The authors appreciate the three 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 italics and our responses following each comment in bold.

Response to reviewer #1

General Comments:

The mass concentrations, chemical composition, and size of submicron particles were measured in Lanzhou, China during the summer of 2012, with the conclusions that BC & HOA from traffic, COA from cooking, LV-OOA/sulfate from photochemical processing, and SV-OOA/nitrate (with some BC) from photochemistry/boundary layer mixing are the major sources of submicron PM. Overall, this paper offers a very thorough analysis of a large and complex dataset, with results that could be useful for developing pollution mitigation strategies. While generally sound, there are a few technical points that should be addressed.
Recommendation: Publish after revisions.

We thank the reviewer for his/her positive comments.

Major Comments:

Section 2.1.2 HR-ToF_AMS Operation: If HEPA-filtered air is used to constrain gaseous CO2 concentrations, HEPA-filtered measurement periods are recommended with much more frequency than shown here (see Jimenez wiki:http://cires.colorado.edu/jimenez-group/wiki/index.php/Field_ToF-AMS_Operation). Ambient CO2(g) concentrations vary, and particulate Org concentrations will be incorrect during times when ambient [CO2(g)] varies from the HEPA measurement period. Also, as outlined in AMS Manual (Pp. 40), the IE calibration should be done every 3-4 days, or each week at maximum, and certainly at the end of the campaign. Yes, the airbeam correction can correct for some fluctuation in ionization efficiency, but the infrequency of calibrations here may impact the quantitativeness of this dataset. There is no remedy for the infrequency of IE calibrations, but it might be advisable to estimate at least the error in Organic quantitation arising from [CO2(g)] uncertainty; for instance, are there any nearby continuous[CO2(g)] measurements? The standard deviation might be used to comment on a general magnitude of error in particulate CO2 quantitaion (also affecting f44, etc.). In all probability this error will not outstrip total AMS quantitation error, but it would be thorough to evaluate this issue. It is stated early and frequently in the paper that the 10:00-13:00 diurnal increase in organics is due to 'downward mixing of a residual layer aloft,' but the temporal progression from a morning HOA maximum to a _10:00 SV-OOA maximum, to a noon-14:00 maximum in LV-OOA suggests photo-oxidative succession. The relatively late maximum in O/C also suggests that organic (photo-) oxidation is occurring, as is acknowledged in Sections 3.5. The nitrate 10:00 maximum does support the downward mixing idea as explained in the text, and that certainly could contribute to the organic diurnal profile as well, but it seems that photochemical SOA formation should be presented earlier and more strongly as a possibility; readers often read abstract, some results, and conclusions, so they may miss the discussions about photochemistry. There is a lot of repeated material; though some repetition is of course inevitable in an analysis involving this many interrelated species/datasets, this paper would benefit greatly from more streamlined introduction of material. For instance, H/C will track the mass fraction of HOA, etc. so you can mention that those are related but don't necessarily need two large paragraphs on each, since they are telling the same story about the nature of the aerosol. Lastly, there is a general pattern of incorrect grammar (mostly in the abstract/

introduction) – often involving incorrect subject reference - that should be remedied by professional editing; some corrections will be provided in the 'Technical Comments' sections, but further proofing is recommended.

We agree with the reviewer that conducting frequent filtered air measurements and IE calibrations during the period of field study is useful and we will try to do so in the future. However, for this study, we conducted IE calibration twice on different days and obtained very similar IE_{NO3}/AB (i.e., IE_{NO3} normalized by air beam signal recorded at the time of the IE calibration). This indicates that the AMS performance was stable during this study and more frequent IE calibration was not necessary.

In terms of applying variable gas phase-CO_2 correction, we do not have ambient CO_2 measurement data nearby our site. We therefore estimate the uncertainty in fCO_2^+ based on typical variation of CO_2 mixing ratio in other urban areas. According to Grimmond et al. (2002), urban CO_2 concentration usually varies in the range of 350–500 ppm, which corresponds to organic-equivalent concentration of CO_2^+ 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 (Allan et al., 2004) and subtracted from measured CO_2^+ signal to determine Org_{CO2} (or Org44). Given that the average Org_{CO2} of this study is ~ 1.1 org-eq μg m^{-3}, we estimate that applying a constant gas phase CO_2 subtraction may introduce ~ -3 – 5% uncertainty in fCO_2^+, which is very small. This point is now mentioned in the updated manuscript.

