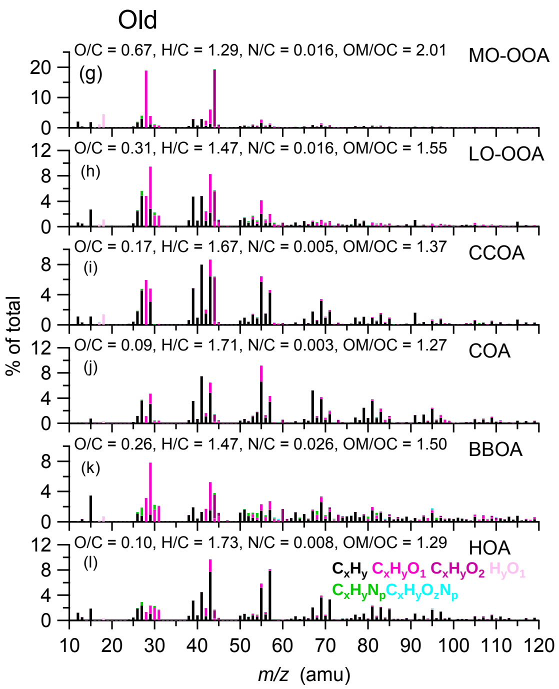
We thank Prof. Eleanor Browne for her very helpful, insightful and constructive comments. We have carefully considered these comments (in italic), and responded to each of them (in blue or red), as appended below:

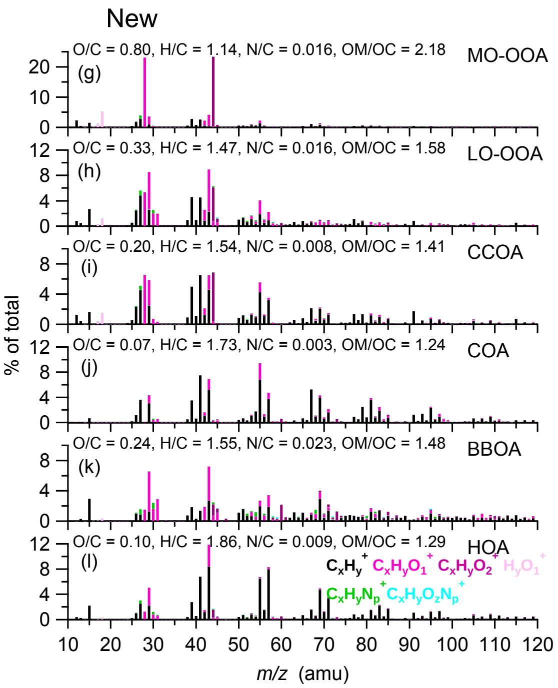
Major comments:

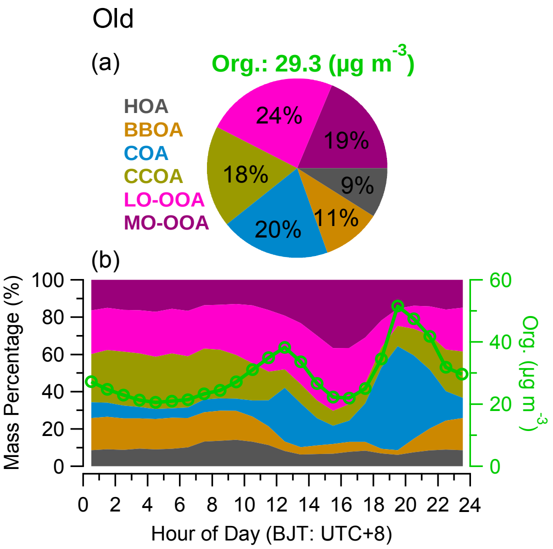
MC1. Referee 2 expressed concerns regarding the removal of highly polluted periods prior to PMF analysis. I agree that this needs to be addressed in greater detail in the manuscript – it appears to me that nothing has been changed. Additionally, I do not understand the authors' reply to this comment. Mainly, I do not understand what is meant by "ion fitting in this period is bad." With high signals, it is typically easier to do ion fitting. Do the authors mean that new ions appear? If so, those should be fitted and addressed. The discussion on PMF results with and without this time period should at least be included in the supplementary material.

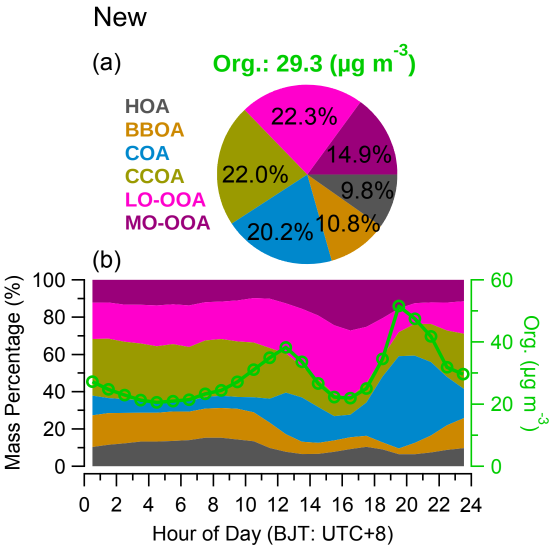
We feel sorry that we didn't elaborate this issue with more details in the previous version. We now have re-visited and investigated this issue. Originally, we did remove a highly polluted short period on Jan. 22-23 from the PMF analyses, as our very preliminary trial on the PMF including this period showed the mass concentrations of the OA on this day was relatively not well reproduced compared with other periods. Thus, we didn't include it in the subsequent PMF treatment. However, we now re-visited this issue, and in fact, after careful PMF matrix and error preparation following the procedure outlined by Zhang et al. (2011) (including removal of a few outlier runs rather than all data on Jan 22-23, removal of highly noisy ions, etc.), the PMF modeling on this polluted day was improved significantly, thus after careful consideration, we think it is better to add back this period. In addition, adding back the polluted day also makes it more reasonable and reliable to compare the source apportionment from carbon isotope analysis results with the AMS-PMF resolved results.

