

We are thankful to the two reviewers for their thoughtful comments and suggestions that help improve the manuscript significantly. We have revised the manuscript accordingly. In particular, we significantly expanded the validation of PMF solution in the revised manuscript. The uncertainties for K quantification were also addressed in the text. Listed below are our point-by-point responses in blue to each reviewer's comments

Response to Reviewer #1

General comments:

The manuscript reports the chemical composition of non-refractory particulate matter with vacuum aerodynamic diameter less than 1 micron (NR-PM₁) measured by an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) in Beijing, China during the Chinese Spring Festival. The field study focuses on evaluating the potential impacts of fireworks and a short-term reduction of population in Beijing during the holiday on air quality as well as the significance of secondary aerosol (both inorganics and organics) formation. The manuscript provide important insight in term of air quality control strategies that reducing primary local emissions only have limited influences to improve the air quality in Beijing, thus highlighting the importance of joint efforts over regional scales for air pollution control in northern China. Overall, I recommend this manuscript to be published after addressing the specific comments below:

We thank the reviewer for his/her positive comments.

Specific comments:

1. Section 2.3: There are a few issues on the quantification of K and Cl ions using ACSM data:

1) Page 20623, line 20-21: The meaning of "PMF modelled m/z 74" is unclear. Figure S2 does not provide any explanation. Please provide details.

The "PMF modeled m/z 74" was clarified in the revised manuscript as: "The PMF modeled m/z 74 refers to the sum of m/z 74 in four OA factors, i.e., HOA, COA, CCOA, and OOA."

2)Page 20623, Line 19: Drewnick et al. (2006) did not calibrate their ToF-AMS using chemical standard and thus estimated the RIE of 2.9 for K⁺, which is based on the potassium to-sulfate ratio (2.76) determined by PIXE analysis on fireworks aerosol sample from Dutcher et al. (1999). The RIE of K⁺ could be instrument dependent. Can the authors perform a similar comparison and comment on the accuracy of using RIE of 2.9 for K⁺ quantification in this study?

We agree with the reviewer that quantification of K with AMS might have large uncertainties, and the RIE_K can be instrument dependent. Unfortunately, we were unable to estimate the RIE_K using the approach described in Drewnick et al.(2006) because we didn't have collocated K⁺ and sulfur (S) measurements in this study. As a result, RIE_K= 2.9 estimated from fireworks by Drewnick et al.(2006)

was used. The AMS quantified K^+ during LFD and LF contributed 4.5% and 4.7% of PM_{10} , respectively, which is very close to $\sim 5\%$ ($PM_{2.5}$) reported by Cheng et al. (2014). The results suggest that $RIE_K = 2.9$ is likely reasonable for our study. The large contribution of K during LNY was likely due to the intensified firework emissions (mainly firecrackers), especially between 0:00 – 3:00 am. Because we don't have

The quantification of K^+ might affect the bulk aerosol composition of fireworks, yet it has negligible effects on our discussions on secondary aerosols and the impacts of reduced emissions on aerosol chemistry during the holiday because the three short-term FW events were all excluded.

3) Page 20623, Line 25-26: Organic aerosol contributes to fragment C_3^+ at m/z 36 in ambient environment. Can the authors estimate the uncertainty due to the presence of fragment C_3^+ to the quantification of HCl^+ based on the non-fireworks period?

Based on our winter (2013-2014) measurements by an Aerodyne High-resolution Time-of-Flight Aerosol Mass Spectrometer at the same location, the C_3^+ contributed a negligible fraction to m/z 36 ($< 0.1\%$, Figure A1). Therefore, C_3^+ should have minor effects on our quantification of HCl^+ although the measurement was conducted in a different year.

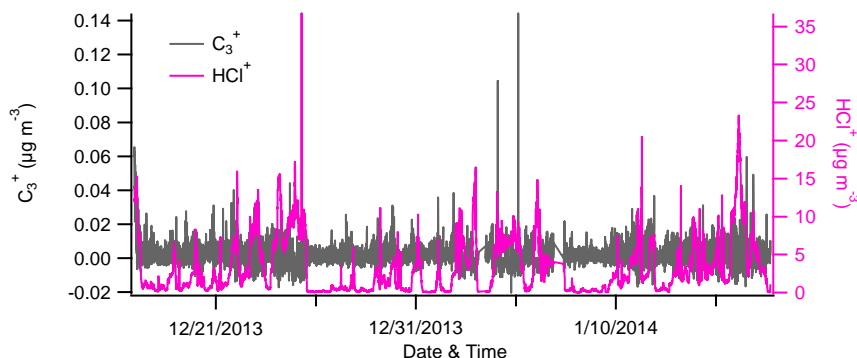


Figure A1. Time series of C_3^+ and HCl^+ at m/z 36. The data was from the W-mode measurement by the HR-ToF-AMS

2. Page 20625, line 2-4 and line 20-23: 1) Please confirm whether the meteorological conditions during LF period were stagnant. The wind direction was changing during LF period (It is hard to read in the print out of Figure 1, please see recommendation in technical comments).

Yes, the meteorological conditions during LF period were stagnant. As shown in Figure S9c, the wind speed remained consistently low ($\sim 1 \text{ m s}^{-1}$) and the wind direction was also consistently from the east.

2) The authors suggest that their estimation represents the upper limits of fireworks aerosol contributed to the total PM due to several reasons. However, it is not clear whether the pollutants in particular PM were carried to or removed from the sampling location due to wind direction changes. Please clarify.