Solar radiation is still relatively weak between 8 - 10 am BJT, which is 7 - 9 am local time. Thus, photooxidation is unlikely an important factor for the sharp increase of SV-OOA during that time period (Fig. R1b). The mechanism of the morning SV-OOA increase is likely similar to that of nitrate which is related with the downward mixing of residual layer.

The description of diurnal variation of OA has been integrated in Section 3.6 as shown following:
"Diurnal changes of the mass fraction of HOA (Fig. 10d) presented an afternoon minimum (less than 5% of OA mass from 12:00 to 18:00), and a morning maximum (more than 20% of OA mass from 0:00 to 8:00), which was due to the photooxidation consumption of fresh aerosol and emissions from traffic activities including late night diesel truck traffic and morning rush hours. This feature can further be supported by the diurnal variations of the elemental ratios (O/C and H/C) of OA (Fig. 10a) that H/C peaked at morning rush hour and evening rush hour/dinnertime, while O/C was particularly elevated in the afternoon."

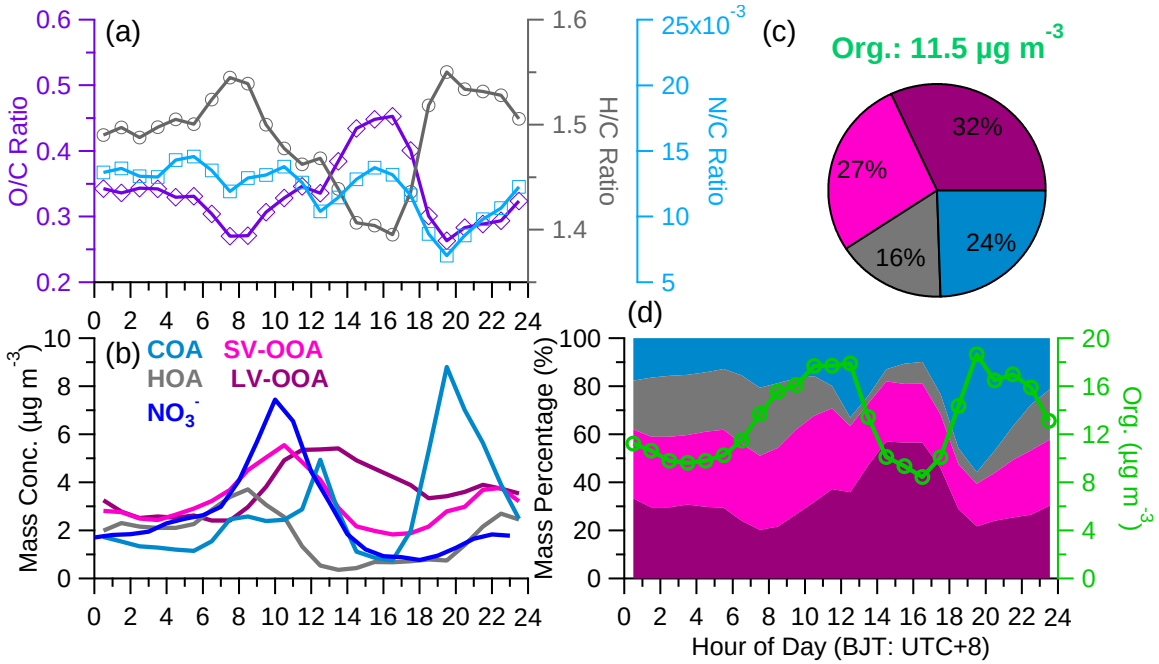

Fig. R1. Diurnal patterns of (a) O/C, H/C, N/C ratios and (b) the mass concentrations of the OA factors; (c) average mass contributions of the four OA factors to the total OA (pie chart) and (d) diurnal cycles of the mass percentages of the four OA factor and the total OA mass concentration.

Specific Comments:

Page 16189, Line 24 through Page 16190, Line 14: This whole paragraph could be omitted for brevity; the same information was summarized wonderfully in the numbered list in the previous paragraph.

Agree. The second paragraph has been removed.

Section 2.1.1 Sampling Sites: Recent studies indicate that PM concentration gradients near roads can be very extreme (e.g. Hagler et al., 2009). These sites appear to be placed approximately the same distance from the road (fig 2), but might prevailing meteorology (if it has any directionality) and/or roadside barriers make these sites have different levels of road influence? Since the BC instrument was not truly co-located with the AMS and you use AMS data to comment on BC sources, I recommend addressing this issue, if briefly.

