The authors appreciate the two reviewers for their constructive comments and suggestions. The manuscript has been revised accordingly. Our point-by-point responses to these comments are provided below. The comments of the reviewers are printed in black italics and our responses following each comment in blue.

Response to Reviewer1

The manuscript by Xu et al. has a comprehensive characterization of wintertime sub-micron aerosols in Lanzhou, a highly polluted city in western China. The sources of organic aerosols were investigated using positive matrix factorization and radiocarbon analysis. The results were also compared with those measured in summer. This is an important study by providing scientific data and improving our knowledge on aerosol chemistry in western China. This manuscript fits within the scope of ACP, and I recommend it for publication after addressing the following minor comments.

Comments:
1. Please consider the usage of SV-OOA and LV-OOA since this study did not measure the OA volatilities.
We have changed the SV-OOA and LV-OOA to LO-OOA and MO-OOA, respectively.

2. As shown in Figure 2, POA showed an elevated contribution to PM at high mass loadings. Is this due to cooking OA, HOA or BBOA?
Yes, the contribution of each POA factor during high mass loadings was discussed in section3.5 (Fig. 10). The results show that COA is a major contributor to the increased mass loading.

3. Line 41-43: please show the range for the changes in fractions of OA, nitrate and sulfate.
Done.

4. Line 46-47: frequently calm and stagnant air conditions during wintertime in Lanzhou.
Done.

5. Line 51: present the O/C value during summer 2012.
Done.

6. Line 60: haze to air pollution events.
Done.

7. Line 61-62: remove "The primary OA sources were more complex during winter than during summer".
Done.

8. Line 84-86: please add one or more citation.
Done.

9. Line 90-93: remove this sentence because of the duplication information with line 85.
Done.

10. Line 99: insert "recent" before "investigated:::"
Done.

11. Line 119: add receptor model after (CMB)
Done.

12. Line 122-125: add the citation.
Done.

13. Line 142-146: combine these two sentences into one sentence.
Done.

14. Line 155: remove the comma after HR-ToF-AMS and add "of" after characteristics
Done.

15. Line 160: change basin to valley
Done.

16. Line 161-164: rephrase these two sentences. Two "thus" have been used which is a little bit confused.
Done.

17. Line 180: add "on average" before 0.82
Done.

18. Line 190: RH appears at the above content.
Done.

19. Line 200: instruments.
Done.

20. Line 246: consisted to consisting.
Done

21. Line 270-271: where is this number (2.14) from, please add the citation.
This sentence has been rewritten and the number has been deleted. The concentration of BC was calculated following the recommend parameters by the manufacturer.

22. Line 275: add the dry the aerosol in the sentence.
Done.

23. Line 310: show the reason to remove the period of Jan.22-23.
Please reference on the response on Reviewer #2.

24. Line 315-317: how about seven solution results, could BBOA be separated?
As can be seen from the following figure, we didn't find clear evidence that BBOA was separated in the seven solution (Fig. R1).

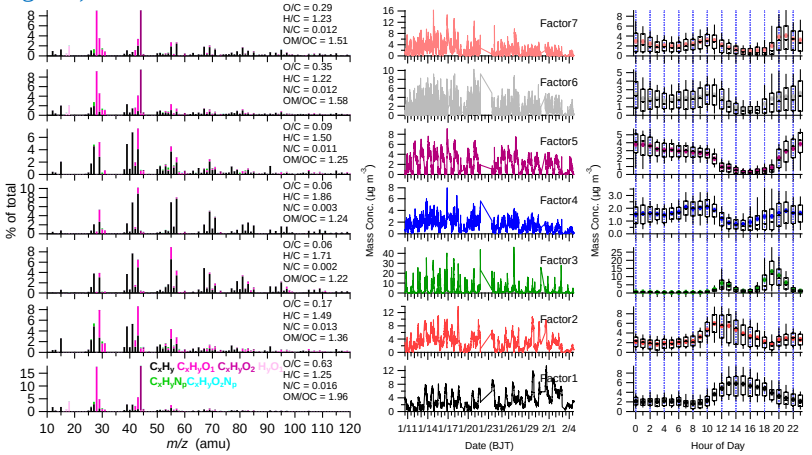

Fig. R1 Seven factors solution analyzed by PMF

25. Line 373-374: there is no wind data in Figure 1, please add them.
Done.

26. Line 412: please show the wind data.

Done.

27. Line 422: primary OA to POA.
Done.

28. Line 443-445: please add the citation.
Done.

29. Line 459: change suggest to could result from
Done

30. Line 485-488: add "from the diurnal variation".
Done.

31. Line 498: is to was.
Done.

32. Line 502-504: Please add the diurnal variation of O/C in summer 2012.
Done.

33. Line 543: add "for all the PMF factor".
Done.
34. Line 544: change "in the past" to "in the recent".
Done.
35. Line 645: add "a little" before "more".
Done.
36. Line 653: change "have been" to "suggest".
Done.

