12 hours, the filter sampling with the duration more than 12 hours will introduce the influences of non-fireworks aerosol. In addition, the meteorological variables may have significant changes during fireworks leading to large variations of non-fireworks aerosol species. The quantification of fireworks based on the average differences before, during, and after fireworks may introduce further uncertainties due to the variations of background aerosol.

We agree with the reviewer that quantification of fireworks using ACSM measurements also has uncertainties, for example, the un-detected components, e.g., metals and the K^+ quantification. For non-refractory aerosol species except K^+ , ACSM measurements can quantify firework influences more accurately than filter measurements owning to the much higher time-resolution.

To avoid confusions, we clarified it in the revised manuscript. Now it reads: "Considering that the fireworks events usually last less than 12 hours, the filter analysis may introduce large uncertainties in accurate quantification of chemical composition of FW particles due to either the interferences of NFW background aerosols or the difficulties to account for meteorological variations."

9) Line 83-84: Reference missing: "Secondary aerosol is of more concern compared to primary aerosol because it is formed over regional scales and exerts impacts on air quality over wider areas (Reference)." Some related references (Matsui et al., 2009;DeCarlo et al., 2010) were added in the revised manuscript.

10) Line89-92: Reference missing: "SIA was observed to contribute a large fraction of PM2.5 and played an enhanced role during haze episodes due to the faster heterogeneous reactions associated with higher humidity. --(Reference) While SIA was relatively well characterized, secondary organic aerosol (SOA) is poorly understood.-- (Reference)"

Some related references (Liu et al., 2013;Sun et al., 2013a;Zhao et al., 2013;Huang et al., 2014;Sun et al., 2014;Wang et al., 2014) were added in the revised manuscript.

11) Line 99: "Despite this, the role of SOA in fine particle pollution in Beijing is not well known, in particular during wintertime" What's kind of role of SOA unknown? If the author talked about the SOA mass contribution to total aerosol or OA. Several papers already talked about the fraction of SOA in Beijing winter (Huang et al., 2014;Sun et al., 2014;Zhang et al., 2014).

Thanks the reviewer's comments. The original description was not clear. We rewrote it in the revised manuscript. Now it reads:

"Recently, the role of SOA in fine particle pollution during wintertime – a season with frequent occurrences of pollution episodes in Beijing was extensively investigated and the results highlighted the similar importance of SOA to SIA (Sun et al., 2013b;Sun et al., 2014;Zhang et al., 2014). However, the role of SOA in particulate pollution during periods with largely reduced anthropogenic activities is not well known yet (Huang et al., 2012). This study happened to take place in a month with the most important holiday in China, i.e., the Spring Festival...."

Comments related to Figures:

Fig.1: Fig.1 shows two blue lines on the top 2 panels without labeling. It is hard to tell the time series of K+ from other traces based on the bottom panel.

The two blue lines were described in the caption of Fig. 1, which is "Also shown in (a) and (b) is the temperature and wind speed at the height of 8 m which are in blue". They are now labeled in the figure.

The time series of K^+ was zoomed by a factor of 5 in Fig. 1 for a better reading. In addition, the original Fig. S9 with a clear time series of K^+ during three FW events was moved into the main text following the reviewer's suggestions.

Fig.3: The label of "chloride" should be consistent with other ions. "Chl" should be "Cl-" The labels of the pie chart in Fig. 3 are now revised to be consistent with those in other figures.

Fig.4: It is not proper to say this is the spectrum of OA in FW, since the pronounced m/z 74 also contains KCl and m/z 37 is enhanced by HCl. It will cause confuses.

The reviewer pointed out a good point. These several m/z's have strong influences from inorganics during fireworks, yet they also have contributions from organic aerosol. For clarification, we added a sentence in the caption which is "Note that five m/z's, 37 (³⁷Cl⁺), 58 (NaCl⁺), 60 (Na³⁷Cl⁺), 74 (KCl⁺), and 76 (K³⁷Cl⁺/ ⁴¹KCl⁺) marked in the figure were dominantly from fragmentation of inorganic salts during fireworks."

Fig.8: Why is PM2.5 enhanced during non-holiday periods but PM1 decrease.

 PM_1 showed an average reduction of 13% during HD while $PM_{2.5}$ showed an increase by 27%. One possible explanation is likely due to the increases of aerosol species in the size range of $1 - 2.5 \mu m$ during HD period (e.g., 13 February).

Fig.12: Uncertainties of ratios for different species between HD/NHD should be added. The uncertainties of ratios (standard errors of the means) are now added in Fig. 9 and Fig. S12.

Fig. S2: Please show the scatter plot of K+ and KCl. The scatter plot of KCl⁺ vs. K⁺ is now shown in Fig. S2c.

Fig.S7 The time series of external tracers corresponding to each factor should be shown as well, since author already described them in the comments. It is more direct to see the graph.

The time series of external tracers including CO, NO_x , BC, NO_3^- , and m/z 60 are shown in Fig. S2. Also shown is the mass spectra of four OA factors identified in our previous study during winter 2011 – 2012 (Sun et al., 2013b).

Fig. S11 There is no shaded areas on the graph as caption described.

The shaded areas were added in updated Fig. S6

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1	Aerosol Composition and Sources during the Chinese Spring
2	Festival: Fireworks, Secondary Aerosol, and Holiday Effects
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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	aerosol composition and sources during the Chinese Spring Festival,- of 2013. Three
23	fireworks-(FW) events exerting significant and short-term impacts on fine particles
24	$(PM_{2.5})$ were observed on the days of Lunar New Year, Lunar Fifth Day, and Lantern
25	Festival. The FW showed major-large impacts on non-refractory potassium, chloride,
26	sulfate, and organics in submicron aerosol (PM1), of which the FW organics appeared
27	to be emitted mainly in secondary with its mass spectrum resembling to that of
28	secondary organic aerosol (SOA). Pollution events (PEs) and clean periods (CPs)
29	alternated routinely throughout the study. Secondary particulate matter (SPM = $SOA +$
30	sulfate + nitrate + ammonium) dominated the total PM ₁ mass on average accounting for
31	63-82% during the nine PEs observed in this study. The elevated contributions of
32	secondary species during PEs resulted in a higher mass extinction efficiency of PM_1
33	$(6.4 \text{ m}^2 \text{ g}^{-1})$ than that during CPs $(4.4 \text{ m}^2 \text{ g}^{-1})$. The Chinese Spring Festival also provides
34	a unique opportunity to study the impacts of reduced anthropogenic emissions on
35	aerosol chemistry in the city. The primary species showed ubiquitous reductions during
36	the holiday period with the largest reduction for cooking OA (69%), nitrogen monoxide
37	(54%), and coal combustion OA (28%). The secondary sulfate, however, remained
38	minor change, and the SOA and the total $PM_{2.5}$ even slightly increased. These-Our
39	results have significant implications that controlling local primary source emissions
40	during PEs, e.g., cooking and traffic activities, might have limited effects on improving
41	air quality during PEs -in megacity Beijing when due to the dominance of SPM that is 2

42 formed over regional scales from regional transport dominates in aerosol particle

43 composition.