The wind directions during Lunar New Year (LNY) and Lantern Festival (LF) were relatively stable. Thus, minor impacts of transport on our estimation are expected during these two holidays. Different from LNY and LF, there was an evident change of wind direction during Lunar Fifth Day (from north to south and then from south to north). All aerosol species (Figure S1b) including nitrate with minor impacts from fireworks showed synchronous increases between 18:00-21:00, and then decreases between 21:00 - 24:00. We would argue that the transport from outside of the location might have played an important role. However, it is difficult to quantify the contributions of transport based on the measurements in this study.

3. Page 20267, line 12-14: Even though LV-OOA component has been considered as aged secondary organic aerosol (SOA) in ambient, it is likely not straight forward to comment on the nature (primary vs. secondary) of OA generated in FW period based on the mass spectral features alone. First, the FW-OOA mass spectra may depend on the manufacturing materials (such as organic component if any) of firework. Furthermore, the freshly formed SOA (i.e. SV-OOA) are generally less oxygenated (i.e. lower O:C) and has a lower organic signal at m/z 44 than LV-OOA. However, it would be interesting to understand if it is possible to create a highly oxidizing ambient environment during fireworks for LV-OOA production.

We agree with the reviewer that the freshly formed SOA is generally less oxygenated. The mass spectral similarity of FW-OA to LV-OOA might be due to the burning of oxygenated organic materials in fireworks though we don't have further evidence to support it. We thank the reviewer for pointing out such an interesting question that should be addressed in future studies.

4. CAPS measurement: Was there any RH control before the CAPS inlet? Secondary inorganic species are hygroscopic in nature, thus resulting in larger size and higher extinction efficiency under high RH. Please clarify.

A silica gel dryer was put in front of the CAPS monitor during this study. The relative humidity after the dryer is generally below 40%. Therefore, the extinction we measured in this study refers to dry particles. This information is now added in the text.

5. HOA and BC in holiday period: 1) Page 20630, line 24-29: In general, heavy-duty vehicles/diesel trucks emit more black carbon (BC) compared to gasoline vehicle. Reduction of BC and slight increasing of HOA during holiday period suggests that significant fraction of BC was emitted from sources (i.e. coal combustion) other than diesel vehicles in Beijing.

We thank the reviewer's suggestion. Indeed, coal combustion is a large source of BC and CO during heating season (Tian et al., 2008;Zhi et al., 2008). This is also consistent with the relatively similar reductions of BC and CO to CCOA. We added this information into the discussions.

2) Page 20631, line 3-4: The authors argue that HOA showed comparably lower concentration during holiday, but it is contradict to the previous discussion (Page 20630, line 24-25) and Figure 8 (Ratio_{HD/NHD} of HOA_1). Please justify.

Thank the reviewer's carefulness. The HOA showed slightly lower concentration during 11 – 17 February than other periods leading to a slight reduction (~5%) during the long holiday period (7 – 19 February, Figure 8). The results suggest that there was a small holiday effect on HOA reduction, but not as significant as other primary species. If the official holiday (9 – 15 February) was used for the comparison, the HOA showed a slight increase instead, mainly due to the large increase of HOA on 9 February – the first day of the official holiday. For the clarification, we revised the statement and now it reads "HOA showed slightly lower concentration during 11 – 17 February than other periods. In fact, the average HOA showed a slight reduction (~5%) during the long holiday period (7 – 19 February) suggesting a small holiday effect on HOA reduction."

Technical comments:

1. Page 20623, line 17: Please change the equation to "m/z 39 – (m/z 43 x 0.45)"

Changed

2. Figure 1 and Figure S9: 1) K and KCl signals are tracers for the fireworks aerosol particles as shown in Figure S9. Since the focus of Section 3.1 and 3.2 is to investigate the short-term impacts of fireworks on air quality, it is recommended to include time series of K and KCl in Figure 1 and refer the zoom in to Figure S9 in order to clearly illustrate the impacts of fireworks on different aerosol species.

We thank the reviewer's suggestions. The time series of K was included in Figure 1. However it is difficult to read the time series of KCl because its concentration was too low. Given that KCl correlates tightly with K ($r^2 = 0.82$), it was not included in Figure 1. Following the reviewer's suggestions, we added "A more detailed time series of aerosol species during the three fireworks events are shown in Fig. S9." in the figure caption of Figure 1.

2) Please also include wind direction plots in Figure S9 as it is often discussed in the text but it is hard to compare different time series in Figure 1.

The wind direction and wind speed were included in Figure S9 in the revised manuscript.

3) The gray color for the clean periods is not clearly showed in Figure 1. Please change the color.

The color for the clean periods was changed to light blue for easy reading.

3. Page 20267, line 21: Change "overestimate" to "underestimate"?

We revised this sentence to avoid confusion. Now it reads “It should be noted that our approach might overestimate the POA components in FW-OA because of the influences of NFW sources, in particular during the FW period of LF when the local HOA, COA, and CCOA happened to have large increases.”

Response to Reviewer #4

The manuscript describes the chemical composition and sources of PM_{10} in Beijing during February 2013, with a focus on the impact of fireworks on PM pollution in the Festival days. The authors measured the aerosol composition with an ACSM and used PMF for source apportionment. Characterization of chemical composition and sources of Chinese particulate matter are certainly of great interest. However, this manuscript is more like a routine study/report. There are quite some weak points (see below), especially the quantification of potassium with ACSM is not convincingly presented and the stability, uncertainty and validation of their PMF solution are lack and/or need substantial improvement. I therefore do not recommend publication in the high profile journal ACP if these issues are not well resolved.