The local environments surrounded for these two sites were indeed different from each other. We mention this kind of potential uncertainty on BC estimation in the updated manuscript (Section 3.7) as shown below:
"Note that the different local environment for BC and AMS measurements could generate a certain uncertainty for the variation of BC and AMS datasets."

Page 16199, Line 14-15: Specify/remind readers that this relatively lower PM in Lanzhou during summer is only for one year; multi-year analysis ensuring that 2012 is not anomalous would be necessary to leave the statement as-is.

We have rewritten this sentence and specify that these results of this study are only for one year.

Page 16200, Line 14-16: There are also time periods (fig. 4b) where the slope <1; non-sphericity (esp. of soot-like particles) can cause this discrepancy – see Zhang et al. (2005). It might be worth constructing average size distributions during the highest discrepancy periods for both SMPS and AMS and seeing whether those support your hypothesis that partial transmission of supermicron particles in the AMS is responsible for the disagreement (_PM1.5 transmission efficiencies are only _0.05 (Liu et al., 2007).

We checked the data during the period with the slope less than 1, and found that the concentration of black carbon increased significantly and the size distribution of NR-PM_1 from AMS shifted to small size range (Fig. R2a), however the size distribution from SMPS shifted to the larger size range, which is consistent with particles being fractal. For the period with the slope above 1, the chemical composition of PM_1 showed an increased contribution of nitrate (Fig. R2b), which could lead to a high collection efficiency of particle (Middlebrook et al., 2012). In addition, we checked the ratio between particle mass in the size range of 800–1200 nm and that in the size range of 40–800 nm and found a ratio of ~0.1 (the size at 800 nm of AMS corresponding to the SMPS at 650 nm). We added these discussions in the updated manuscript (Section 3.2) as shown below:
"This relatively large discrepancy, which was observed previously (Ge et al., 2012b), was likely caused by the different response between these two instruments in their maximum transmission sizes (e.g., the AMS has partial transmission for particles up to ~1.0 μm whereas the upper cut off size of the SMPS is only ~650 nm) and the variation of CE of AMS depending on the chemical composition of particle."

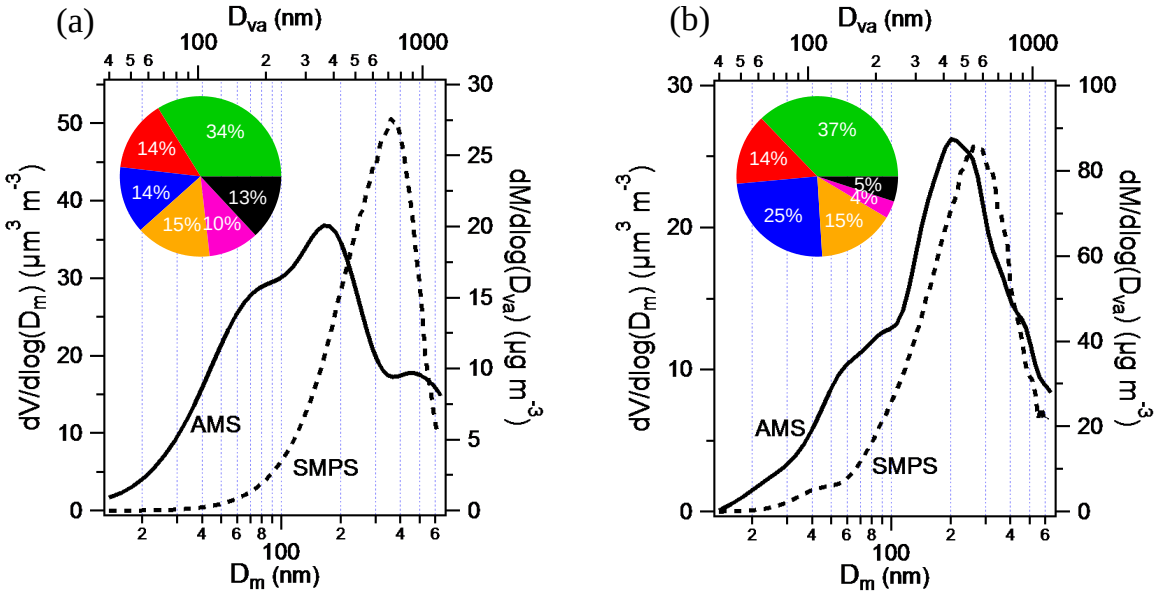

Fig. R2. Comparisons of size distribution (measured by AMS and SMPS) and chemical composition during the different periods with the slope of PM_1 mass concentrations vs. SMPS particle volumes of (a) lower than 1 (b) higher than 1.