44 1 Introduction

45 Air pollution caused by fine particles $(PM_{2.5})$ is of great concern in densely populated megacities because of its adverse effects on human health and regional air 46 quality (Molina and Molina, 2004; Chan and Yao, 2008). The health risk of air 47 48 pollution is greater than expected leading to around 7 million people's death in 2012 according to the latest report by World Health Organization 49 (http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/). The Beijing 50 51 metropolitan area is one of the most populous megacities in the world with the population reaching 20.69 million by the end of 2012 (Beijing Municipal Bureau of 52 53 Statistics). According to Beijing Municipal Environmental Protection Bureau, the annual average concentration of $PM_{2.5}$ was 89.5 µg m⁻³ in 2013, which is about 2.5 54 times the National Ambient Air Quality Standards of China (35 µg m⁻³ for annual 55 average), which This indicates suggests severe fine particle pollution in Beijing. As-56 57 a result, eExtensive studies have been made recently to investigate the chemical 58 composition and sources of PM_{2.5}. The results showed that secondary inorganic aerosol (SIA = sulfate + nitrate + ammonium), coal combustion, traffic emissions 59 (gasoline and diesel), biomass burning, cooking emissions and dust are the major 60 sources of PM_{2.5} (Zheng et al., 2005;Song et al., 2006;Zhang et al., 2013), and. 61 62 However, the source contributions varied significantly among different seasons-, 63 Despite this, therefore improving air quality in Beijing remains a great challenge due to the very complex sources and dynamic evolution processes of aerosol particles. 64 65 Fine particles from various sources emissions can be either primary from direct emissions, e.g., fossil fuel combustion and biomass burning, or secondary from 66 atmospheric oxidation of gas-phase species. The fireworks (FW) is one of the most 67

68	important primary sources that can exert significant and short-time impacts on air
69	quality. The fireworks burning emits a large amount of gaseous pollutants, e.g., sulfur
70	dioxide (SO ₂) and nitrogen oxide (NO _x) (Vecchi et al., 2008;Huang et al., 2012), and
71	also fine particles comprising organic/elemental carbon, sulfate, potassium, chloride
72	and various metals, e.g., copper (Cu), barium (Ba), strontium (Sr) and magnesium
73	(Mg) (Moreno et al., 2007;Wang et al., 2007;Li et al., 2013). The enhanced short-term
74	air pollution by fireworks can substantially increase health risk levels (Godri et al.,
75	2010;Yang et al., 2014) and reduce visibility for hours (Vecchi et al., 2008)However,
76	^p Previous studies on chemical characterization of fireworks in China were mostly
77	based on filter measurements with a time resolution of 12 h or 24 h (Wang et al.,
78	2007;Zhang et al., 2010;Feng et al., 2012;Huang et al., 2012;Cheng et al., 2014;Zhao
79	et al., 2014),. Considering that the fireworks events usually last less than 12 hours, the
80	filter analysis may which may have introduce large uncertainties in accurate
81	quantification of chemical composition of FW particles due to either the interferences
82	of non-FW (NFW) background aerosols or the difficulties to account for
83	meteorological variations. Drewnick et al. (2006) first conducted real-time
84	size-resolved chemical composition measurements during the New Year's period in
85	Mainz, Germany using an Aerodyne Time-of-Flight Aerosol Mass Spectrometer
86	(ToF-AMS). To our knowledge, there are no such real-time measurements of
87	chemical composition of aerosol particles during fireworks events in China yet, which
88	limits our understanding on the rapid formation and evolution of fireworks events,
89	and also their impacts on air-particulate matter (PM) pollution.
90	Secondary aerosol is of more concern compared to primary aerosol because it is
91	formed over regional scales and exerts impacts on air quality over wider areas (Matsui

92	et al., 2009;DeCarlo et al., 2010). Therefore, extensive studies have been conducted in
93	recent years to characterize the sources and formation mechanisms of secondary
94	aerosol (Yao et al., 2002;Duan et al., 2006;Sun et al., 2006;Wang et al., 2006;Guo et
95	al., 2010;Yang et al., 2011;Sun et al., 2013b;Zhang et al., 2013;Zhao et al., 2013).
96	SIA was observed to contribute a large fraction of PM _{2.5} and played an enhanced role
97	during haze episodes due to the faster heterogeneous reactions associated with higher
98	humidity (Liu et al., 2013;Sun et al., 2013a;Zhao et al., 2013;Sun et al., 2014;Wang et
99	al., 2014). While SIA was relatively well characterized, secondary organic aerosol
100	(SOA) is poorly not well understood (Huang et al., 2014). The recent deployments of Formatted: Font color: Blue
101	Aerodyne Aerosol Mass Spectrometers (AMS) greatly improved our understanding
102	on sources and evolution processes of organic aerosol (OA) in China, and also the
103	different roles of primary organic aerosol (POA) and SOA by positive matrix-
104	factorization (PMF) of organic mass spectra in PM pollution (Huang et al., 2010;Sun
105	et al., 2010;He et al., 2011;Sun et al., 2012;Sun et al., 2013b;Zhang et al., 2014). SOA
106	was found to play different roles among different seasons. While SOA is more
107	significant in summer (Huang et al., 2010;Sun et al., 2010;Sun et al., 2012), POA
108	generally plays a more important role during wintertime (Sun et al., 2013b). Recently,
109	the role of SOA in fine particle pollution during wintertime – a season with frequent
110	occurrences of pollution episodes in Beijing was extensively investigated and the
111	results highlighted the similar importance of SOA to SIA (Sun et al., 2013b; Sun et al., Formatted: Font color: Blue
112	2014;Zhang et al., 2014). Despite this However, the role of SOA in fine-
113	particle particulate pollution during periods with largely reduced anthropogenic
114	activities in Beijing is not well known yet (Huang et al., 2012), in particular during
115	wintertime, a season with frequent occurrences of pollution episodes (Sun et al.,
116	2013b;Zhang et al., 2014) . Of particular interest, tThis study happened to took-take

117	place in the a month with the most important holiday in China, i.e., the Spring	
118	Festival. The source emissions (e.g., traffic and cooking) had have significant changes	
119	due to a large reduction of population and anthropogenic activities in the city. This	
120	provides a unique opportunity to investigate how source changes affect aerosol	
121	chemistry including primary emissions and secondary formation in Beijing. Although	
122	Huang et al. (2012) investigated such a holiday effect on aerosol composition and	
123	optical properties in Shanghai, the data analyses were limited by daily average	
124	composition measurements and also the significantly different meteorological	
125	conditions between holiday and non-holiday periods.	
126	In this study, an Aerosol Chemical Speciation Monitor (ACSM) along with	
127	various collocated instruments was deployed in Beijing during February 2013. The	
128	chemical composition of submicron aerosol (PM ₁) from fireworks is first quantified in	Formatted: Not Superscript/ Subscript
129	Beijing-based on the highly time – resolved – resolution-measurements of	
130	non-refractory submicron aerosol (NR-PM) species (organics, sulfate, nitrate,	Formatted: Subscript
131	ammonium, chloride, and potassium) and black carbon. The impacts of fireworks on	
132	PM pollution during Chinese Lunar New Year (LNY), Lunar Fifth Day (LFD), and	
133	Lantern Festival (LF) are investigated, and the roles of secondary formation in PM	
134	pollution are elucidated. Further, the impacts effects of reduced anthropogenic	
135	emissions during the holiday on primary and secondary aerosols in the city are	
136	illustrated, which has significant implications for making air pollution control	
137	strategies in Beijing.	
138	2 Experimental	
139	2.1 Sampling site	
140	The measurements in this study were conducted at the Institute of Atmospheric 7	

141	Physics (IAP), Chinese Academy of Sciences (39°58'28''N, 116°22'16''E), an urban	
142	site located between the north third and fourth ring road in Beijing (Sun et al., 2012).	
143	Aerosol eharaeterization measurements was were performed from 1 February to 1	
144	March 2013, during which when three episodes with significant influences of	
145	fireworks, i.e., Lunar New Year (LNY), Lunar Fifth Day (LFD), and Lantern Festival	
146	(LF), were observed (Fig. 1). The meteorological conditions during the measurement	
147	period are reported in Fig. 1. Winds at the ground surface were generally below 2 m	
148	s^{-1} and temperature averaged 0.6 °C. Relative humidity (RH) varied periodically with	
149	higher values generally associated with higher particulate matter (PM) pollution.	
150	2.2 Aerosol sampling	
151	The chemical composition of non-refractory submicron aerosol particles (NR-PM ₁)	
152	including organics, sulfate, nitrate, ammonium, and chloride were measured on linein	
153	situ by an Aerodynethe Aerosol Chemical Speciation Monitor (ACSM) at an	
154	approximate 15-min time intervals (Ng et al., 2011b). The ACSM has been widely	
155	used for long-term and routine aerosol particle composition measurements due to its	
156	robustness (Sun et al., 2012;Budisulistiorini et al., 2014;Petit et al., 2014) despite its	
157	lower sensitivity and mass resolution compared to previous versions of research-grade	
158	AMS (Jayne et al., 2000; DeCarlo et al., 2006). The ACSM shares the same aerosol-	Field Code Changed
159	sampling, vaporization and ionization modules as The ACSM was built upon previous-	
160	versions of AMS (Jayne et al., 2000;DeCarlo et al., 2006). However, the ACSM has-	
161	no size information and also a lower sensitivity and lower mass resolution due to the	
162	use of a commercial grade quadrupole mass analyzer (Ng et al., 2011b). The-	
163	advantage of the ACSM is its robustness for long-term and routine aerosol particle-	
164	composition measurements. In this study, the ambient air was drawn inside the	