We thank the reviewer's comments. In the revised manuscript, we significantly expanded the validation of PMF solution and also addressed the uncertainties for the quantification of K.

We disagree with the reviewer that this manuscript is a routine report. Our study not only characterized the short-term fireworks impacts on PM pollution, but also investigated the role of secondary aerosols in PM pollution and the impacts of reduced anthropogenic emissions on aerosol chemistry. In particular, we found that reducing primary local emissions have limited influences to improve the air quality in Beijing, thus highlighting the importance of joint efforts over regional scales for air pollution control in northern China.

Major comments:

1. Quantification of potassium (K^+) with AMS (ACSM) is very difficult. Potassium RIEs are very sensitive to instrument tuning and can change by orders of magnitude (personal experience: range 5-50). Slowik et al. (2010) for example reported $RIE_K = 10$ and stated that quantification is very difficult. The reason for that is the tendency of potassium for surface ionization. Depending on the tuning of the spectrometer and exact temperature of the vaporizer the ratio between potassium ions from surface ionization (SI) and electron impact (EI) ionization can vary a lot. Large differences between different instruments (especially different types like ToF-AMS and Q-ACSM) are expected. Even within a measurement campaign RIE_K could change significantly due to changes at the vaporizer surface. The stability of SI/EI during a campaign can be checked by correlating K^+ to Na^+ that is affected similarly.

We agree with the reviewer that quantification of K with AMS is challenging because of surface ionization issue. Slowik et al. (2010) reported a $RIE_K = 10$ based on the calibration of pure KNO_3 particles, while Drewnick et al. (2006) obtained $RIE_K = 2.9$ based on the comparisons of K/S from fireworks and AMS measurements. Unfortunately we didn't have collocated K measurement, $RIE_K = 2.9$ that was estimated from fireworks was therefore used in this study. The quantified K during LFD and LF contributed 4.5% and 4.7% of PM_{10} , respectively, which is close to ~5% ($PM_{2.5}$) reported by Cheng et al. (2014). These results suggest that $RIE_K = 2.9$ appear to be reasonable for our study. The large contribution of K during LNY was likely due to the intensified firework emissions (mainly firecrackers), especially between 0:00 – 3:00 am.

Following the reviewer's suggestions, we checked the ratio of m/z 39/23 throughout the study. 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. Higher ratio of m/z 39/23 (=26.4) was observed during LNY. One explanation is the fireworks burned during LNY were mainly firecrackers which are different from those during LF.

In addition, the K in this study is primarily used to identify the FW events. Although the quantification of K has some uncertainties and might affect the bulk aerosol composition of fireworks to some extent, it has negligible effects on our discussions on secondary aerosol and the impacts of reduced emissions on aerosol chemistry during the holiday because the three short-term FW events were all excluded.

2. The second major issue is related to the stability of their PMF solution and the uncertainty.

The authors did not provide any error or uncertainty of their PMF results, while they should at least present the average of a few seed runs and the standard deviations (for both factor profiles and factor time series). In the present manuscript, only profiles and time series are shown. What external tracers were used for the validation of the PMF factors?

We thank the reviewer's comments. In the revised manuscript, we significantly expanded the supplementary materials on the chosen of PMF solutions.

Fig. A2 shows the mass fraction of OA components as a function of f_{peak} values. Overall, the contribution of each OA factor was relatively stable across different f_{peak} values (average $\pm 1\sigma$; min – max): HOA ($14\pm 1.6\%$; 12 – 16%); COA ($14\pm 2.8\%$; 11 – 17%); CCOA ($19\pm 2.7\%$; 15 – 22%); OOA ($51\pm 1.7\%$; 49 – 55%). As shown in Fig. A3a, the time series of four OA factors for 3 different f_{peak} values (-1, 0, and 1) agree overall well. However, the mass spectra of OA factors have large differences (Fig. A3b). Most mass spectra of OA factors at $f_{\text{peak}} > 1$ are largely different from the standard mass spectra reported in Ng et al. (2011) and those resolved in winter 2011-2012 (Sun et al., 2013b). Figure A4 showed that the mass spectra of OA factors at $f_{\text{peak}} = -1$ presented the best correlation with those identified in winter 2011-2012 (Sun et al., 2013b). Therefore, four factor solution with $f_{\text{peak}} = -1$ was chosen in this study.

The external tracers used to validate the PMF factors include NO_3 , SO_4 , SO_4+NO_3 , Chl, BC, NO_x , CO, m/z 60, etc. Similar to previous studies (Sun et al., 2013b), OOA shows the best correlation with NO_3 ($r^2 = 0.90$) and good correlation with SO_4+NO_3 ($r^2 = 0.87$). It's interesting to note that the correlations between HOA+CCOA and BC ($r^2 = 0.88$), NO_x ($r^2 = 0.77$), and CO ($r^2 = 0.63$) are higher than those between HOA and BC/ NO_x /CO ($r^2 = 0.36 - 0.47$). These results suggest that coal combustion emissions during wintertime are also important sources of CO, BC and NO_x (Tian et al., 2008; Zhi et al., 2008). CCOA also correlates with m/z 60 ($r^2 = 0.77$). Although COA didn't have external tracers to validate, it is very distinct as suggested by its unique diurnal patterns (two peaks corresponding to meal time) and high m/z 55/57 ratio.