Response to Reviewer2

Summary
This manuscript summarizes measurements of aerosol composition, concentration, and size distributions carried out during wintertime in Lanzhou, China. A large majority of the data is derived from HR-ToF-AMS measurements, although data from an aethalometer, a SP2, a SMPS, and a TEOM are also reported in the manuscript. While the data are interesting and of high quality, there are several aspects of the interpretation of the results that will need to be improved before publication. These issues are discussed in the comments below. In addition, I found the manuscript long, and more importantly, unfocused and encourage the authors to revise the manuscript in order to highlight their most important findings.
Thank you very much for your positive comments. The manuscript is indeed a little bit long as most of AMS paper did. In this manuscript, we mainly focus on the winter-time PM_1 chemical composition, processes and sources in Lanzhou. The most important findings in this study include, firstly, the chemical composition and diurnal patterns of PM_1 species showed difference with those of summer. Secondly, the sources of organic aerosol (OA) were more complex during winter and the primary OA including HOA, BBOA, COA and CCOA was the major contributor during high air pollution. Finally, based on carbon isotopic analyses, we evaluated the contribution of fossil and modern carbon to primary and secondary organic carbon which are important to understand the source and chemical evolution of OA. In order to clarify the findings in this manuscript, we rephrase the conclusion section in the updated manuscript and emphasize on these three points.

General comments

Introduction:

There are two sections of the introduction which should be better referenced.

These sections are listed below.

- *Lines75 –77: references are needed that describe the implementation of the new air quality improvement strategies mentioned in the text.*

We add a reference in this sentence.

- *Lines 84 –93: references are needed to support the description of past air pollution issues in Lanzhou, especially for illustrating the sources of pollution as well as the meteorology discussed by the authors.*

Done.

Presentation:

Overall the quality of the writing is adequate, but copy-editing will be necessary to correct grammar and syntax errors.

Positive matrix factorization (PMF) analyses: Why were the highly polluted periods removed from the PMF analysis? More justification is needed to explain why this step was taken. It is concerning to see a dataset altered in such a way before a statistical analysis since that will add subjective bias. Were the highly polluted episodes removed for both the unconstrained and the constrained analyses or just for the unconstrained analyses with PMF2.exe?

The highly polluted period was removed during both constrained and unconstrained analyses, mainly based on the three points below. First, ion fitting in this period is bad. The reconstructed time series of organics during this period was offset from the measured time series and the residual is significant. Second, the mass spectra and diurnal pattern of PMF factor were influenced by this high polluted period. The mass spectra of PMF factor were more reasonable when removed this period which were more comparable with that during summer study such as HOA, COA and LO-OOA. The standard deviation of diurnal pattern on each time point of PMF factors were bigger than those after removing this period.

In addition, the classification of the CCOA factor is not convincing. The correlation of this factor with the various tracers measured is very similar to HOA. In addition, there were no tracers measured that are specific to coal emissions for validating the assignment of this factor to coal combustion. The fact that this factor has a mass spectrum similar to HOA with a larger contribution at m/z 44 suggests that the factor may represent instead "aged" HOA.

Response: The CCOA factor has been found in several studies in China during winter-time study (Hu et al., 2013; Elser et al., 2016; Sun et al., 2016), as in north China, coal is a major energy and heating source during wintertime that is different from developed countries. The major feature in its mass spectrum is the increased signals at m/z 91 and 115 as observed in laboratory study (Dall'Osto et al., 2013), and the high contribution at m/z 44 is related to the production of organic acids during coal combustion (Zhang et al., 2008). These spectral features were indeed observed in this study, which is similar to other observed CCOA factors in China (Hu et al., 2013; Sun et al., 2016). In addition, although the mass spectra of HOA and CCOA are similar below m/z 120, they are significant different above m/z 120, and many of them are found to be PAH-related ions (Hu et al., 2013; Sun et al., 2016).

^{14}C analysis: Only four 24 h samples were collected. Therefore, the conclusions drawn from the ^{14}C analysis may not be representative and are highly dependent on the choice of days sampled. The ^{14}C analysis is difficult to perform, so it is understandable that only a small number of samples can be analyzed, but the manuscript must include some discussion of how aerosol properties during the four days with ^{14}C data compare to the rest of the measurement period.

Response: The sentence below has been added in the updated manuscript (section 2.3.3): "Here, we use the results of these four filter samples to represent the average situation of the field sampling. During the field study period, the air mass and aerosol source are pretty stable which mainly originated from regional sources as illustrated from the consistent variations of chemical composition (section 3.1.3). This can also be evidenced from the relative calm meteorological conditions during the whole sampling period (section 3.1.1)."