165	sampling room at a flow rate of 3 L min ⁻¹ , of which ~ 0.1 L min ⁻¹ was sub-sampled	
166	into the ACSM and 0.85 L min ⁻¹ into a Cavity Attenuated Phase Shift Spectrometer	Formatted: Superscript
167	(CAPS) particle extinction monitor (Massoli et al., 2010). A PM _{2.5} cyclone (Model:	
168	URG-2000-30ED) was supplied in front of the sampling line to remove coarse	
169	particles with aerodynamic diameters larger than 2.5 μ m. The aerosol particles were	
170	dried by a silica gel dryer (RH $<$ 40%) before entering the ACSM and the CAPS. The	
171	ACSM was operated at a scan rate of 500 ms amu ⁻¹ for the mass spectrometer from	
172	m/z 10 – 150. Because ACSM cannot detect refractory components, e.g., BC and	
173	mineral dust, Aa two-wavelength Aethalometer (Model AE22, Magee Scientific Corp.)	
174	was therefore used to measure refractory black carbon (BC in PM _{2.5}) that the ACSM-	
175	eannot detect. The light extinction of dry fine particles (b_{ext} , 630 nm) was measured at	
176	1 s time resolution with a precision (3 σ) of 1 M m ⁻¹ by the CAPS monitor.	
177	The light extinction of dry fine particles (b_{ext} , M m ⁻¹ , 630 nm) was measured by a \star	Formatted: Indent: First line: 0.25"
178	Cavity Attenuated Phase Shift Spectrometer particle extinction monitor, CAPS PMex-	
179	(Massoli et al., 2010). The CAPS PMex was measured at 1 s time resolution with a	
180	precision (3 σ) of 1 M m⁻¹. In addition, – T the mass concentration of PM _{2.5} was	
181	determined by a heated Tapered Element Oscillating Microbalance, TEOM, and the	
182	collocated gaseous species (including CO, SO ₂ , NO, NO _x and O_3) were measured by	
183	various gas analyzers (Thermo Scientific) at 1 min time resolution. The-A more	
184	detailed descriptions of aerosol and gas measurements were given in Sun et al.	
185	(2013b).	
186	2.3 ACSM data analysis	
187	The ACSM data were analyzed for the mass concentrations and chemical	

188 composition of NR-PM₁ using standard ACSM software (v 1.5.3.2) written within

9

189	Igor Pro (WaveMetrics, Inc., Oregon USA). A composition-dependent collection
190	efficiency (CE) recommended by Middlebrook et al.(2012), $CE = max (0.45, 0.0833)$
191	+ 0.9167 × ANMF), was used to account for the incomplete detection due to the
192	particle bouncing effects (Matthew et al., 2008) and the influences caused by high
193	mass fraction of ammonium nitrate (ANMF). Because aerosol particles were overall
194	neutralized (NH ₄ ⁺ _{measured} /NH ₄ ⁺ _{predicted} = 1.01, r^2 = 0.99) and also dried before entering
195	the ACSM, the effects of particle acidity and RH would be minor (Matthew et al.,
196	2008;Middlebrook et al., 2012). The default relative ionization efficiencies (RIEs)
197	except ammonium (RIE = 6.5) that was determined from the IE calibration were used
198	in this study, $except$ ammonium (RIE = 6.5) that was determined from IE calibration.

Quantification of K⁺ is challenging for ACSM because of a large interference of 199 organic $C_3H_3^+$ at m/z 39 and also uncertainties caused by surface ionization (Slowik et 200 al., 2010). In this work, we found that m/z 39 was tightly correlated with m/z 43 that is 201 completely organics during non-fireworks (NFW) periods ($r^2 = 0.87$, slope = 0.45, Fig. 202 S1). However, higher ratios of m/z 39/43 during FW periods were observed due to the 203 elevated K^+ signal from burning of fireworks. Assuming that m/z 39 was primarily 204 contributed by organics during NFW periods, the excess m/z 39 signal, i.e., K⁺, can 205 then be estimated as $m/z 39 - m/z 43 \times 0.45$. The ⁴¹K⁺ (at m/z 41) was calculated using 206 its an isotopic ratio of 0.0722, i.e., ${}^{41}K^+ = 0.0722 \times K^+$. The K⁺ signal was converted to 207 mass concentration with a RIE of 2.9 that was reported by Drewnick et al. (2006). It 208 should be noted that the quantification of K⁺ in this study might have a large 209 210 uncertainty because of the unknown RIE of K^+ (RIE_K). The RIE_K can vary a lot 211 depending on the tuning of the spectrometer and the temperature of the vaporizer. For example, Slowik et al. (2010) reported a $RIE_{K} = 10$ based on the calibration of pure 212 KNO3 particles using an Aerodyne time of flight aerosol mass spectrometera 213 (e-ToF-AMS), which is much higher than the $RIE_K = 2.9$ obtained from the 214 215 comparisons of K/S from fireworks and AMS measurements (Drewnick et al., 2006).

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216	In addition, the stability of surface ionization (SI) and electron impact (EI) also affects
217	RIE _K . We then checked the variations of the ratio of m/z 39/ m/z 23 (two m/z 's with
218	similar surface ionization issues). The average ratio of m/z 39/23 during LFD and LF
219	is 8.7 and 11.1, respectively, which is close to 9.0 during the NFW periods. The
220	results suggest that the SI/EI ratio was relatively stable throughout the study. Because
221	we didn't have collocated K measurement, $RIE_K = 2.9$ that was estimated from
222	fireworks was used in this study. The quantified K^+ during LFD and LF on average
223	contributed 4.5% and 4.7% of PM ₁ , respectively, which is close to \sim 5% (PM _{2.5})
224	reported by Cheng et al. (2014). Also, the large contribution of K^+ to PM_1 (20.5%)
225	during LNY, likely due to the intensified firework emissions (mainly firecrackers), is
226	consistent with that (17.3%) observed during LNY 2014 in megacity Tianjin (Tian et
227	al., 2014). Using $RIE_{K}=10$ will decrease the K ⁺ concentration by a factor of more than
228	3, which appears to underestimate K^+ a lot. Therefore, $RIE_K = 2.9$ for the
229	quantification of K^+ in our study appear to be reasonable-for our study. The large-
230	contribution of K during LNY was likely due to the intensified firework emissions-
231	(mainly firecrackers), especially between 0:00 – 3:00 am.
232	The KCl ⁺ (m/z 74) and ⁴¹ KCl ⁺ /K ³⁷ Cl ⁺ (m/z 76) were estimated by the differences
233	between the measured and PMF modeled m/z 74 (see Fig. S2 for details). Not
234	surprisingly, the quantified KCl ⁺ highly correlates with K ⁺ ($r^2 = 0.82$, Fig. S2c). The
235	chloride concentration was also biased at m/z 35 during some periods (e.g., LNY, Fig.
236	S3) mainly), which is likely due to the inappropriate default fragment ratio at m/z -
237	35 interferences of NaCl from fireworks. Therefore, Cl^+ (m/z 35) was recalculated
238	based on its correlation with m/z 36 (mainly HCl ⁺ with negligible C ₃ ⁺ and ³⁶ Ar), i.e.,
239	m/z 35 = 0.15 × m/z 36 , . The and ³⁷ Cl ⁺ was calculated using an isotopic ratio of 0.323,
240	i.e., ${}^{37}Cl^+ = 0.323 \times {}^{35}Cl^+$. A-The comparison of the time series of the reconstructed
241	chloride from the default values and recalculated chloride is shown in Fig. S3b.
242	The positive matrix factorization (PMF) with the algorithm of PMF2.exe in robust