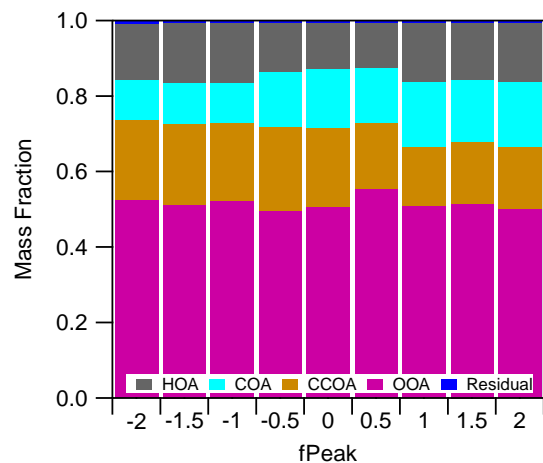


Figure A2. Mass fraction of four OA factors (from 6-factor solution; three OOA factors were combined into one OOA factor) as a function of fpeak values.

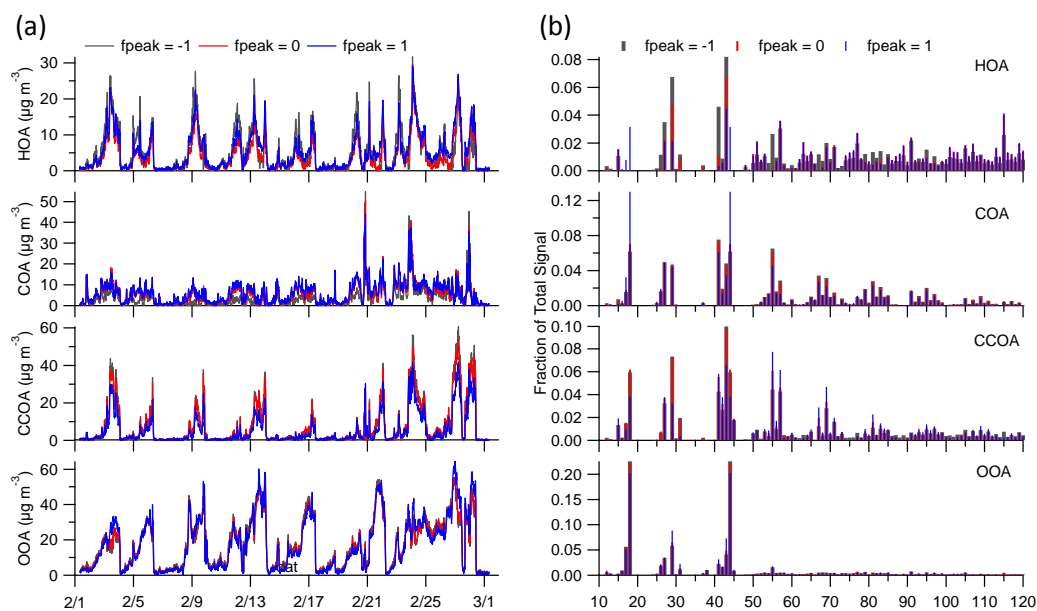


Fig. A3. Time series and mass spectra of four OA factors for three different fpeak values (-1, 0, and 1)

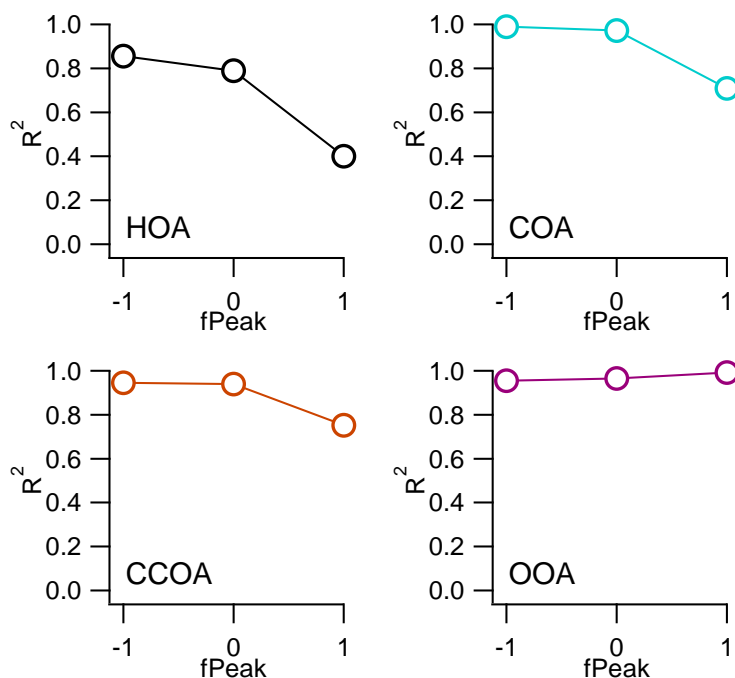


Fig. A4. Mass spectra correlations between this study and those identified in winter 2011-2012 (Sun et al., 2013b).

3. The OM fireworks contribution is apportioned to all the primary factors, this most probably means that they need one more factor at least to explain the firework episodes.

We tried to extend the number of PMF factors to 10. Unfortunately, the fireworks factor still cannot be resolved. One of the reasons is because fireworks is a small component of OA, while the true components with <5% of the mass are unlikely to be resolved accurately (Ulbrich et al., 2009).