The results of the new PMF analysis results covering the whole sampling period are overall similar with previous results but there are some changes between them, such as the element ratios of the factor mass profiles, mass contributions and time series of the PMF factors (as shown in the Figures below). For example, the O/C ratio of MO-OOA increased from 0.67 to 0.80 due to that a bit more $C_xH_y^+$ signals are apportioned in the primary factors including COA, BBOA and OOA, and the mass contribution of MO-OOA correspondingly decreased from 18% to 15% on average. The diurnal pattern of HOA also shows differences with the previous one – the new HOA pattern showed relatively high level throughout the nighttime (Fig. R1). The characteristics of other PMF factors are similar with previous results. We have listed the key figures that compare the old and new PMF results for reference below. Correspondingly, all PMF results (including figures and relevant text) in the manuscript were updated in this modified version.


Old
O/C = 0.67, H/C = 1.29, N/C = 0.016, OM/OC = 2.01
MO-OOA
(g)
O/C = 0.31, H/C = 1.47, N/C = 0.016, OM/OC = 1.55
LO-OOA
(h)
O/C = 0.17, H/C = 1.67, N/C = 0.005, OM/OC = 1.37
CCOA
(i)
O/C = 0.09, H/C = 1.71, N/C = 0.003, OM/OC = 1.27
COA
(j)
O/C = 0.26, H/C = 1.47, N/C = 0.026, OM/OC = 1.50
BBOA
(k)
O/C = 0.10, H/C = 1.73, N/C = 0.008, OM/OC = 1.29
HOA
(l)
C_xH_y $C_xH_yO_1$ $C_xH_yO_2$ H_yO_1
$C_xH_yN_p$ $C_xH_yO_zN_p$
% of total
m/z (amu)
10 20 30 40 50 60 70 80 90 100 110 120


New
O/C = 0.80, H/C = 1.14, N/C = 0.016, OM/OC = 2.18
MO-OOA
(g)
O/C = 0.33, H/C = 1.47, N/C = 0.016, OM/OC = 1.58
LO-OOA
(h)
O/C = 0.20, H/C = 1.54, N/C = 0.008, OM/OC = 1.41
CCOA
(i)
O/C = 0.07, H/C = 1.73, N/C = 0.003, OM/OC = 1.24
COA
(j)
O/C = 0.24, H/C = 1.55, N/C = 0.023, OM/OC = 1.48
BBOA
(k)
O/C = 0.10, H/C = 1.86, N/C = 0.009, OM/OC = 1.29
HOA
(l)
$C_xH_y^+$ $C_xH_yO_1^+$ $C_xH_yO_2^+$ $H_yO_1^+$
$C_xH_yN_p^+$ $C_xH_yO_zN_p^+$
% of total
m/z (amu)
10 20 30 40 50 60 70 80 90 100 110 120


Old
(a)
Org.: 29.3 (μg m-3)
HOA
BBOA
COA
CCOA
LO-OOA
MO-OOA
24%
19%
18%
9%
11%
20%
(b)
Mass Percentage (%)
Org. (μg m-3)
Hour of Day (BJT: UTC+8)
0 2 4 6 8 10 12 14 16 18 20 22 24


New
(a)
Org.: 29.3 (μg m-3)
HOA
BBOA
COA
CCOA
LO-OOA
MO-OOA
22.3%
14.9%
22.0%
9.8%
10.8%
20.2%
(b)
Mass Percentage (%)
Org. (μg m-3)
Hour of Day (BJT: UTC+8)
0 2 4 6 8 10 12 14 16 18 20 22 24