Correlation of OOA and Odd-Oxygen (OX): There are several aspects of this analysis (on lines 800 to 822) that are seriously flawed. First, only the correlation between LV-OOA and OX has been plotted in the manuscript. It is necessary to include similar plots of SV-OOA versus OX as well as (SV-OOA + LV-OOA) versus OX. Otherwise the conclusions drawn from this type of analysis regarding SOA sources are not valid. Second, it is suggested that "aromatic VOCs may be a large contributor to SOA formation". It is not clear how such a conclusion can be made based on the slope of the LV-OOA to OX plot alone, and it is impossible to draw such conclusions regarding the contributions of compounds to SOA formation without a detailed analysis of product yields and volatilities. Third, the suggestion that sources that emit modern carbon may emit more aromatic VOCs than traffic and coal combustion together is speculative. It is difficult to follow the logic behind this suggestion, but it seems to be based on the unjustified assumption that aromatic VOCs are the dominant SOA precursors at this site, which completely ignores primary semi- and intermediate volatility organic compounds (P-S/IVOCs) that are likely to be important contributors to SOA formation [e.g. Robinson et al., 2007].

Response: We did not include LO-OOA (named SV-OOA in the original manuscript) in the correlation of OOA and Ox owning to the different synchronization of LO-OOA and Ox (Fig. R2). It seems LO-OOA varied two to three hours earlier than Ox possibly due to other origination for LO-OOA such as down mixing of mixing-layer aerosol, which is a popular phenomenon in the mountain-valley city (Chen et al., 2009). We add this content in the updated manuscript (section 3.7). MO-OOA was suggested to be mainly from photochemical processes under the low RH and air temperature conditions. The different slope between OOA and Ox had been suggested to result from the different VOCs precursor in the photochemical process. In the manuscript, we didn't consider the volatility of VOCs which P-S/IVOCs can also be aromatic. This conclusion is indeed too strong only based on this slope. For the origination of modern SOC, we remove these contents in the updated manuscript.

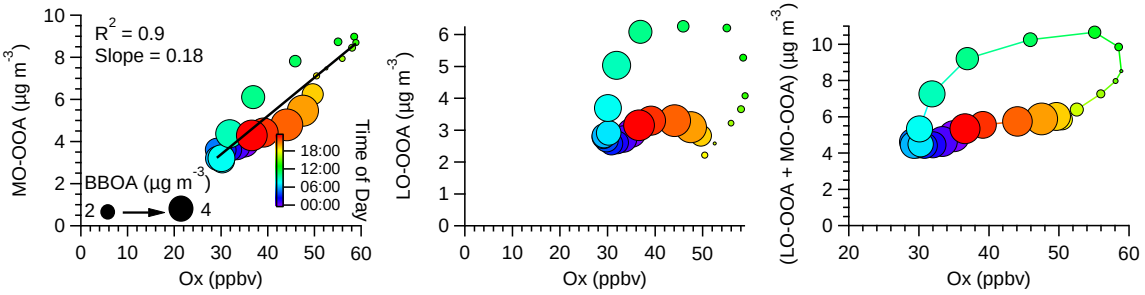

Fig. R2 The scatter plots for MO-OOA, LO-OOA, and (LO-OOA + MO-OOA) versus Ox.

Specific Comments

Lines 109 –112: This sentence is confusing the authors should re-word the sentence to make its meaning clearer.

Response: The sentence has been revised.

Line 132 –134: I agree that the formation and evolution mechanisms of secondary species are poorly understood, but more than one reference should be provided to support this statement. There is a large

body of published work that discusses the lack of agreement between various atmospheric models and measurements of secondary aerosol species. More of this work should be cited here.
Response: Thanks. The sentence has been revised to "However, the formation and evolution mechanisms of those secondary species were poorly understood which previous models tended to underestimate the secondary species budget in polluted regions (e.g., Volkamer et al., 2006)".

Line 134 –136: More details, including references, should be given regarding the possible "fast chemical reactions" proposed in the manuscript. At the moment, these reactions are not described and seem rather mysterious.
Response: This sentence has been deleted.

Line 138 –142: I don't agree with the phrase "the most advanced". While I do think the development of new online mass spectrometry techniques including the AMS has been very valuable, I disagree strongly with the absolute nature of this statement. One could, for example, list a variety of recently developed instruments based on optical techniques that have also been valuable.
Response: Agree. The sentence has been revised to "…appear to be advance on probing…".

Lines 274 –276: I'm not very familiar with TEOM instruments, but it seems strange that the instrument was operated at 40℃ to "minimize mass loss due to volatilization of semi-volatile aerosol compounds". This temperature is warmer than room temperature and would likely cause the evaporation of ammonium nitrate and organics.
Response: The TEOM was normally operated at 50°C to remove the water vapor. In this study, we changed the inlet temperature to 40°C to minimize the loss of semi-volatile species. The sentence has been revised to "The TEOM was operated at a temperature of 40 °C other than normal operation condition (50 °C) in order to minimize mass loss due to volatilization of semi-volatile aerosol compounds".