4. Some factors seem to be mixed, e.g. high m/z 44 in COA, m/z 60 in HOA and COA. This shows the possible mixing between HOA and CCOA. I would have suspected m/z 91 or m/z 95 to be higher in CCOA than in HOA. Diurnal variations also indicate mixing of HOA and CCOA. HOA does not show any rush hour peaks while CCOA shows an evening peak. A short view at the BC diurnal of Fig S8 suggests a higher correlation of CCOA with BC than HOA. A BC source partitioning in BC from traffic and BC from wood/coal burning along the lines of Sandradewi et al. (2008) may improve the picture.

High m/z 44 in COA has been observed many times in previous studies, e.g., Allan et al. (2010), Crippa et al. (2013), Sun et al. (2011), Sun et al. (2013b), Mohr et al. (2012), etc. The high m/z 60 in COA is also consistent with the results from characterization of mass spectra of fresh cooking emissions (Allan et al., 2010; He et al., 2010). The m/z 60 in HOA is small. It's possible that HOA and CCOA were not well separated as the reviewer mentioned. The diurnal profile of HOA in Beijing didn't present a morning rush peak as those observed at other urban sites. One of the major reasons is that HOA in Beijing is primarily from the emissions of diesel trucks and heavy duty vehicles that are not allowed inside the city

between 6:00 am – 10:00 pm. Similarly, the BC didn't show the morning rush peak as well (Han et al., 2009). More detailed discussions on diurnal profile of HOA in Beijing can be found in Sun et al. (2013a). HOA and CCOA might be mixed. Unfortunately, we don't have more external tracers to further separate them like what has been done in Sandradewi et al. (2008). The reviewer is right that CCOA correlates better with BC than HOA. Indeed, HOA+CCOA correlates better than each individual HOA or CCOA, suggesting that coal combustion and traffic emissions are both important sources of BC in Beijing during wintertime (Tian et al., 2008;Zhi et al., 2008).

A recent study in central eastern China by a high resolution aerosol mass spectrometer (Hu et al., 2013) showed obvious m/z 91 and 95 peaks in HOA, but only a pronounced m/z 91 in CCOA, which is overall consistent with our results. Similarly, Zhang et al. (2014a) also observed higher m/z 91 in HOA than CCOA from the high resolution aerosol mass spectrometer measurements in Beijing during wintertime.

5. No BBOA? Why? There seems to be m/z 60 in the HOA and COA. Is f_{60} above the threshold of 0.3% (Cubison et al. 2011)?

BBOA was not resolved in this study. The BBOA is generally a small component of OA during heating season (Huang et al., 2014) although it is important during the harvest seasons in China (Zhang et al., 2014b). The small fraction of BBOA, the low sensitivity of unit mass resolution spectra of OA from ACSM measurements plus the relatively similar spectra between BBOA and CCOA (Zhang et al., 2014b) all limit the identification of BBOA factor in this study.

High m/z 60 in COA is consistent with previous results that are described above. The m/z 60 in HOA is small, but there's possibility that HOA is partially mixed with CCOA. The average f_{60} for the entire study is ~0.42%, which is slightly higher than 0.3% reported by Cubison et al. (2011). Other factors contributing to the high m/z 60 include: 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). The contributions of m/z 's > 120 were not included because of the limitation of ACSM commercial-grade mass spectrometer.

Specific comments:

1. L139: "same aerosol sampling, vaporisation and ionisation" is not correct. ACSM switches between ambient and filter while in the AMS the beam is periodically blocked by a chopper wheel. In addition, the vaporizer chamber is smaller in the ACSM and the filaments are made of different material.

The reviewer is right. We revised this sentence as "The ACSM was built upon previous versions of AMS"

2. L141: not only lower sensitivity but also lower mass resolution.

The lower mass resolution was added in the text.

3. L163: default RIE for SO₄ may significantly differ from the real SO₄_RIE (values between 0.5 and 1.4 were measured). How does the ion balance look like? Is the aerosol neutralized? Budisulistiorini et al (doi:10.5194/amt-7-1929-2014) presented a way to estimate RIE_SO₄ from the dataset.

We agree with the reviewer that the real SO₄²⁻ RIE might significantly differ from the default value. In this study, the aerosol particles appear to be fully neutralized based on the ion balance in Figure A5. Therefore, the ACSM SO₄²⁻ concentration might be reasonably determined. It should be noted that the approach proposed by Budisulistiorini et al. (2014) is based on a full balance of aerosol particles. Such an approach might introduce large uncertainties when aerosol particles are highly acidic.

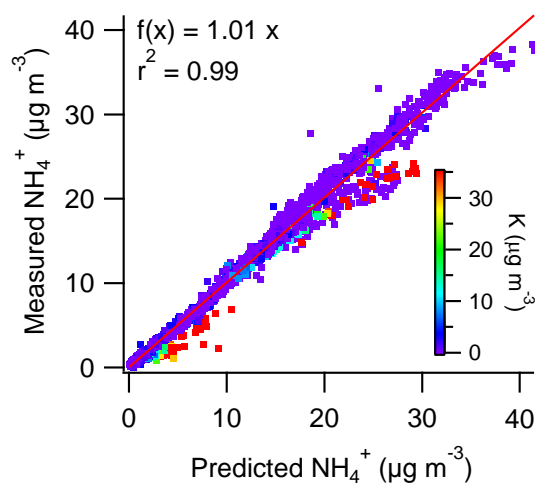


Figure A5. Correlation plot of measured NH₄⁺ versus predicted NH₄⁺ that needs to fully neutralize the aerosol particles. $\text{predicted NH}_4^+ = 18 \times (\text{SO}_4^{2-}/96 \times 2 + \text{NO}_3^-/62 + \text{Cl}/35.5)$ (Zhang et al., 2007).