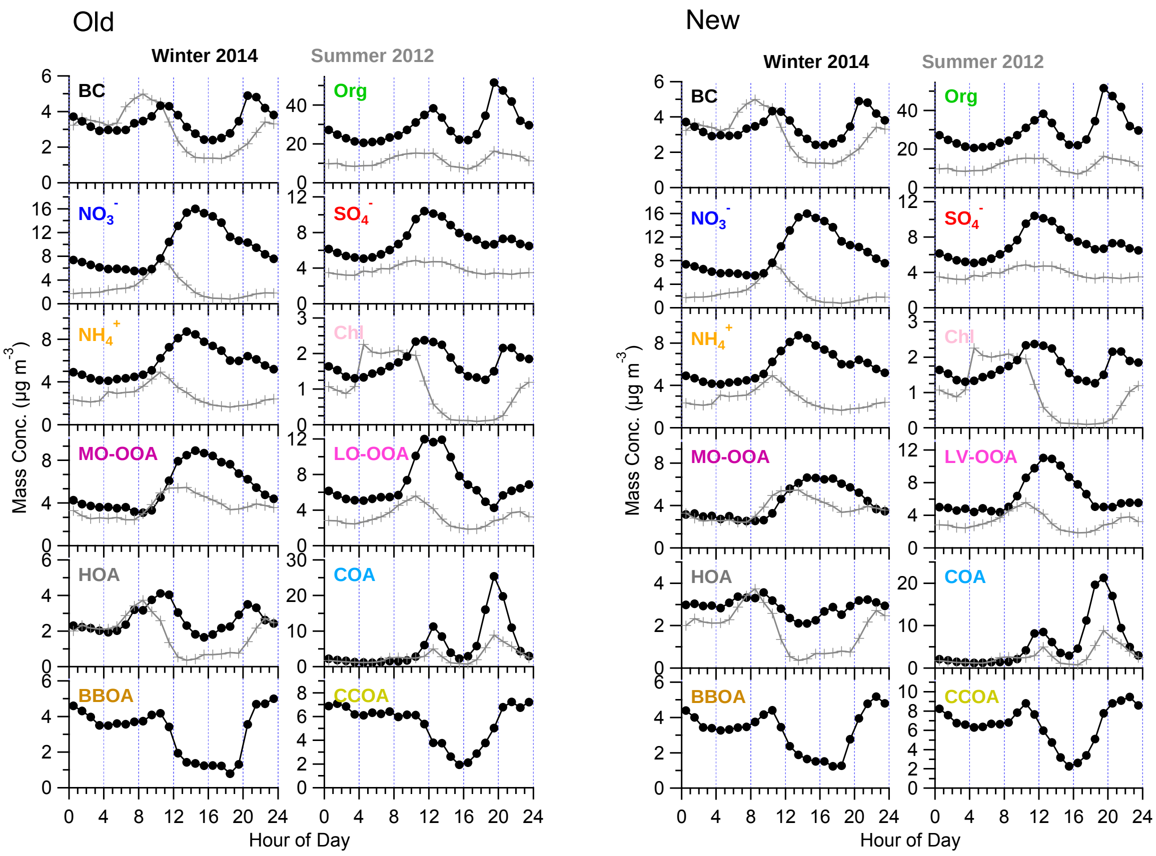


Fig. R1 The comparison between the old and new PMF results

MC2. Referee 2 also expressed concerns regarding the conclusions drawn from the 14C analysis and the representativeness of the samples. I think that the authors should reconsider their interpretation of these results. In particular, I am hesitant to agree with the statement that the "…air mass and aerosol source are pretty stable…" when applied to the 14C measurements. One of the 14C measurements (January 23) was performed on a day that was removed from the data analysis due to "highly polluted" conditions, and another (January 3) was during a period before the start of the AMS measurements. Given that 50% of the filters are from time periods where the AMS data is not even considered in the manuscript, how much can we really learn from this analysis and is this analysis even appropriate?

We thanks for the comment. Indeed, as carbon isotope analysis is difficult for us to perform, we in this study aimed to make a try to combine it with the AMS-PMF results to probe more information regarding the fossil/non-fossil sources for the OA in Lanzhou. Currently, we selected four days in every week during the sampling month, trying to use this daily filter sample to represent a week average by assuming the meteorological conditions aerosol sources are relatively stable. We agree with the editor and the reviewer that the sampling schedule should be improved and it will be better if more samples were included so as to make the analysis more robust. This certainly will be the subject of our future work. For the current analyses, the wind speed was very low (on average ranging from 0.6 to 1.1 m s^{-1}) and the prevailing wind direction was basically typical (north and northeast) during the field study (Fig. R2). This in some extent verifies our assumption that the daily sample are in some extent representative. In addition, we have added back the PMF analysis on Jan 23, so the analysis is improved from the previous version. In addition, the very similar carbon isotopic results (the total average of f_{NF} in these four filters was 55 ± 3%) further confirm the representativeness of the samples. Nevertheless, cautions and limitation of our results should be clearly stated and open to the readers, thus, we have added a few sentences in the MS to make it clear, as following:

"Here, we use the results of these four filter samples to roughly represent the average situation of the field sampling because of the relative stable meteorological conditions (section 3.1.1) and similar aerosol sources during the field study (section 3.1.3). Due to the limitation of the small amount of filter samples, the results based on this carbon isotopic data are preliminary and comprehensive validation is an ongoing work."