243	mode (Paatero and Tapper, 1994) was performed on organic aerosol (OA) mass
244	spectra (m/z 12 – 120) to resolve distinct OA components from different sources. The
245	PMF results were evaluated with an Igor Pro-based PMF Evaluation Tool (PET, v
246	2.04) (Ulbrich et al., 2009) following the procedures detailed in Zhang et al. (2011).
247	After a careful evaluation of the spectral profiles, diurnal variations and correlations
248	with external tracers, a 6-factor solution with rotational forcing parameter fpeak $= -1$
249	$(Q/Q_{exp} = 4.3)$ was chosen, yielding a hydrocarbon-like OA (HOA), a cooking OA
250	(COA), a coal combustion OA (CCOA), and three oxygenated OA (OOA)
251	components. Because of the absence of collocated measurements to validate the
252	different OOA components, the three OOA components were , which were-
253	recombined into one OOA component. The contributions of four OA factors were
254	relatively stable across different fpeak values (average $\pm 1\sigma$; min – max, Fig. S4):
255	HOA (14±1.6%; 12 – 16%), COA (14±2.8%; 11 – 17%), CCOA (19±2.7%; 15 –
256	22%), and OOA (51 \pm 1.7; 49 – 55%). However, considering the mass spectra of OA
257	factors at fpeak = -1 presented the best correlation with those identified in winter
258	2011-2012 ($r^2 = 0.86 - 0.99$, Fig. S5) (Sun et al., 2013b), the four OA factors with
259	fpeak = -1 was chosen in this study. The four OA components show very similar mass
260	spectral profiles ($r^2 = 0.86 - 0.99$) and diurnal variations (Fig. S4) to those observed-
261	during winter 2011-2012 (Sun et al., 2013b). The HOA spectrum resembles to that
262	identified by PMF analysis of high resolution OA mass spectra in Beijing in January
263	2013 (Zhang et al., 2014) which are both characterized pronounced m/z 91 and 115.
264	Although the CCOA spectrum doesn't present similar pronounced m/z 's (e.g., 77, 91,
265	105, and 115) as that resolved at a rural site in Central Eastern China (Hu et al., 2013),
266	it shows more similarity to that resolved in Beijing (Zhang et al., 2014). Also, CCOA
267	correlates better with chloride with an importance source from coal combustion
	12

268	(Zhang et al., 2012) than HOA ($r^2 = 0.41$ vs. 0.24), and also correlates well with m/z
269	60 ($r^2 = 0.77$, Fig. S6) a tracer m/z for biomass burning (Cubison et al., 2011). Note
270	that better correlations between HOA+CCOA and BC ($r^2 = 0.88$), NO _x ($r^2 = 0.77$),
271	and CO ($r^2 = 0.63$) than HOA ($r^2 = 0.36 - 0.47$) were observed in this study, which
272	might suggest that coal combustion emissions are also important sources of CO, BC
273	and NO _x during wintertime (Tian et al., 2008;Zhi et al., 2008).(Cubison et al., 2011).
274	Although COA didn't have external tracers to validate, it is very distinct as suggested
275	by its unique diurnal patterns (two peaks corresponding to meal time) and high m/z
276	55/57 ratio. Similar to our previous study (Sun et al., 2013b), the OOA shows a tight
277	correlation with NO ₃ ($r^2 = 0.90$) and also a good correlation with SO ₄ ⁻²⁻ +NO ₃ ⁻ ($r^2 =$
278	0.87). The mass spectral profiles and time series of four OA factors are shown in Fig.
270	86
279	
279	No biomass burning OA (BBOA) was resolved in this study. One of the reasons is
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3 Results and discussion

3.1 Identification and quantification of fireworks events

293	Burning of fireworks has been found to emit a large amount of K^+ , which can be
294	used to identify the FW events (Drewnick et al., 2006; Wang et al., 2007). As shown
295	in Fig. 1 and Fig. $\frac{892}{100}$, three FW events with significantly elevated K ⁺ were observed
296	on the days of Lunar New Year (LNY, 9-10 February), Lunar Fifth Day (LFD, 14
297	February), and Lantern Festival (LF, 24 February), respectively. All three FW events
298	started approximately at 18:00 and ended at midnight except LNY with a continuous
299	FW impact until 4:00 on the second day (Fig. 2). Fig. 1 shows that the relative
300	humidity was generally below 30% during LNY and LFD. While the wind speed at
301	the ground surface remained consistently below 2 m s ⁻¹ , it was increased to ~ 4 m s ⁻¹
302	at the height of 100 m. Also note that there was a wind direction change in the middle
303	of the two events. The meteorological conditions during LF were stagnant with wind
304	speed generally below 2 m s ⁻¹ across different heights. The relative humidity was \sim 50%
305	and the temperature averaged 3.5°C.
305 306	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background
305 306 307	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is -
305 306 307 308	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference
305 306 307 308 309	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then assumed as the contribution from
 305 306 307 308 309 310 	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It
 305 306 307 308 309 310 311 	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It should be noted that this approach might significantly overestimate the FW
 305 306 307 308 309 310 311 312 	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It should be noted that this approach might significantly overestimate the FW contributions of primary species (e.g., HOA, COA, CCOA, and BC) that were largely
 305 306 307 308 309 310 311 312 313 	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It should be noted that this approach might significantly overestimate the FW contributions of primary species (e.g., HOA, COA, CCOA, and BC) that were largely enhanced during the typical FW periods (18:00 – 24:00) due to the increased local
 305 306 307 308 309 310 311 312 313 314 	and the temperature averaged 3.5°C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then-assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It should be noted that this approach might significantly overestimate the FW contributions of primary species (e.g., HOA, COA, CCOA, and BC) that were largely enhanced during the typical FW periods (18:00 – 24:00) due to the increased local emissions (see Fig. S11-for diurnal variations of aerosol species). However, it should
 305 306 307 308 309 310 311 312 313 314 315 	and the temperature averaged 3.5° C. To estimate the contributions of fireworks, we first assume that the background concentration of each species has a linear variation during FW period. A linear fit is- was then performed on the 6 h data before and after FW events. The difference between the measured and the fitted value is then-assumed as the contribution from FW. The typical examples for estimating FW contributions are shown in Fig. S10. It should be noted that this approach might significantly overestimate the FW contributions of primary species (e.g., HOA, COA, CCOA, and BC) that were largely enhanced during the typical FW periods ($18:00 - 24:00$) due to the increased local emissions (see Fig. S11-for diurnal variations of aerosol species). However, it should have a minor impact on secondary species (e.g., SO4 ²⁻ , NO3-, and OOA) because of