Page 16202, Line 23-24: I guess I don't see clear evidence of ammonium sulfate here. Restate? Is there evidence in the timeline, etc. of periods with clear ammonium sulfate in the morning? It looks more like ammonium chloride, if anything. Check the stoichiometry – is there ammonium in excess of balance with sulfate and nitrate in the morning?

Agree. It was ammonium chloride during the morning. The diurnal pattern of the excess ammonium balancing from sulfate and nitrate was similar to that of chloride peaking at early morning. The sentence has been revised in the manuscript.

Page 16206, Line 25-27: (1) most would not consider 0.46 a 'high' correlation, and (2) relationships between fresh emissions and SV-OOA could also arise from condensation of SVOCs on the newly-available POA surface area.

The correlation efficiency (r =0.68 and r^2 = 0.46) between HOA and SV-OOA time series is the highest among these four OA factors, and the value of 0.68 is reasonable high. The relatively high correlation between these two factors could arise from the condensation of SVOCs on POA as well as the transport of mixed air mass, which has been emphasized in the original manuscript. This sentence has been revised in the updated manuscript as following:
"Another reason for relatively oxidized HOA was likely due to the fact that a fraction of the HOA mass was likely transported from aged OA from regional source and/or the condensation of SV-OOA on HOA as suggested by the significant positive correlation (r^2 = 0.46) between time series of HOA and SV-OOA (Fig. S5d)."

Page 16210, Line 8-10: SV formation through in-situ photo-oxidation is also likely and should be mentioned.

As described above, photooxidation is unlikely a significant mechanism for the observed morning increase of SV-OOA.

Page 16210, Line 14-18: This sentence is confusing, not strictly accurate, grammatically incorrect, and could be deleted. The better correlation between SOA and nitrate+ sulfate (than between SV-/LV- and the inorganics individually) does not necessarily indicate the 'same' processing (you have already argued that LV/sulfate and SV/nitrate processing are different) – it could reflect the increased precision of organic reconstruction when removing the somewhat arbitrary mass division between SV- and LV-OOA in PMF. I do not mean to imply that such divisions in PMF not meaningful, but rather emphasize that PMF factors are static representations of average composition patterns.

The sentence has been deleted.

Page 16210, Line 25-: Might be good to emphasize that these are only for one year, and not contemporaneous. The wording may imply that these are statistical comparisons, but these data could be outliers, as they are each from a single year, and are non-contemporaneous.

The sampling date of each study has been added.

Page 16211, Line 8: Citation/name for the linear decomposition algorithm?

Done.

Technical Comments:
Page- and Line-referenced comments on specific points of grammar and style.

*** indicates additions.*
{ } indicates deletions.
[] indicates referee notes on grammar.
"Recommend: : :" indicates a style/preference comment, while "Replace/Change" indicates a real grammatical error.

Abstract/Introduction Comments:

Page 16188, beginning Line 10: change "The organics was consisted of: : :" to "Organic matter was comprised of: : :" or "Organics consisted of: : :"

Done.

*Page 16188, beginning Line 13: ": : :(PMF) of the high-resolution *organic* mass spectra {of organic aerosols (OA)} identified: : :"*

Done.

Page 16188, Line 24: Recommend replacing "peak" with "maximum"

Done.

Page 16188, Line 24: Replace "mixing down" with "downward mixing"

Done.

*Page 16188, Line 25: "The mass spectrum of SV-OOA was *similar to that of coal combustion aerosol and likely influenced* by coal combustion activities: : :"*

Done.

Page 16189, Line 6: Replace "quick" with "rapid"

Done.

*Page 16189, Line 10: subject reference, correct with, e.g.: "With a population of ~3.6 million (including its surroundings), Lanzhou *is* {has been viewed as} one of the most: : :"*

Done.

Page 16189, Line 14: subject reference, correct with, e.g.: "Generally, poor air quality in Lanzhou is attributed to several important factors:: : :"
Done.

*Page 16189, Line 15, list item (1): "{easily} *often* results in: : :"*

Done.

*Page 16189, Line 22, list item (4): ": : : vehicle *usage* [singular], which *was*: : :"*

Done.