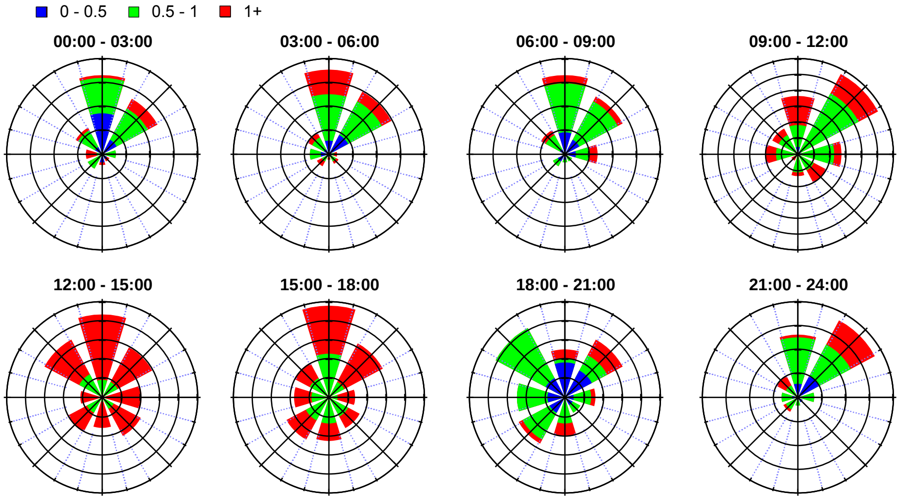


Fig. R2 Three hourly average wind rose plot during the field study

MC3. The comment from Referee 2 regarding the CCOA factor also needs to be addressed further in the manuscript. Given the similar time trends of CCOA and BBOA, how certain is it that m/z 91 and the PAH-related ions are related to CCOA and not BBOA, especially given that the BBOA spectrum is somewhat constrained? Please include discussion addressing these points in the manuscript.

Because the similar conventional usage of biomass and coal for heating during cold seasons in Lanzhou, it is understandable that the BBOA and CCOA had similar temporal variations. In addition, the biomass usage is relatively in a small amount, thus by using the general PMF (unconstrained mass spectra), it is difficult to separate the BBOA and CCOA. Instead, we constrained the BBOA mass spectrum (MS) using ME2, and thus we can extract the BBOA and separated another factor that is CCOA. As shown in Fig. R3, indeed the temporal variations of BBOA and CCOA were similar (R^2 = 0.77), yet their mass spectra were significantly different (R^2 = 0.35). As is well known, the standard MS of BBOA is usually characterized by the high signals at m/z 60 and 73, and the BBOA factor shows this feature. On the other hand, a high signal fraction of m/z 91, is a common feature of CCOA found in previous studies (Dall'Osto et al., 2013). The CCOA MS was sometimes also characterized by high signal of CO_2^+ due to the emission of organic acids directly from coal combustion (Zhang et al., 2008). In northern China, coal combustion had been thought to be a very important source of OA due to the large and widespread use of coal for heating and other purposes during wintertime (accounted for 60% of the energy consumption), and the CCOA factor was also found in several other Chinese cities (Elser et al., 2016; Hu et al., 2016; Sun et al., 2016). In general, our CCOA factor is highly similar to those CCOA identified earlier.

Furthermore, although both the MS of BBOA and CCOA can contain the PAHs-related ions (m/z), e.g., m/z 152, 165, 178, 189, 202, 215, 226, 239, 252, 276, 300, 326, etc (Sun et al., 2016). Due to widely and more enhanced use of coal in the northern China, coal combustion was found to be the most important source of PAH during wintertime (Okuda, et al., 2006). A study by Sun et al. (2016) also suggested that 66% of PAH was from coal combustion and only 18% was from biomass burning in Beijing. To address referee's concerns, in this section we have added the following sentences to explain our PMF results:

"Note that although the similar temporal variations between BBOA and CCOA, the significant differences between their MS (in particular, m/z 91) suggested their different origins. In addition, high PAH signals had been observed in the CCOA MS, and this is consistent with previous results that the coal combustion could be a dominate source of PAHs in China (Okuda et al., 2006; Sun et al., 2016). "

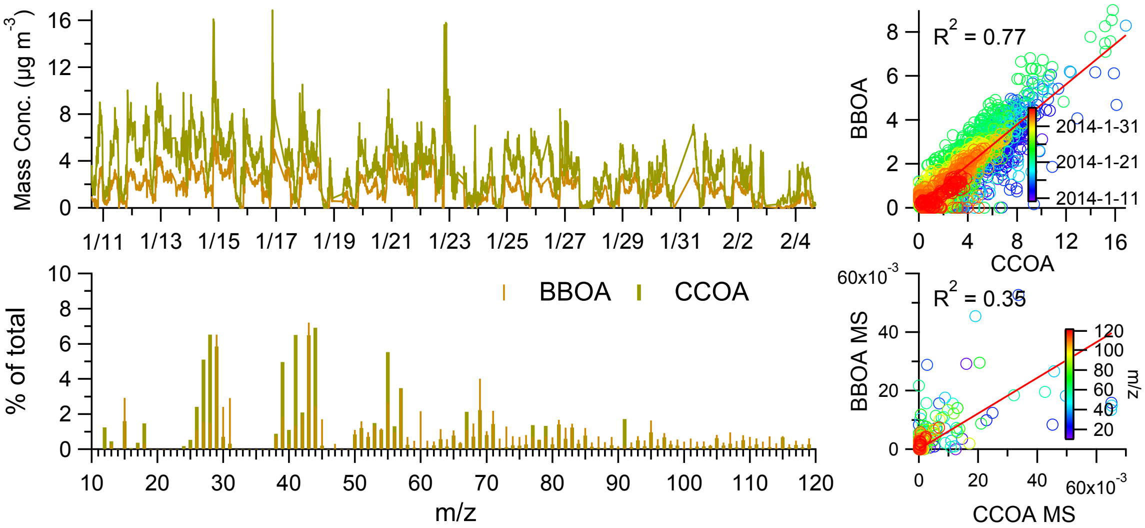


Fig. R3 The comparisons of MS and time series between BBOA and CCOA

Specific Comments

SC1. Line 120: "…applying thousands of individual species…" Given that vaporization and ionization in the AMS result in extensive fragmentation, the AMS does not really measure "individual species." I recommend revising since this is somewhat misleading.