316	their relatively stable variations between 18:00-24:00. As shown in Fig+2, all aerosol
317	species showed substantial increases from 15:00 to 21:00 on the day of LNY which
318	coincidently corresponded to a gradual change of wind direction. Therefore, regional
319	transport might have played dominant roles for the evolution of chemical species
320	during this period. For these reasons, only the FW contributions between 23:30, 9
321	February and 3:30, 10 February when the meteorological conditions were stable were
322	estimated. The FW contributions during LFD might also be overestimated due to the
323	influences of regional transport as suggested by the wind direction change in the
324	middle. Considering above, the contributions estimated in this work would represent
325	the upper limits of FW.
326	3.2 Mass concentration and chemical composition of FW aerosols
327	Figure 1 shows the time series of mass concentrations of PM_1 , $PM_{2.5}$, and
328	submicron aerosol species from 1 February to 1 March 2013. Because ACSM cannot
329	measure the metals (e.g., Sr, Ba, Mg, etc.) that were significantly enhanced during
330	FW periods (Wang et al., 2007; Vecchi et al., 2008) , the PM_1 in this study refer to
331	
332 I	$NR-PM_1 (= Org + SO_4^{2} + NO_3^{3} + NH_4^{4} + Chl + K^{4} + KCl) + BC.$ The $PM_{2.5}$ showed
	NR-PM ₁ (= $Org + SO_4^{} + NO_3^{} + NH_4^{-+} + Chl + K^{-+} + KCl) + BC$. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30
333	NR-PM ₁ (= Org + SO ₄ ^{$-$} + NO ₃ ^{$-$} + NH ₄ ^{$+$} + Chl+ K ^{$+$} + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of
333 334	NR-PM ₁ (= Org + SO ₄ ^{z} + NO ₃ ^{z} + NH ₄ ^{z} + Chl+ K ^{z} + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM _{2.5} during LNY (775 μ g m ⁻³) is more than 10 times higher than the China National
333 334 335	NR-PM ₁ (= Org + SO ₄ ^{z} + NO ₃ ^{z} + NH ₄ ^{z} + Chl+ K ^{z} + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM _{2.5} during LNY (775 μ g m ⁻³) is more than 10 times higher than the China National Ambient Air Quality Standard (75 μ g m ⁻³ , 24 h average). The average FW-PM _{2.5} mass
333 334 335 336	NR-PM ₁ (= Org + SO ₄ + NO ₃ + NH ₄ + Chl+ K ⁺⁻ + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM _{2.5} during LNY (775 μ g m ⁻³) is more than 10 times higher than the China National Ambient Air Quality Standard (75 μ g m ⁻³ , 24 h average). The average FW-PM _{2.5} mass concentrations during three FW events all exceeded 100 μ g m ⁻³ . These results suggest
333 334 335 336 337	NR-PM ₁ (= Org + SO ₄ + NO ₃ + NH ₄ + Chl+ K ⁺⁻ + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM _{2.5} during LNY (775 μ g m ⁻³) is more than 10 times higher than the China National Ambient Air Quality Standard (75 μ g m ⁻³ , 24 h average). The average FW-PM _{2.5} mass concentrations during three FW events all exceeded 100 μ g m ⁻³ . These results suggest that fireworks have large impacts on fine particle pollution, yet generally less than
 333 334 335 336 337 338 	NR-PM ₁ (= Org + SO ₄ ^{u} + NO ₃ ^{u} + NH ₄ ^{u} + Chl+ K ^{u} + KCl) + BC. The PM _{2.5} showed three prominent FW peaks with the maximum concentration occurring at ~00:30 during LNY and ~21:30 during LFD and LF, respectively. The peak concentration of PM _{2.5} during LNY (775 μ g m ⁻³) is more than 10 times higher than the China National Ambient Air Quality Standard (75 μ g m ⁻³ , 24 h average). The average FW-PM _{2.5} mass concentrations during three FW events all exceeded 100 μ g m ⁻³ . These results suggest that fireworks have large impacts on fine particle pollution, yet generally less than half day (approximately 10 h for LNY, and 6 h for LFD and LF). The PM ₁ also

340	correlation of PM_1 versus $PM_{2.5}$ shows much lower $PM_1/PM_{2.5}$ (0.08 – 0.19) ratios
341	during three FW events than that observed during NFW periods (0.90) (Fig. 23). One
342	of the reasons is likely due to the mineral dust component and metals from fireworks
343	that ACSM did not measure. However, the ACSM un-detected metals (e.g., Mg, Sr,
344	and Ba) that were are largely enhanced during FW periods generally contribute a
345	small fraction of PM (<2%) (Wang et al., 2007; Vecchi et al., 2008; Kong et al., 2014).
346	Therefore, our results might suggest that a large fraction of aerosol particles from the
347	burning of fireworks was emitted burning of fireworks has the most impact on aerosol-
348	particles in the size range of $1 - 2.5 \mu$ m. Consistently, Vecchi et al. (2008) found the
349	best correlation between the fireworks tracer, Sr, and the particles between 700-800
350	nm (mobility diameter, $D_{\rm m}$) which is approximately equivalent to $1.9 - 2.2 \ \mu {\rm m}$ in $D_{\rm va}$
351	(vacuum aerodynamic diameter, D_{va}) with a density of 2.7 g cm ⁻³ (Zhang et al., 2010).
252	Discuss 2.4 shares the second schemical second sitisms (DDM and OA from
352	Figure $\frac{3}{4}$ shows the average chemical composition of PM ₁ and OA from
352 353	Figure $\frac{3}{3}$ shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The
352 353 354	Figure $\frac{3}{4}$ shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean
352 353 354 355	Figure 3-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> \sim 50%) (Sun et al., 2012;Sun et al., 2013b),
352 353 354 355 356	Figure 3-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> \sim 50%) (Sun et al., 2012;Sun et al., 2013b), whereas that during LF was dominated by SIA (52%). As a comparison, organics
352 353 354 355 356 357	Figure 5-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> ~50%) (Sun et al., 2012;Sun et al., 2013b), whereas that during LF was dominated by SIA (52%). As a comparison, organics constituted the major fraction of FW-PM ₁ , contributing 44 – 55% on average. During
352 353 354 355 356 357 358	Figure 5-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> \sim 50%) (Sun et al., 2012;Sun et al., 2013b), whereas that during LF was dominated by SIA (52%). As a comparison, organics constituted the major fraction of FW-PM ₁ , contributing 44 – 55% on average. During LNY, FW exerted large impacts on potassium and chloride whose contributions were
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352 353 354 355 356 357 358 359 360	Figure 3-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> ~50%) (Sun et al., 2012;Sun et al., 2013b), whereas that during LF was dominated by SIA (52%). As a comparison, organics constituted the major fraction of FW-PM ₁ , contributing 44 – 55% on average. During LNY, FW exerted large impacts on potassium and chloride whose contributions were elevated to 21% and 15% of PM ₁ , respectively, from less than 7% (Chl) in the background aerosols. The large increases of potassium and chloride were also
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352 353 354 355 356 357 358 359 360 361 362	Figure 3-4 shows the average chemical composition of PM ₁ and OA from fireworks and also the background composition during LNY, LFD and LF. The background PM ₁ during LNY and LFD showed typical characteristics of clean periods with high fraction of organics (> ~50%) (Sun et al., 2012;Sun et al., 2013b), whereas that during LF was dominated by SIA (52%). As a comparison, organics constituted the major fraction of FW-PM ₁ , contributing 44 – 55% on average. During LNY, FW exerted large impacts on potassium and chloride whose contributions were elevated to 21% and 15% of PM ₁ , respectively, from less than 7% (Chl) in the background aerosols. The large increases of potassium and chloride were also observed during LFD and LF, and previous studies in Beijing (Wang et al., 2007;Cheng et al., 2014). As shown in Fig. 34, FW also emitted a considerable

364	SO_2 during all three FW events (r ² = 0.49 -0.92). Given that the relative humidity was	
365	low, $< 30\%$ during LNY and LFD, and $\sim 50\%$ during LF, aqueous-phase oxidation of	
366	SO ₂ depending on liquid water content could not play significant roles for the sulfate	
367	formation (Sun et al., 2013a). Therefore, sulfate in $FW-PM_1$ was mainly from the	
368	direction emissions of FW. Compared to sulfate, FW appeared to show minor impacts	
369	on nitrate, for example, 4% and 2% during LNY and LF, respectively. Although	
370	nitrate contributed 12% of FW-PM ₁ during LFD, most of it was likely from local-	
371	sources and/or-regional transport as supported by synchronous increases of all aerosol	
372	species the large contributions of local HOA and COA in OA (Fig. 3b) and	
373	alsoassociated with a wind direction change in the middle (Fig. 2).	
374	The OOA contributed dominantly to OA during LNY, which is 79% on average	
375	(Fig. 3a4 a). As shown in Fig. 45, the mass spectrum of FW-organics is highly similar	
376	to that of low-volatility OOA (LV-OOA, $r^2 = 0.94$; $r^2 = 0.89$ by excluding m/z 18 and	
377	m/z 44) (Ng et al., 2011a) indicating that the FW-organics is likely emitted in	Forma
378	secondary. Consistently, Drewnick et al. (2006) also found large enhancements of the	
379	OOA-related m/z 's (e.g., m/z 44) during New Year's fireworks, but the HOA-related	
380	m/z's (e.g., m/z 57) are not significant contributors to FW organics. OOA contributed-	
381	accounted for a much smaller fraction of OA during LF (28%) due to the large	
382	contributions of POA components (72%). Although the OOA contributions varied	
383	during three FW events, their absolute concentrations were relatively close ranging	
384	from 5.8 to 7.9 μ g m ⁻³ . It should be noted that our approach might overestimate the	
385	POA components in FW-OA were likely from because of the influences of NFW	
386	sources that were overestimated by the approach in this work, in particular during the	
387	FW period of LF when the local HOA, COA, and CCOA happened to have large	

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increases. By excluding the POA components in FW-OA, FW on average contributed $15 - 19 \ \mu g \ m^{-3} PM_1$ during three FW events.