Lines 423 –425: The similar size distributions for inorganics and SOA indicate that the aerosols are internally mixed rather than externally mixed. This statement should be corrected, as well as similar statements that are made in the abstract and the conclusions.
Response: Revised.

Lines 478 –479: To my eye, it appears that the concentration of NO2 stays the same or even increases slightly after 14:00 rather than decreasing.
Response: This sentence has been revised to "NO_2 increased from 10:00 which formed from NO consumed by OH radical and slightly decreased from 14:00 to 18:00 corresponding to the formation of nitrate and O_3 during afternoon."

Lines 578 –589: The entire discussion of BBOA aging in this paragraph is not well-supported by the measurements presented in the manuscript. It is suggested that the BBOA factor is due to oxidation of gas phase emissions. However, the measured O/C ratio for BBOA is consistent with unoxidized primary emissions [Ortega et al., 2013]. In addition, the relatively large particle sizes could be due to internal mixing or coagulation of particles.
Response: Thanks for your suggestion. The content of BBOA aging has been deleted and the updated manuscript only focused on the primary feature of BBOA.

Lines 680–682: The pertinent figure for this sentence is 13b and not 13a. Also, it is not correct to say that the SV-OOA component is situated in the upper left corner. In fact, this component is positioned rather low in the two triangle diagrams.
Response: Revised.

Lines 698 –703: It is difficult to understand the significance of the discussion on these lines, which seems to mostly review well-known facts regarding secondary aerosol formation. A stronger connection should be made to the OOA data or these lines should be deleted.
Response: The sentence has been deleted.

Line 734: I think this percentage should be 59%.
Response: Revised.

Lines 736–740: This sentence is very speculative. It should either be deleted or supported by measurements of actinic flux and OH radicals.
Response: The speculative part of the sentence has been deleted.

Lines 748 –751: Why would emissions of COA decrease during the holiday? Presumably people still cook during the Chinese New Year.
Response: During the Chinese New Year holiday (up to four weeks), many people (about half of the population of Lanzhou) would leave for their hometown, and most of the restaurants in the city were closed. For example, the students' canteen in Lanzhou University had been closed. A sentence has been added in this part to explain this reason.

Line 771: The values for SOC in the text are different from those displayed in Figure 12.
Response: The values in the text have been revised.

Lines 853 –855: Please delete the term "VOC" from this sentence. SOA could also be formed from lower volatility P-S/IVOCs and 14C measurements do not provide any information regarding the amount SOA specifically formed from VOCs.
Response: Done.

Figures1 & 2: I assume that the BC measurements in these figures come from the aethalometer rather than the SP2. Nevertheless, the instrument that was used should be specified in the figure caption.
Response: The information for instrument of BC has been added in the captions.

Figure 3: It would be very interesting to include in this figure the rBC size distribution measured using the SP2.
Response: Unfortunately, we did not process the data of SP2.

Figure s1a: The text in the map is very small and difficult to read.
Response: The size of text has been revised.

Reference:

Chen, Y., Zhao, C., Zhang, Q., Deng, Z., Huang, M., and Ma, X.: Aircraft study of Mountain Chimney Effect of Beijing, China, J. Geophys. Res., 114, D08306, 10.1029/2008JD010610, 2009.

Dall'Osto, M., Ovadnevaite, J., Ceburnis, D., Martin, D., Healy, R. M., O'Connor, I. P., Kourtchev, I., Sodeau, J. R., Wenger, J. C., and O'Dowd, C.: Characterization of Urban Aerosol in Cork City (Ireland) Using Aerosol Mass Spectrometry, Atmos Chem Phys, 13, 4997-5015, 10.5194/acp-13-4997-2013, 2013.

Elser, M., Huang, R. J., Wolf, R., Slowik, J. G., Wang, Q., Canonaco, F., Li, G., Bozzetti, C., Daellenbach, K. R., Huang, Y., Zhang, R., Li, Z., Cao, J., Baltensperger, U., El-Haddad, I., and Prévôt, A. S. H.: New Insights into Pm2.5 Chemical Composition and Sources in Two Major Cities in China During Extreme Haze Events Using Aerosol Mass Spectrometry, Atmos Chem Phys, 16, 3207-3225, 10.5194/acp-16-3207-2016, 2016.

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.

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Zhang, Y., Schauer, J. J., Zhang, Y., Zeng, L., Wei, Y., Liu, Y., and Shao, M.: Characteristics of Particulate Carbon Emissions from Real-World Chinese Coal Combustion, Environmental Science & Technology, 42, 5068-5073, 10.1021/es7022576, 2008.