We have reworded this sentence as following:

"Source apportionment techniques, such as the positive matrix factorization (PMF) allow us to use thousands of fragment ions for source identification and use the real measurement uncertainties to constrain the fitting, and would thus appear more suitable to identify and apportion PM to their sources".

SC2. Line 131-132: I think a few more references would be appropriate. Particularly some that represents the more recent advancements in models.

We have added two more references in this sentence as following:

"However, the formation and evolution mechanisms of those secondary species were poorly understood, and previous models tended to underestimate the secondary species budget in polluted regions (e.g., Volkamer et al., 2006; Carlton et al., 2010; Hodzic et al., 2016)."

Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. O., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model Representation of Secondary Organic Aerosol in Cmaqv4.7, Environ. Sci. Technol., 44, 8553-8560, doi:10.1021/es100636q, 2010.

Hodzic, A., Kasibhatla, P. S., Jo, D. S., Cappa, C. D., Jimenez, J. L., Madronich, S., and Park, R. J.: Rethinking the Global Secondary Organic Aerosol (Soa) Budget: Stronger Production, Faster Removal, Shorter Lifetime, Atmos. Chem. Phys., 16, 7917-7941, doi:10.5194/acp-16-7917-2016, 2016.

Volkamer, R., Jimenez, J. L., San Martini, F., Dzepina, K., Zhang, Q., Salcedo, D., Molina, L. T., Worsnop, D. R., and Molina, M. J.: Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected, Geophys. Res. Lett., 33, L17811, doi:10.1029/2006GL026899, 2006.

SC3. Line 135: "…appear to be advance…" This does not make sense. Please revise.

We have reworded this sentence as following:

"Online instruments based on mass spectrometric techniques, such as Aerodyne aerosol mass spectrometer (AMS) (Jayne et al., 2000), has advantages on probing the fast aerosol chemical processes because that the instrument can output data with a large amount of chemical information and its fine time resolution (in minutes) and mass sensitivity (in ng m^{-3}) (Canagaratna et al., 2007)."

SC4. Line 156-157: The aerosols are also influenced by very different meteorological processes between the two seasons.

Revised as suggested.

"Thus aerosols are influenced largely by very different meteorological conditions and chemical processes between the two seasons."

SC5. Line 219: "ionic path" This is non-standard. Please consider more standard wording.

We have reworded this sentence as following:
"The mass spectrometer in the detection section works in two modes based on the shape of the ion path, i.e., V-mode and W-mode, with high sensitivity and high chemical resolution (~6000 m/Δm), respectively."

SC6. Line 231: If a background was determined only once in the study, how was the gas-phase CO2 correction determined for other times during the campaign? Using a constant value is likely not appropriate, and given the emphasis put on CO2+, this needs to be explained.

We agree with the referee that conducting frequent filtered air measurements during the period of field study is useful and we will try to do so in the future. For AMS study, it is better to apply on-line measurement of atmospheric CO_2 concentrations for gas-phase CO_2 correction (Collier et al., 2013); however, we do not have CO_2 measurement data nearby our site. We have estimated the uncertainty of this artifact for organic-CO_2 in previous study (Xu et al., 2014). For a range of 350–500 ppm of gas phase CO_2, the organic-equivalent concentration of CO_2^+ is in the range of 0.22–0.31 μg m^{-3}. Based on our filtered air measurement, we estimate an average CO_2 concentration of 400 ppm during this study, which corresponds to ~ 0.25 org-eq μg m^{-3} of CO_2^+. This value was incorporated in the fragmentation table and subtracted from measured CO_2^+ signal to determine $OrgCO_2$ (or Org44). Given that the average $OrgCO_2$ of this study is ~ 1.7 org-eq μg m^{-3}, we estimate that applying a constant gas phase CO_2 subtraction may introduce ~ −2 – 4% uncertainty in fCO_2^+, which is very small. This point is now mentioned in the updated manuscript as following:

"Note that since no in-situ measurement of gas phase CO_2, the subtraction of a constant CO_2 signal (400 ppm based on filtered-air measurement in this study) may introduce uncertainties in the quantification of the organic-CO_2^+ signal. However, this artifact was expected to be small (less than 5% error in organic-CO_2^+ quantification) due to the high OA concentration (Xu et al., 2014)."

SC7. Sect. 2.2.1: How was the size measurement of the AMS calibrated? Significant time is spent discussing diameter later in the manuscript so this must be addressed.

The size calibration was performed following the general protocol used in the AMS community. We used standard polystyrene latex (PSL) spheres (Duke Scientific Corp., Palo Alto, CA) (100-700nm) and mono-dispersed ammonium nitrate particles (100-300nm), respectively. This information was now added in the updated manuscript:

"The instrument was calibrated for ionization efficiency (IE), inlet flow rate, and particle sizes using the standard procedure described by (Jayne et al., 2000). For example, the size calibration was performed following the general protocol used in the AMS community. We used standard polystyrene latex (PSL) spheres (Duke Scientific Corp., Palo Alto, CA) (100-700nm) and mono-dispersed ammonium nitrate particles (100-300nm), respectively. "

SC8. Line 248-249: "…refractory mass of the particle quantified by detection of the main light-absorbing component is rBC." This is unclear and should be rephrased.