4. L170: Is this assumption valid? Under winter conditions, one of the most important sources of K⁺ is biomass burning. As mentioned below, K⁺ contributes to 7% of the total PM₁ during normal days.

The assumption is rationale because biomass burning is a small fraction of organics (<6%) during wintertime (Zheng et al., 2005; Huang et al., 2014) although it is significant during harvest seasons (Zhang et al., 2014b). We didn't estimate the K⁺ during normal days because our assumption is only used to calculate the contribution of K⁺ during fireworks events. The 7% below refers to the fraction of chloride not potassium. We clarified this point in the revised manuscript.

5. L178: "inappropriate fragmentation ratio at m/z35". What was the issue? What is the reason for negative m/z 35 values? Any speculation? Could this be an indication of (slowly evaporating) sea salt?

The exact reason is still not clear yet. But very likely, it is due to the interferences of sodium chloride from fireworks which has a slow evaporation process in the oven of ACSM. Indeed, we also observed a

similar increase of m/z 23 as m/z 39 during the fireworks events. Figure 4b further shows two evident peaks at m/z 58 and 60, which is likely NaCl^+ and $\text{Na}^{37}\text{Cl}^+$, respectively.

6. L189: what external tracers were used for the validation of the PMF? Why are these correlations not shown? The validation of the PMF results is very weak! Only profiles and time series are shown.

The external tracers used to validate the PMF factors include NO_3 , SO_4 , SO_4+NO_3 , Chl, BC, NO_x , CO, m/z 60, etc. The profiles and time series of OA factors for different m/z values were also compared and presented in Figure S6 – Figure S8. Also see our detailed response to major comment 2.

7. FigS1b: Is this behavior also seen in m/z 41 (potassium isotope).

The behavior is much weaker for m/z 41 because the potassium isotope at m/z 41 is only ~7.2% of K (m/z 39).

8. FigS2: Do residuals of m/z 76 (KCl isotope) have the same pattern? And how does the residual of a purely organic fragment look like? Assumptions are only valid if peaks do not show up at organic fragments. Otherwise it is a PMF artifact. PMF does have issues with outliers. Extra caution is necessary for large singular short term plumes. They are routinely excluded from the PMF. See http://www.epa.gov/heads/documents/PMF_5.0_User_Guide.pdf: "Impacts such as fireworks represent a challenge for PMF and multivariate models because they are infrequent short duration events with high concentrations." and "The user should also examine time series for extreme events that should be excluded from modeling (for example, elevated potassium concentrations on the Fourth of July from fireworks)."

Yes, m/z 76 has the same residual pattern as m/z 74, yet the pure organic fragment m/z 's, e.g., m/z 42 and m/z 43 don't have such residual patterns (Figure A6). The results suggest that our assumption is rationale, and it's not a PMF artifact. We did check the PMF results without fireworks data. The time series and mass spectral profiles are almost identical to those by including fireworks data into PMF analysis. Since we can approximately estimate KCl^+ and $\text{K}^{37}\text{Cl}^+/\text{K}^{41}\text{Cl}^+$ based on the differences between measured and PMF modeled values, the PMF results by including fireworks data were used in this study.

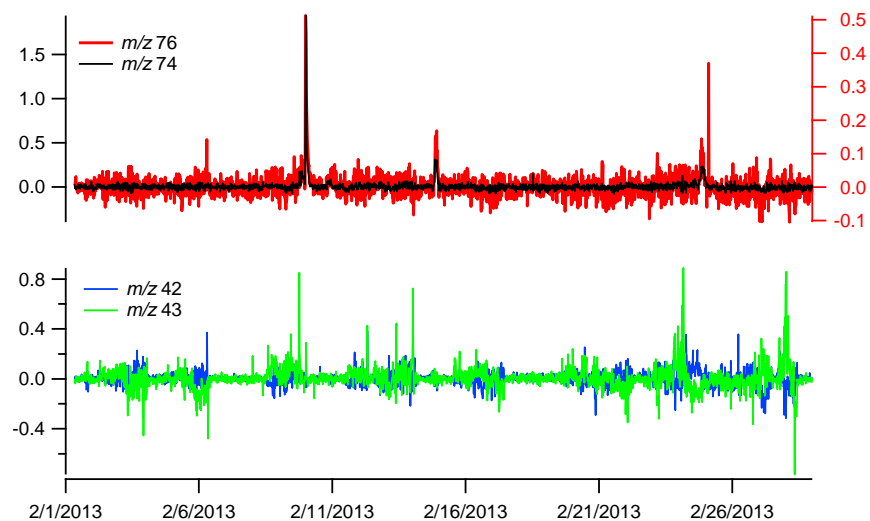


Figure A6. Time series of the PMF residuals of m/z 74, m/z 76, m/z 42, and m/z 43.

9. Fig4b: Correlation between MS_FW and LV-OOA and the following conclusion that most FW-OA is secondary is invalid!! m/z 44 and m/z 18 dominate the regression and make it almost a two point fit (18 & 44 vs rest) with misleading high R^2 . Correlation of MS_NFW or MS_BG with LV-OOA is probably also close to $R^2=0.94$. Correlation should be shown excluding 44 and 18. Spectra in Fig 4b do show significant differences, e.g. ratios 41/42, 55/57.