390 3.3 Secondary aerosol and PM pollution

391 The PM_1 (NR- $PM_1 + BC$) varied largely across the entire study with daily average mass concentration ranging from 9.1 to 169 μ g m⁻³. The average PM₁ mass 392 concentration was 80 (\pm 68) µg m⁻³, which is approximately 20% higher than that 393 394 observed during winter 2011-2012 (Sun et al., 2013b). Organics composed the major fraction of PM₁, accounting for 43%, followed by nitrate (22%), sulfate (14%), 395 396 ammonium (13%), BC (5%) and chloride (3%). The OA composition was dominated by OOA (53%) with the rest being POA. Compared to winter 2011-2012 (Sun et al., 397 398 2013b), this study showed significantly enhanced OOA (53% vs. 31%) and secondary 399 nitrate (22% vs. 16%), indicating that secondary formation have played important 400 roles in the formation of pollution episodes. 401 Figure 1d shows that submicron aerosol species alternated routinely between pollution events (PEs) and clean periods (CPs) throughout the entire study. The PEs 402 generally lasted $\sim 1 - 2$ days except the one on 23 - 28 February that lasted more than 403 5 days, whereas the CPs were shorter, generally less than 1 day. In total, 9 PEs and 9 404 405 CPs were elassified-identified in this study (Fig. 1). A statistics of the mass concentrations and mass fractions of aerosol species during the 9 PEs is presented in 406 Fig. 56. The average PM₁ mass concentration ranged $68 - 179 \mu \text{g m}^{-3}$ during PEs with 407 the total secondary particulate matter (SPM = OOA + SO_4^2 + NO_3^2 + NH_4^+) 408 accounting for 63 - 82%. The average mass concentration of SPM for the 9 PEs was 409 86 (\pm 32) µg m⁻³, which is nearly 3 times that of primary PM (PPM = HOA + COA 410 +CCOA + BC + Chl) $(30 \pm 9.5 \ \mu g \ m^{-3})$. SPM consistently dominated PM₁ across 411

413	during daytime (Fig. 6b7b). The diurnal cycle of SPM presented a gradual increase
414	from 50 to 70 μ g m ⁻³ between 10:00 – 20:00, indicating evident photochemical
415	production of secondary species during daytime. It should be also noted that all
416	secondary species showed ubiquitously higher mass concentrations than those of
417	primary species (Fig. 5a 6a).
418	The SOA generally contributed more than 50% to OA with an average of 55%
419	during the PEs except the episode on 3 February (35%). It's interesting to note that
420	the contribution of POA increased as a function of organic loadings which varied
421	from ~35% to 63% when organics was above 80 μ g m ⁻³ (Fig. 6e 7c). Such behavior is
422	mainly caused by the enhanced CCOA at high organic mass loadings, which was also
423	observed during winter 2011 – 2012 (Sun et al., 2013b). These results suggest that
424	POA played more important roles than SOA in PM pollution during periods with high
425	organic mass loadings (e.g., $> 60 \ \mu g \ m^{-3}$). In fact, POA showed even higher mass
426	concentrations than OOA at nighttime $(0:00 - 8:00)$ due to the intensified local
427	emissions, e.g., coal combustion for heating. Despite this, the role of POA in PM
428	pollution was compensated by the elevated secondary inorganic species as a function
429	of PM loadings (Fig. 6a7a) leading to the consistently dominant SPM across different
430	pollution levels. Figure 7a-8a shows an evidently lower contribution of organics to
431	PM1 during PEs than CPs. The elevated secondary inorganic species during PEs were
432	closely related to the increase of RH (Fig. 1). For example, during the pollution
433	episode on 3 February, the sulfate concentration increased rapidly and became the
434	major inorganic species when RH was increased from $\sim 60\%$ to $> 90\%$. The gaseous
435	SO ₂ showed a corresponding decrease indicating aqueous-phase processing of SO ₂ to

different PM levels (69 - 75%), but generally with higher contributions (up to 81%)

436	form sulfate, consistent with our previous conclusion that aqueous-phase processing
437	could contribute more than 50% of sulfate production during winter 2011-2012 (Sur
438	et al., 2013a).

The compositional differences between PEs and CPs also led to different mass 439 extinction efficiency (MEE, 630 nm) of PM₁ (Fig. 7b8b). The higher MEE (6.4 m² g⁻¹) 440 during PEs than CPs (4.4 m² g⁻¹) is primarily due to the enhanced secondary species, 441 442 and also likely the increases of aerosol particle sizes although we don't have size data 443 to support it. Similar increases of mass scattering efficiency from clean periods to relatively polluted conditions were also observed previously in Beijing and Shanghai 444 445 (Jung et al., 2009;Huang et al., 2013). It should be noted that the MEE of PM₁ in this study refers to PM2.5 bext/PM1. Considering that PM1 on average contributes ~60-70% 446 of PM_{2.5}If assuming PM₁ on average contributed 70% of PM_{2.5} in Beijing (Sun et al., 447 2012; Sun et al., 2013b), the real MEE of PM₁ during PEs and CPs would be \sim 3.8 - 4.5 448 and ~ 2.6 - 3.1 m² g⁻¹, respectively. 449

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450 3.4 Holiday Effects on PM Pollution

Figure 8-9 shows a comparison of aerosol species, gaseous species, and 451 452 meteorological parameters between holiday (HD) and non-holiday (NHD) periods. 453 The official holiday for the Spring Festival was 9 - 15 February. However, we noted a large decrease of cooking aerosols from 7 February until 19 February (Fig. S46eb), 454 455 whose emissions were expected to be stable under similar meteorological conditions. The decrease of COA was likely due to the reduction of the number of population in 456 457 Beijing, which agreed with the fact that most migrants from outside Beijing were leaving for hometown before the official holiday. Therefore, 7-19 February was 458 459 used as a longer holiday for a comparison. It was estimated that approximately half of

460 population (9 million) left Beijing before the Spring Festival

461

462 reduction in human activities would exert a large impact on aerosol composition and 463 sources in the city during holidays. To better investigate the HD effects on PM pollution, the data shown in Fig. 8-9 excluded the CPs marked in Fig. 1. The data with 464 the CPs included are presented in Fig. S12. 465 466 The differences between HD and NHD for primary species varied largely among 467 different species. COA showed the largest reduction (69%) among aerosol species with the average concentration decreasing from 5.8 μ g m⁻³ during NHD to 1.8 μ g m⁻³ 468 469 during HD. The contribution of COA to OA showed a corresponding decrease from 470 12% to 4%. Given the similar meteorological conditions between HD and NHD, e.g., RH (46% vs. 52%) and wind speed (1.3 m s⁻¹ vs. 1.2 m s⁻¹), the reduction of COA 471 472 clearly indicated a large decrease of population and the number of restaurants opened during HD. The CCOA showed approximately 30% reduction during HD, and its 473 474 contribution to OA decreased from 23% to 18%. Not surprisingly, chloride showed a similar reduction as CCOA because it was primarily from coal combustion emissions 475 476 during wintertime (Sun et al., 2013b). Figure 8-9 also shows a significant reduction (54%) for NO₃ indicating much less traffic emissions in the city during HD. The HOA, 477 478 however, even showed a slight increase during HD, which appeared to contradict with 479 the reduction of two combustion-related tracers, BC and CO ($\sim 20\%$). The results This 480 can be explained by the fact that coal combustion is a large source of BC and CO 481 during heating season (Tian et al., 2008; Zhi et al., 2008). Consistently, BC and CO showed relatively similar reductions to CCOA. Therefore, the minor variations of 482 HOA might indicate that the number of heavy-duty vehicles and diesel trucks that 483

(http://news.xinhuanet.com/yzyd/local/20130208/c 114658765.htm). Such a great
484	dominated HOA emissions (Massoli et al., 2012;Hayes et al., 2013) remained little
485	change during HD period although that of gasoline vehicles was largely decreased. It
486	should be noted that HOA showed a large peak on 9 February – the first day of the
487	official holiday (Fig. S46b) when more traffic emissions were expected due to many
488	people leaving for hometown. After that, HOA showed comparablyslightly lower
489	concentration during $11 - 17$ February than other periods indicating. In fact, the
490	average HOA showed a slight reduction (~5%) during the long holiday effects period
491	(7 – 19 February) suggesting a small holiday effect on HOA reduction. Together, the
492	total primary aerosol species (PPM) showed an average reduction of 22% because of
493	holiday effects.