We have reworded this sentence as following:
"The SP2 uses an intra-cavity Nd:YAG laser at 1064 nm to determine the light scattering and laser-induced incandescence of individual rBC (namely material associated with a strongly absorbing component at 1064 nm)."

SC9. Line 283: What density was assumed in making the comparison with the SMPS? Is a slope of 1.48 really good? Or was no density applied?

Sorry that we didn't make it clear here. We did not apply the density in the comparison between AMS and SMPS data. The slope (1.48) is the scatter plot between AMS mass concentrations and SMPS-determined volumes (assuming spherical particles). This slope can generally be used to represent the average density of the PM_1 and is comparable with other studies. The detail discussion is in the section 3.1.2.

We have reworded this sentence as following:
"This CE value was further validated by the consistency and reasonable slope between HR-ToF-AMS measured mass concentrations and SMPS-determined particle volumes (section 3.1.2, R^2 = 0.9, slope = 1.48)."

SC10. Line 335: "pretty stable" is not very meaningful and should be quantified better. Please see general comments for other concerns regarding this section.

We have reworded this sentence as response in MC2.

SC11. Line 344: Where does the factor of 1.03 come from?

A reference has been added as following:
"Zhang, Y. L., Huang, R. J., El Haddad, I., Ho, K. F., Cao, J. J., Han, Y., Zotter, P., Bozzetti, C., Daellenbach, K. R., Canonaco, F., Slowik, J. G., Salazar, G., Schwikowski, M., Schnelle-Kreis, J., Abbaszade, G., Zimmermann, R., Baltensperger, U., Prévôt, A. S. H., and Szidat, S.: Fossil vs. Non-fossil sources of fine carbonaceous aerosols in four chinese cities during the extreme winter haze episode of 2013, Atmos. Chem. Phys., 15, 1299-1312, 10.5194/acp-15-1299-2015, 2015b."

SC12. Line 363: Should fNF_BBOC be less than 1? That is my understanding from the wording. Please clarify.

Thanks. The original sentence was used to describe the possible source of biomass burning from soft coal, but we could not find the exact ratio of this kind of burning emission. So we assume the contribution of this soft coal to non-fossil fuel carbon was negligible .

This sentence has been revised as following:
"BBOC is estimated to be originated from biomass burning, i.e., $f_{\mathrm{NF\_BBOC}} = 1$;"

SC13. Line 376: Some of the average values (particularly temperature) are not very convincing in support of the statement of stable conditions. I assume this is due to diurnal variation. Perhaps it would be better to present average lows and highs.

Agree, the description for meteorological conditions have been changed to the average diurnal variation as following:

"The measurement site mainly received air masses from northern and northeastern directions associated with low wind speeds (WS) ranging from 0.6 to 1.1 m s^{-1} (on daily average: 0.8 ± 0.2 m s^{-1}). The mountains to the north and south of the city could significantly reduce the wind speeds. Air temperature ranged from −5.0 to 6.6 °C (average = 0.6 ± 3.9 °C) for the diurnal variation during the campaign, but had an evident increase after the Chinese New Year (January 31, 2014) (Fig. 1a). No precipitation event occurred during the campaign, and RH was pretty low ranging from 16.8 to 39.5% (on daily average = 27.5 ± 7.4%) for the diurnal variation. Overall, the meteorological conditions during the campaign were much stable and dryer than those during summer 2012 (on average: 1.2 ± 0.6 m s^{-1} for WS and 60 ± 17 % for RH)."

SC14. Lines 394-396: Do you mean to say that you have measured a lower limit?

Yes, this value represents a lower limit.

SC15. Sect 3.1.4: The size distribution of chloride seems more similar to the size distribution of the organics than to the other inorganics. What are the implications of this and is it consistent with your factor analysis?

Due to lack of other data, we don’t know exactly the reason why the consistent size distribution between chloride and organics. All the fragments of chloride show similar size distribution (Fig. R4). One possible reason may be related with organochlorine which could be emitted from coal combustion and trash burning. The temporal variations of chloride in this study indeed showed good correlations with BBOA, CCOA, and LO-OOA (R^2, 0.45 – 0.52).

We have added a sentence in this section for explaining this phenomenon:

"Note that chloride also showed a wider distribution which was more similar with organics other than sulphate and nitrate. This was not observed during 2012 summer and could be related with OA emitted from coal combustion and biomass burning during wintertime."

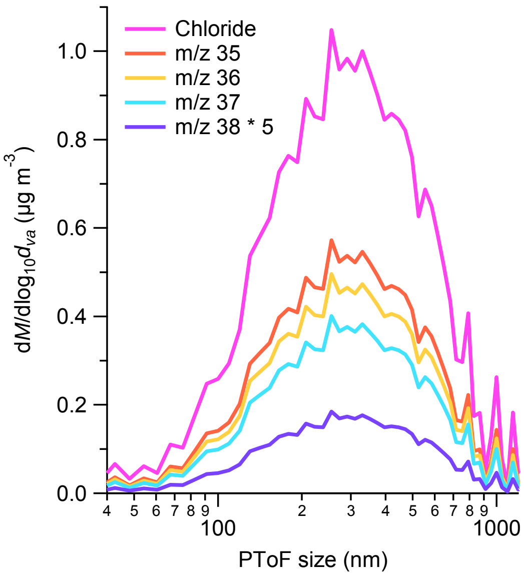


Fig. R4 The size distribution chloride and its corresponding fragments

SC16. Line 460-462: I find this highly speculative since the oxidizing capacity is greatly reduced in the winter. Is the increase in SO2 really enough to account for changes in sulfate given lower oxidizing conditions?