The R^2 recalculated by excluding m/z 18 and 44 is 0.89 indicating that there are still many similarities between FW-OA and LV-OOA. We would not expect the exact same spectra between FW-OA and LV-OOA. In fact, the mass spectrum of LV-OOA varied significantly at different sites as indicated by the error bar of the standard LV-OOA (see Figure 1 in Ng et al., 2011). The m/z ratios also varied largely depending on the sources and aging processes. The main feature of secondary LV-OOA is the high m/z 44 (mainly CO_2^+) in the spectrum. Higher fraction of m/z 44 (f_{44}) associated with higher oxygen-to-carbon ratio (Aiken et al., 2008) generally indicates more oxidized organic aerosol. Compared to secondary OA, the primary OA, e.g., hydrocarbon-like OA, cooking OA, biomass burning OA etc. generally show much lower f_{44} . Considering above, the high f_{44} in FW-OA does suggest oxidized OA in this work. Although the exact sources and formation mechanisms are not very clear yet, they are likely from the burning or oxygenated organic compounds in fireworks. Our results are also consistent with those reported by Drewnick et al. (2006) that the OOA-related m/z 's (e.g., m/z 44) showed large enhancements during New Year's fireworks, yet the HOA-related m/z 's (e.g., m/z 57) didn't.

10. Fig3: Potassium quantification with extremely high uncertainties in pie charts.

We agree that the potassium quantification has uncertainties. However, based on the comparisons with previous studies (see our response to major comment 1), the $\text{RIE}_K = 2.9$ and quantified K^+ appear to be reasonable for this work.

11. FigS9: same as Fig8.

Fig. 8 shows the average ratios of HD/NHD by excluding both clean periods (Figure 1) and three fireworks events. Fig. S9 presents the average ratios of HD/NHD by excluding only three fireworks events. They look similar, but different.

References:

- Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with High-Resolution Time-of-Flight Aerosol Mass Spectrometry, *Environ. Sci. Technol.*, **42**, 4478-4485, 2008.
- Allan, J. D., Williams, P. I., Morgan, W. T., Martin, C. L., Flynn, M. J., Lee, J., Nemitz, E., Phillips, G. J., Gallagher, M. W., and Coe, H.: Contributions from transport, solid fuel burning and cooking to primary organic aerosols in two UK cities, *Atmos. Chem. Phys.*, **10**, 647-668, 2010.
- Budisulistiorini, S. H., Canagaratna, M. R., Croteau, P. L., Baumann, K., Edgerton, E. S., Kollman, M. S., Ng, N. L., Verma, V., Shaw, S. L., Knipping, E. M., Worsnop, D. R., Jayne, J. T., Weber, R. J., and Surratt, J. D.: Intercomparison of an Aerosol Chemical Speciation Monitor (ACSM) with ambient fine aerosol measurements in downtown Atlanta, Georgia, *Atmos. Meas. Tech.*, **7**, 1929-1941, 10.5194/amt-7-1929-2014, 2014.
- Cheng, Y., Engling, G., He, K.-b., Duan, F.-k., Du, Z.-y., Ma, Y.-l., Liang, L.-l., Lu, Z.-f., Liu, J.-m., Zheng, M., and Weber, R. J.: The characteristics of Beijing aerosol during two distinct episodes: Impacts of biomass burning and fireworks, *Environ. Pollut.*, **185**, 149-157, <http://dx.doi.org/10.1016/j.envpol.2013.10.037>, 2014.
- Crippa, M., DeCarlo, P. F., Slowik, J. G., Mohr, C., Heringa, M. F., Chirico, R., Poulain, L., Freutel, F., Sciare, J., Cozic, J., Di Marco, C. F., Elsasser, M., Nicolas, J. B., Marchand, N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E., Zimmermann, R., Jaffrezo, J. L., Prévôt, A. S. H., and Baltensperger, U.: Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris, *Atmos. Chem. Phys.*, **13**, 961-981, 10.5194/acp-13-961-2013, 2013.
- Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., Brune, W. H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J., Mikoviny, T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J., Wisthaler, A., and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies, *Atmos. Chem. Phys.*, **11**, 12049-12064, 10.5194/acp-11-12049-2011, 2011.