494 Nitrate showed the largest reduction among secondary species by 22% during HD, primarily due to a reduction of its precursors NO and NO₂. The Rresults here suggest 495 496 that reducing traffic emissions would help mitigate the nitrate pollution in the city. Compared to nitrate, sulfate showed minor changes (2%) between HD and NHD, and 497 498 OOA even showed a slight increase (6%) during HD. One of the reasons is that 499 secondary sulfate and OOA were mainly formed over regional scale and less affected 500 by local production, consistent with their relatively flat diurnal cycles (Fig. S11). 501 Ammonium showed a reduction between nitrate and sulfate because ammonium mainly existed in the form of (NH₄)₂SO₄ and NH₄NO₃. Overall, secondary species 502 showed generally lower reductions than primary species with the total secondary 503 504 species (SPM) showing an average reduction of 9% during HD. The joint reductions 505 of PPM and SPM led to an average reduction of 13% for PM1 during HD. However, 506 these reductions did not help alleviate the fine particle pollution during HD. The PM_{2.5} excluding FW impacts even showed 27% increase from 96 µg m⁻³ during NHD 507

508	to 122 $\mu gm^{\text{-3}}$ during HD. One possible reason is likely due to the increases of aerosol
509	species in the size range of $1 - 2.5 \mu m$ during HD period. The longer holiday (LHD, 7
510	- 19 February) showed similar influences on both primary and secondary species as
511	the official holiday (9 – 15 February). COA, CCOA, and NO are the three species
512	with the largest reductions during LHD (> 50%). However, HOA, $SO_4^{\frac{2}{2}}$, OOA, and
513	$PM_{2.5}$ showed rather small changes (< ±7%). Therefore, results in this study suggest
514	that controlling the primary source emissions, e.g., cooking and traffic emissions in
515	the city can reduce the primary particles largely, yet has limited effects on secondary
516	species and the total fine particle mass. One of the reasons is that because the severe
517	PM pollution in Beijing is predominantly contributed by secondary species (see
518	discussions above in section 3.3) that are formed over regional scales. Reducing the
519	primary source emissions in local areas would have limited impacts on mitigation of
520	air pollution in the city. Similarly, Guo et al. (2013) reported a large reduction of
521	primary organic carbon (OC) from traffic emissions and coal combustion during the
522	2008 Olympic Summer Games when traffic restrictions and temporary closure of
523	factories were implemented. However, secondary OC was not statistically different
524	between controlled and non-controlled periods. Our results highlight the importance
525	of implementing joint efforts over regional scales for air pollution control in north
526	China.

527 **4** Conclusions

528 We have characterized the aerosol particle composition and sources during the 529 Chinese Spring Festival,— in 2013. The average PM₁ mass concentration was 80 (\pm 68) 530 µg m⁻³ for the entire study with organics being the major fraction (43%). Nine 531 pollution events and nine clean periods with substantial compositional differences

532	were observed. The secondary particulate matter (= SOA+ sulfate + nitrate +
533	ammonium) played a dominant role for the PM pollution during the-nine PEs. The
534	contributions of SPM to PM_1 varied from 63% to 82% with SOA on average
535	accounting for \sim 55% of OA. As a result, the average mass extinction efficiency of
536	PM_1 during PEs (6.4 m ² g ⁻¹) was higher than that during CPs (4.4 m ² g ⁻¹). Three FW
537	events, i.e., LNY, LFD, and LF, were identified, which showed significant and
538	short-term impacts on fine particles, and non-refractory potassium, chloride, and
539	sulfate in PM ₁ . The FW also exerted a large impact on organics that presented mainly
540	in secondary as indicated by its similar mass spectrum to that of oxygenated OA. The
541	holiday effects on aerosol composition and sources were also investigated by
542	comparing the differences between holiday and non-holiday periods. The changes of
543	anthropogenic source emissions during the holiday showed large impacts on reduction
544	of cooking OA (69%), nitrogen monoxide (54%), and coal combustion OA (28%) in
545	the city, yet presented minor-much smaller influences on secondary species. The
546	average SOA and the total PM _{2.5} even increased slightly during the holiday period.
547	Results here have significant implications that controlling the local primary source
548	emissions, e.g., cooking and traffic activities, might have limited effects on improving
549	air quality during polluted days when SPM from regional transport dominated aerosol
550	composition for most of time. Our results also highlight the importance of
551	implementing joint measures over regional scales for mitigation of air pollution in
552	megacity Beijing.

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802 Figure Captions:

803	Fig. 1. Time series of meteorological parameters (a) relative humidity (RH) and	
804	temperature (T); (b) wind direction (WD) and wind speed (WS) at the height of 8m	
805	and 100 m; mass concentrations of (c) PM _{2.5} and NR-PM ₁ + BC and (d) submicron	
806	aerosol species. Also shown in (a) and (b) is the temperature and wind speed at the	
807	height of 8 m which are in blue. The extinction coefficient (b_{ext}) at 630 nm is shown	
808	in (c). Three events, i.e., Lunar New Year (LNY), Lunar Fifth Day (LFD) and Lantern	
809	Festival (LF) with significant influences of fireworks are marked in (c). In addition,	
810	the classified clean periods (CPs) and polluted events (PEs) are marked as shaded	
811	graylight blue and pink areas, respectively A more detailed time series of aerosol-	
812	species during the three fireworks events are shown in Fig. S9.	
813	Fig. 2. Time series of PM species (Org, SO4, NO3, NH4, Chl, K KCl, and BC) and	
814	meteorological variables (wind direction (100 m) and wind speed (8 m)) during three	
815	firework events, i.e., (a) Lunar New Year, (b) Lunar Fifth Day, and (c) Lantern	
816	Festival. The two blue arrow lines represent the starting and ending times of fireworks	
817	events.	
818	Fig. 23. Correlation of PM_1 vs. $PM_{2.5}$ with the data segregated into three fireworks	
819	events (LNY, LFD, and LF) and non-fireworks periods (NFW). The blank circles	
820	represent FW data between 18:00 – 23:30 on 9 February which had large influences	
821	from NFW sources.	
822	Fig. 34. Average chemical composition of PM_1 and OA from fireworks and	
823	background during three FW events.	
824	Fig. 45. (a) Average mass spectra (MS) of OA during the firework period of Lunar	
825	New Year (23:30, 9 February – 3:30, 10 February) and the period of background (BG,	
826	4:30 – 11:00, 10 February). (b) Comparison of the difference spectrum from (a), i.e.,	
827	$MS_{FW+BG} - MS_{BG}$, with the average LV-OOA spectrum in Ng et al.(2011a). Note that	
828	five m/z 's, 37 (³⁷ Cl ⁺), 58 (NaCl ⁺), 60 (Na ³⁷ Cl ⁺), 74 (KCl ⁺), and 76 (K ³⁷ Cl ⁺ / ⁴¹ KCl ⁺)	
829	marked in the figure were dominantly from fragmentation of inorganic salts during	
830	fireworks. Five m/z^2 , 37 (³⁷ Cl [±]), 58 (NaCl [±]), 60 (Na ³⁷ Cl [±]), 74 (KCl [±]), and 76 (K ³⁷ Cl [±] /	
831	⁴¹ KCl ⁺) with significant influences of fireworks are marked.	
832	Fig. 56. Box plots of (a) mass concentrations and (b) mass fractions of aerosol species	
833	for 9 pollution events marked in Fig. 1. The mean (cross), median (horizontal line),	
834	25 th and 75 th percentiles (lower and upper box), and 10 th and 90 th percentiles (lower	

and upper whiskers) are shown for each box.