We agree that the oxidizing capacity is greatly reduced during the wintertime, which has recently aroused widely concerns such as WINTER field campaign in USA. Although, the production of OH from ozone photolysis is reduced by more than an order of magnitude, recent studies have shown that nitryl chloride (ClNO2), together with nitrous acid (HONO), can be an important source of OH radicals in the wintertime (Young et al., 2012). In addition, the increased aerosol volume/surface is another potential factor contributing the sulfate formation during wintertime in Beijing, China (Zheng et al., 2015). The production rate of sulfate through heterogeneous reactions can be estimated by

$$\frac{dC_{S(IV)}}{dt} \approx k[S(IV)(aq)] \cdot [oxidants\ (aq)] \cdot V_{aerosol}$$

In which $C_{S(IV)}$ is the sulfate concentration, k is the effective rate coefficient, $[S(IV)(aq)]$ is the $S(IV)$ concentration in the aqueous phase of aerosol, $[oxidants\ (aq)]$ is the concentration of oxidants in the aqueous phase of aerosols, and $V_{aerosol}$ is the volume concentration of humidified aerosol at ambient.

We have reworded this sentence in the updated manuscript as following:

"The significantly higher concentration of sulphate during winter than summer could result from a higher amount of precursor SO_2 emission, wintertime hydroxyl radical formation, and the increased aerosol particle surface due to high PM loadings that facilitated the heterogonous conversion of SO_2 to sulphate in winter (Yong et al., 2012; Puaede et al., 2015; Zheng et al., 2015)."

Young, C. J., Washenfelder, R. A., Roberts, J. M., Mielke, L. H., Osthoff, H. D., Tsai, C., Pikelnaya, O., Stutz, J., Veres, P. R., Cochran, A. K., VandenBoer, T. C., Flynn, J., Grossberg, N., Haman, C. L., Lefer, B., Stark, H., Graus, M., de Grouw, J., Gilman, J. B., Kuster, W. C., and Brown, S. S.: Vertically resolved measurements of nighttime radical reservoirs in Los Angeles and their contribution to the urban radical budget, Environ. Sci. Technol., 46, 10965–10973, doi:10.1021/es302206a, 2012.

Pusede, S. E., VandenBoer, T. C., Murphy, J. G., Markovic, M. Z., Young, C. J., Veres, P. R., Roberts, J. M., Washenfelder, R. A., Brown, S. S., Ren, X., Tsai, C., Stutz, J., Brune, W. H., Browne, E. C., Wooldridge, P. J., Graham, A. R., Weber, R., Goldstein, A. H., Dusanter, S., Griffith, S. M., Stevens, P. S., Lefer, B. L., and Cohen, R. C.: An Atmospheric Constraint on the No2 Dependence of Daytime near-Surface Nitrous Acid (Hono), Environ. Sci.Technol., 49, 12774-12781, doi:10.1021/acs.est.5b02511, 2015.

Zheng, G. J., Duan, F. K., Su, H., Ma, Y. L., Cheng, Y., Zheng, B., Zhang, Q., Huang, T., Kimot, T., Chang, D., Poschl, U., Cheng, Y. F., and He, K. B.: Exploring the severe winter haze in Beijing: the impact of synoptic weather, regional transport and heterogeneous reactions. Atmos. Chem. Phys., 15(6), 2969-2983 doi:10.5194/acp-15-2969-2015, 2015.

SC17. Line 479: NO2 is not formed from the reaction of NO with OH. Please correct.

We have reworded this sentence as following:
"NO_2 increased from 10:00 which formed from NO consumed by O_3 and slightly decreased from 14:00 to 18:00 corresponding to the photolysis of NO_2 and the formation of nitric acid during afternoon."

SC18. Line 486-489: These comments regarding the inorganic species can be constrained (at least somewhat) with the appropriate analysis. I recommend either performing this analysis, or removing some of this from the paper as the major point of the paper is the analysis of the organic aerosol.

After careful consideration, we have removed this part.

SC19. Line 533: If the HOA and BC are thought to be mainly from the same source than why is the HOA peak in the evening small than the morning while the BC ones are about equal?

We agree that BC was not only from traffic emission. The source of BC could be originated and/or associated with different sources such as biomass burning, coal combustion, traffic emission etc. In our previous study (Xu et al., 2014), we found ~50% of BC was associated with traffic emission during 2012 summer. During wintertime in Lanzhou, the temporal variation of BC had tight correlation with HOA (R^2 = 0.71), BBOA (R^2 = 0.56), and CCOA (R^2 = 0.56). The higher evening peak of BC could be related with the higher evening peak of BBOA and CCOA. In the updated manuscript, we emphasis the morning and evening peaks of BC were contributed from multiple sources.