- Drewnick, F., Hings, S. S., Curtius, J., Eerdeken, G., and Williams, J.: Measurement of fine particulate and gas-phase species during the New Year's fireworks 2005 in Mainz, Germany, *Atmos. Environ.*, **40**, 4316-4327, 10.1016/j.atmosenv.2006.03.040, 2006.
- Han, S., Kondo, Y., Oshima, N., Takegawa, N., Miyazaki, Y., Hu, M., Lin, P., Deng, Z., Zhao, Y., Sugimoto, N., and Wu, Y.: Temporal variations of elemental carbon in Beijing, *J. Geophys. Res.*, **114**, D23202, doi:23210.21029/22009JD012027, 2009.
- He, L. Y., Lin, Y., Huang, X. F., Guo, S., Xue, L., Su, Q., Hu, M., Luan, S. J., and Zhang, Y. H.: Characterization of high-resolution aerosol mass spectra of primary organic aerosol emissions from Chinese cooking and biomass burning, *Atmos. Chem. Phys.*, **10**, 11535-11543, 10.5194/acp-10-11535-2010, 2010.
- Hu, W. W., Hu, M., Yuan, B., Jimenez, J. L., Tang, Q., Peng, J. F., Hu, W., Shao, M., Wang, M., Zeng, L. M., Wu, Y. S., Gong, Z. H., Huang, X. F., and He, L. Y.: Insights on organic aerosol aging and the influence of coal combustion at a regional receptor site of central eastern China, *Atmos. Chem. Phys.*, **13**, 10095-10112, 10.5194/acp-13-10095-2013, 2013.
- Huang, R.-J., Zhang, Y., Bozzetti, C., Ho, K.-F., Cao, J.-J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., Haddad, I. E., and Prevot, A. S. H.: High secondary aerosol contribution to particulate pollution during haze events in China, *Nature*, advance online publication, 10.1038/nature13774
<http://www.nature.com/nature/journal/vaop/ncurrent/abs/nature13774.html#supplementary-information>, 2014.
- Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, *Atmos. Chem. Phys.*, **12**, 1649-1665, 10.5194/acp-12-1649-2012, 2012.
- Ng, N. L., Canagaratna, M. R., Jimenez, J. L., Zhang, Q., Ulbrich, I. M., and Worsnop, D. R.: Real-Time Methods for Estimating Organic Component Mass Concentrations from Aerosol Mass Spectrometer Data, *Environ. Sci. Technol.*, **45**, 910-916, 10.1021/es102951k, 2011.
- Sandradewi, J., Pröhner, A. S. H., Alfarra, M. R., Szidat, S., Wehrli, M. N., Ruff, M., Weimer, S., Lanz, V. A., Weingartner, E., Perron, N., Caseiro, A., Kasper-Giebl, A., Puxbaum, H., Wacker, L., and Baltensperger, U.: Comparison of several wood smoke markers and source apportionment methods for wood burning particulate mass, *Atmos. Chem. Phys. Discuss.*, **8**, 8091-8118, 2008.
- Slowik, J. G., Stroud, C., Bottenheim, J. W., Brickell, P. C., Chang, R. Y. W., Liggio, J., Makar, P. A., Martin, R. V., Moran, M. D., Shantz, N. C., Sjostedt, S. J., van Donkelaar, A., Vlasenko, A., Wiebe, H. A., Xia, A. G., Zhang, J., Leitch, W. R., and Abbatt, J. P. D.: Characterization of a large biogenic secondary organic aerosol event from eastern Canadian forests, *Atmos. Chem. Phys.*, **10**, 2825-2845, 2010.

- Sun, Y., Wang, Z., Fu, P., Yang, T., Jiang, Q., Dong, H., Li, J., and Jia, J.: Aerosol Composition, Sources and Processes during Wintertime in Beijing, China, *Atmos. Chem. Phys. Discuss.*, **13**, 2077-2123, doi:10.5194/acpd-13-2077-2013, 2013a.
- Sun, Y. L., Zhang, Q., Schwab, J. J., Demerjian, K. L., Chen, W. N., Bae, M. S., Hung, H. M., Högrefe, O., Frank, B., Rattigan, O. V., and Lin, Y. C.: Characterization of the sources and processes of organic and inorganic aerosols in New York City with a high-resolution time-of-flight aerosol mass spectrometer, *Atmos. Chem. Phys.*, **11**, 1581-1602, 10.5194/acp-11-1581-2011, 2011.
- Sun, Y. L., Wang, Z. F., Fu, P. Q., Yang, T., Jiang, Q., Dong, H. B., Li, J., and Jia, J. J.: Aerosol composition, sources and processes during wintertime in Beijing, China, *Atmos. Chem. Phys.*, **13**, 4577-4592, 10.5194/acp-13-4577-2013, 2013b.
- Tian, L., Lucas, D., Fischer, S. L., Lee, S. C., Hammond, S. K., and Koshland, C. P.: Particle and Gas Emissions from a Simulated Coal-Burning Household Fire Pit, *Environ. Sci. Technol.*, **42**, 2503-2508, 10.1021/es0716610, 2008.
- Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, *Atmos. Chem. Phys.*, **9**, 2891-2918, 2009.
- Zhang, J. K., Sun, Y., Liu, Z. R., Ji, D. S., Hu, B., Liu, Q., and Wang, Y. S.: Characterization of submicron aerosols during a month of serious pollution in Beijing, 2013, *Atmos. Chem. Phys.*, **14**, 2887-2903, 10.5194/acp-14-2887-2014, 2014a.
- Zhang, Q., Jimenez, J. L., Worsnop, D. R., and Canagaratna, M.: A case study of urban particle acidity and its effect on secondary organic aerosol, *Environ. Sci. Technol.*, **41**, 3213-3219, 2007.
- Zhang, Y. J., Tang, L. L., Wang, Z., Yu, H. X., Sun, Y. L., Liu, D., Qin, W., Zhang, H. L., and Zhou, H. C.: Insights into characteristics, sources and evolution of submicron aerosols during harvest seasons in Yangtze River Delta (YRD) region, China, *Atmos. Chem. Phys. Discuss.*, **14**, 9109-9154, 10.5194/acpd-14-9109-2014, 2014b.
- Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C. S., Zhang, Y., and Cass, G. R.: Seasonal trends in PM_{2.5} source contributions in Beijing, China, *Atmos. Environ.*, **39**, 3967-3976, DOI: 10.1016/j.atmosenv.2005.03.036, 2005.
- Zhi, G., Chen, Y., Feng, Y., Xiong, S., Li, J., Zhang, G., Sheng, G., and Fu, J.: Emission characteristics of carbonaceous particles from various residential coal-stoves in China, *Environ. Sci. Technol.*, **42**, 3310-3315, 2008.