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- Fig. 67. Left panel: variations of chemical composition of (a) organics, SNA (=sulfate + nitrate + ammonium), and others (the rest species in PM₁); (b) SPM and PPM; and
 (c) SOA and POA as a function of PM₁ and organics loadings, respectively. The
 middle and Rright panels show their corresponding the diurnal profiles of
 compositions and mass concentrations, respectively.
- 841 **Fig. 78.** (a) Average mass fraction of organics (f_{Org}) as a function of PM₁ mass, and (b)
- 842 correlations of extinction coefficients ($PM_{2.5}$) vs. PM_1 for 9 pollution events (PEs) and
- 843 9 clean periods (CPs) marked in Fig. 1. The error bar represents one standard
- 844 deviations of the average for each event.
- **Fig. 89.** The average ratios of aerosol species, gaseous species, PM mass
- 846 concentrations, extinction coefficient, and meteorological parameters between holiday
- 847 (HD) and non-holiday (NHD) periods. Two different holidays, i.e., the official
- holiday of 9 15 February and the longer holiday of 7 20 February were used for
- 849 averages. Also note that the averages were made by excluding clean periods and
- 850 firework events during both HD and NHD days. The error bars are the standard errors
- 851 of the ratios.





Fig. 1. Time series of meteorological parameters (a) relative humidity (RH) and temperature (T); (b) wind direction (WD) and wind speed (WS) at the height of 8m



firework events, i.e., (a) Lunar New Year, (b) Lunar Fifth Day, and (c) Lantern

Festival. The two blue arrow lines represent the starting and ending times of fireworks

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events.



Fig. 23. Correlation of PM₁ vs. PM_{2.5} with the data segregated into three fireworks

events (LNY, LFD, and LF) and non-fireworks periods (NFW). The blank circles

represent FW data between 18:00 – 23:30 on 9 February which had large influences

876 from NFW sources.



Fig. 34. Average chemical composition of PM₁ and OA from fireworks and

880 background during three FW events.





882	Fig. 45. (a) Average mass spectra (MS) of OA during the firework period of Lunar
883	New Year (23:30, 9 February – 3:30, 10 February) and the period of background (BG,
884	4:30 - 11:00, 10 February). (b) Comparison of the difference spectrum from (a), i.e.,
885	$MS_{FW+BG} - MS_{BG}$, with the average LV-OOA spectrum in Ng et al.(2011a). Note that
886	five m/z 's, 37 (³⁷ Cl ⁺), 58 (NaCl ⁺), 60 (Na ³⁷ Cl ⁺), 74 (KCl ⁺), and 76 (K ³⁷ Cl ⁺ / ⁴¹ KCl ⁺)
887	marked in the figure were dominantly from fragmentation of inorganic salts during
888	fireworks. Five <i>m/z</i>'s, 37 (³⁷Cl⁺), 58 (NaCl⁺), 60 (Na³⁷Cl⁺), 74 (KCl⁺), and 76 (K³⁷Cl⁺/
889	⁴¹ KCl ⁺) with significant influences of fireworks are marked.



Fig. 56. Box plots of (a) mass concentrations and (b) mass fractions of aerosol species

- for 9 pollution events marked in Fig. 1. The mean (cross), median (horizontal line),
- 893 25^{th} and 75^{th} percentiles (lower and upper box), and 10^{th} and 90^{th} percentiles (lower
- and upper whiskers) are shown for each box.



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Fig. 67. Left panel: variations of chemical composition of (a) organics, SNA (=sulfate

+ nitrate + ammonium), and others (the rest species in PM₁); (b) SPM and PPM; and

- 899 (c) SOA and POA as a function of PM_1 and organics loadings, respectively. The
- 900 middle and Rright panels show their corresponding the diurnal profiles of
- 901 compositions and mass concentrations, respectively.



Fig. 78. (a) Average mass fraction of organics (f_{Org}) as a function of PM₁ mass, and (b)

- correlations of extinction coefficients ($PM_{2.5}$) vs. PM_1 for 9 pollution events (PEs) and
- 905 9 clean periods (CPs) marked in Fig. 1. The error bar represents one standard
- 906 deviations of the average for each event.



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Fig. 89. The average ratios of aerosol species, gaseous species, PM mass
concentrations, extinction coefficient, and meteorological parameters between holiday
(HD) and non-holiday (NHD) periods. Two different holidays, i.e., the official
holiday of 9 – 15 February and the longer holiday of 7 – 20 February were used for
averages. Also note that the averages were made by excluding clean periods and
firework events during both HD and NHD days. The error bars are the standard errors
of the ratios.

1	Supporting Information
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3	Aerosol Composition and Sources during the Chinese Spring
4	Festival: Fireworks, Secondary Aerosol, and Holiday Effects
5	
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Fig. S1. (a) Time series of signals of m/z 39 and K⁺, (b) correlation of m/z 39 vs. m/z

43. The data in (b) are segregated into three FW events, i.e., Lunar New Year (LNY),

24 Lunar Fifth Day (LFD), and Lantern Festival (LF), and NFW periods. The data during

the FW period of 18:00 – 23:30, 9 February (LNY, BF) that have large influences of

26 NFW sources are also shown for a comparison. The K^+ signal in (a) was calculated as

27 $m/z 39 - m/z 43 \times (m/z 39/43)_{NFW}$, i.e., $m/z 39 - m/z 43 \times 0.45$.





m/z 74 in four OA factors, i.e., HOA, COA, CCOA, and OOA. (c) shows the









37 0.15 and ${}^{37}\text{Cl}^+ = 0.323 \times {}^{35}\text{Cl}^+$.

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Fig. S64. Mass fraction of four OA factors (from 6-factor solution; three OOA factors were combined into one OOA factor as discussed in the text) as a function of fpeak values. Overall, the contribution of each OA factor was relatively stable across different fpeak values (average $\pm 1\sigma$; min – max): HOA (14 \pm 1.6%; 12 – 16%); COA (14±2.8%; 11 – 17%); CCOA (19±2.7%; 15 – 22%); OOA (51±1.7; 49 – 55%).

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Fig. S85. Mass spectra correlations between this study and those identified in Beijing 46 in winter 2011-2012 (Sun et al., 2013). The mass spectra of OA factors at fpeak = -1 47 presented the best correlation with those identified in winter 2011-2012 (Sun et al., 2013). Therefore, four factor solution with fpeak = -1 was chosen in this study.



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53 Fig. S4S6. (a) Mass spectra, (b) diurnal profiles, and (eb) time series of the four OA components, i.e., HOA, COA, CCOA, and OOA. The comparisons of mass spectra of 54 55 four OA factors with those resolved during winter 2011-2012 (Sun et al., 2013) are







Fig. S558. Summary of key diagnostic plots of the PMF results for 6-factor solution: (a) Q/Q_{exp} as a function of number of factors, (b) Q/Q_{exp} as a function of FPEAK, (c) the box and whiskers plot showing the distributions of scaled residuals for each m/z, (d) variations of the residual (= measured – reconstructed), (e) Q/Q_{exp} for each point in time, (f) time series of 6 factors and (h) factor profiles of 6 factors. The three OOA factors, i.e., OOA-1, OOA-2, and OOA-3 were combined into one OOA factor that is shown in Figure S4.

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73 Fie 74 ₩ 75 stable across $\mathbf{V}\mathbf{0}$ 76 diff COA

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Fig. **S7S9.** Time series and mass spectra of four OA factors for three different fpeak 79 80 values (-1, 0, and 1). The time series of four OA factors for different fpeak values agree overall well. However, the mass spectra of OA factors have large differences. 81 Note that most mass spectra of OA factors at fpeak > 1 are largely different from the 82 83 standard mass spectra reported in Ng et al. (2011) and those resolved in Beijing in 84 winter 2011-2012 (Sun et al., 2013).



Fig. S8. Mass spectra correlations between this study and those identified in Beijing
 in winter 2011-2012 (Sun et al., 2013). The mass spectra of OA factors at fpeak = -1
 presented the best correlation with those identified in winter 2011-2012 (Sun et al.,
 2013). Therefore, four factor solution with fpeak = -1 was chosen in this study.



Fig. S10. Estimation of firework contributions (red shaded areas) for selected species $(SO_4^{2}, Chl, OOA, and SO_2)$ during LNY, LFD, and LF.







96 **Fig. S11.** Diurnal cycles of SO_4^{2-} , NO₃⁻, BC, Chl, OOA, CCOA, HOA, and COA for

97 the entire study. The shaded areas show the typical time intervals with fireworks98 impacts.

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