SC20. Lines 597-598: There are so many points in Fig. S10 that I cannot clearly see at all any time of day dependence. Please consider some sort of data reduction to make this clear. For Fig. S10, where do the COA and HOA lines come from and why does HOA fall on the COA line in panel B? It would also be helpful to see the plot for the CxHy+ ions at m/z 55 and 57 as well.

Fig. S10 has been revised to be clearly. We change the $C_xH_yO^+$ ions plot to the $C_xH_y^+$ ions plot which is used to diagnose the PMF results of COA and HOA. The reference for HOA and COA lines has been added.

SC21. Line 638: The CCOA is not "high and left." This was pointed about by Referee 2 as well. Please fix.

We have reworded this sentence as following:
"The CCOA also locates in a lower left position in the triangle plot defined by Ng et al. (2010) (Fig. 9a)."

SC22. Line 660: Please correct the spelling of Jimenez.

Done.

SC23. Line 673-674: "These results indicate that the atmospheric oxidation capacity during the winter was still very strong." This statement is very vague (what precisely is meant by "very strong"?) and I think somewhat strong given that there is no exploration of the oxidative budget. Please reconsider.

We have reworded this sentence as following:
"These results indicate that the atmospheric oxidation capacity during winter was still somewhat strong."

SC24. Line 716-717: It is not at all clear to me that a correlation exists for POA less than 15 ug/m3. At 15 ug/m3 SOA varies between ~4 and 12 ug/m3!

Sorry for that we didn't make it clear here. In this sentence, we try to emphasize the consistent variation of POA and SOA during a low PM loading period. As shown in the figure, the points were relative tightly correlated below ~15 μg m^{-3}, while they were very scatted above this concentration value. Because there are too many points, the mass concentrations of points have a wide range. This sentence has been reworded as following:

"It is clear that POA and SOA show relative tight correlation during the periods of POA less than ~15 μg m^{-3} associated with low mass fractions of OA."

SC25. Lines 798-803: This part still seems rather speculative (as pointed out by Referee 2). I do not know what we are really learning from this discussion/comparison since there is no analysis of gas-phase organics in the present manuscript.

In this part, we try to get insight of the formation mechanism of SOA based on the results of PMF and carbon isotope analyses. Although the gas-phase organics were not measured during this study, the content in this part could shine some lights for understanding the sources of SOA which is useful for further study in this region. In this case, we have kept this part in the manuscript, but we also modified some sentences to make it clear and avoid over-interpretation.

SC26. Lines 824-826: I encourage the authors to remember that the boundary layer explanation is still rather speculative since simultaneous measurements of the boundary layer or vertical structure do not exist. Even in Fig. S8 only early morning and evening temperature profiles are shown and no data is given for noon.

We have reworded this sentence as following:

"Further analysis indicated that the first peak was resulted from the contribution of multiple combustion sources and could be related with the variations of the boundary layer heights during morning which accumulated the air pollutants from early morning and until the break-up at around noon time (such influences should be further verified in the future with simultaneous measurements from boundary layer heights).

SC27. Table 2 & multiple places in the text): Please be consistent and use either r or r2.

This has been consistent to R^2 in the updated manuscript.

SC28. Fig 1: I find this very difficult to read due to the small size and the large amount of information. Please consider making more figures. It would also be useful to see the wind direction as a wind rose.

Sorry for it is unclear. Because this is a combo plot which is very helpful to compare the data with each other, we think it is better to keep this figure as a whole, but we also try to adjust it to make it more clear. In addition, we add a wind rose plot in Fig. S1.

SC29. Fig 5: How is there 50 ppbv of NO at night and still 10 ppbv of O3? I would have thought that O3 would have been titrated away.

We noticed this issue, but don't know its exact reason. These data was provided and downloaded from the MEP-China station. Although the value is somewhat strange, we think the diurnal variations should be correct.

SC30. Fig. 7: Please use the standard AMS colors for the ion families. Please also identify them as ions by including “+” in each name. Also, the y-axis labels are overlapping and difficult to read.

We add "+" in each family ion name and the display of this figure are more clear now. We keep the color for each family ion. Although there are default colors in the PIKA panel for each family ion, different AMS group has applied different color (Hu et al., 2016). The ion colors used in this manuscript have been applied in many published AMS paper.

SC31. Fig. 8: I am unsure how useful this figure is given that most (if not all) of this data already appears in Fig. 4.

Thanks for your suggestion. After careful consideration, we keep this figure as the diurnal contributions of each factor in this figure are frequently used in section 3.4. The diurnal contribution of each factor is important for understanding the source variation during the different time of day.

SC32. Fig. 9: What is the R2 value representing? A value of 0.46 seems unrealistically high for any sort of line in panel A. Is OOA supposed to be SOA?

R^2 represents the correlation between POA and SOA in this figure. OOA is a surrogate of SOA, and in the updated figure, OOA has been changed to SOA. After using the new PMF results, the R^2 is 0.1 now.

SC33. Fig 13: Would a van Krevelen plot be more meaningful? Also, please update LV-OOA and SV-OOA to their correct names. Also Fig. 13 is referenced in the text before figures 9-12 are referenced. Please reorder the figures to be referenced in order.

Agree. We apply a Krevelen plot in the updated Fig. 14 and use the slope of H:C versus O:C of AMS data to explain the formation mechanisms of organic aerosol. The terms of OOA factors have been updated. In addition, the order of figures has been updated too.

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