*Page 16190, Line 3: "In the late 1980's, field studies {were conducted to} investigat*ing* the physical structure of *the* atmospheric boundary layer in Lanzhou {and had} led to a better understanding of the transport and diffusion of pollutants in the valley basin and the relationships between air quality and topographically driven circulations (Hu et al., 1989; Chen and Huang, 1993).*

This paragraph has been removed.

Page 16190, Line 15: subject reference

A website address has been added.

Page 16190, Line 17: Recommend replace "." with ";" (and de-capitalize "Another")

Done.

*Page 16190, Line 25: ": : :has *been* reduced: : :"*

Done.

Page 16190, Line 27: Replace "fast" with "rapid"

Done.

Page 16190, Line 23: Replace "In this regard" with, e.g. "To address this, we conducted a measurement campaign in summer 2012: : :"

Done.

Methods Comments:

*Page 16194, Line 15: "Instrumentation details of *the* HR-ToF-AMS are described elsewhere"*

Done.

Page 16194, Line 16: Replace "briefly" with "brief"

Done.

*Page 16194, Line 20: "_6000 *m/Δm* in this study" [include units]*

Done.

Page 16194, Line 24-26: This last sentence is redundant – this information was stated earlier in text.

Deleted.

Sections 2.2.1 and 2.2.2: These sections are very thorough, but most of this is standard practice; it could be condensed if length is of concern.

We are keeping these detailed descriptions because this paper is the first AMS study from the Lanzhou group, and it might be useful as a reference of our future studies/papers.

Results/Discussion Comments:

Page 16198, Line 21: "The concentrations of all PM1 species varied dynamically during this study, with: : :" [insert "," between 'study' and 'with']

Done.

Page 16200, Line 25: Replace "mixing down" with "downward mixing"

Done.

Page 16201, Line 24: "observations at other cities where nitrate usually peaks ["peaks" is plural] during nighttime, during which low temperature and high RH favor {the} nitrate partitioning to the particle phase (e.g., Sun et al., 2011)."

Done.

Page 16202, Line 27: Replace "maintained" with "remained", and replace "till" with "until or 'til"

Done.

*Page 16202, Line 28: "associated with the {similar} *same* boundary layer evolution as {that} for nitrate."*

Done.

Page 16206, Line 15: Replace "reasonable" with "reasonably"

Done.

*Page 16208, Line 4: ": : :during dinner time {situating} *are situated* around the COA {side} *line. *"*

Done.

*Page 16209, beginning Line 5: Replace with, e.g.: "Two sub-types of OOA, {i.e.,} semi-volatile OOA (SV-OOA) and low-volatility OOA (LV-OOA), *are* frequently {determined} *identified* using PMF, and {the} SV-OOA is {usually} less oxidized than the LV-OOA." PMF-derived SVOOA is by definition (not 'usually') less oxidized than low-volatility OOA.*

Done.

Page 16211, Line 18-19: This sentence mixes passive and active tenses; revise.

Done.

*Page 16211, Line 24: "Meanwhile, these results also suggest that organics {was a major species} *were* internally: : :"*

Done.

Page 16212, Line 20: Replace "period" with "periods"

Done.

Page 16212, Line 21: Replace "highly" with "somewhat" or erase entirely

Done.

*Page 16213, Line 11: "which were related to local traffic emission during rush hours, *photooxidation, turbulent mixing, * and cooking emissions during meal times"*

Done.

*Page 16213, Line 15: ": : :of *the* urban boundary layer: : :"*

Done.

Table 2 could be omitted for brevity.

Done.

Comments on Figures:

*Figure 1: Caption, recommend, "*Summary of* AMS measurements in China. In each site, total mass concentration, mass fractions (pie chart), and OA components based on PMF analysis (column) *are shown*. The O/C ratios for each OA component{s} {have} *are* also {been} shown {in} *for* several studies which used HR-ToF-AMS."*

Done.

*Figure 3: Axis text too small (whole figure could be bigger). Caption:: : :(Precip{i}.) : : :SO2 and NO2{of} *from* the two EPA-China stations: : :*

The Figure has been enlarged and the caption has been revised accordingly.

Figure 4: descriptions of a) and b) are switched in caption.

The order of Figure 4a and Figure 4b has been switched.

*Figure 6: "{The box plots} *Diurnal variations* of oxidation ratios of (a) sulfur and (b) nitrogen defined as fS = nSO2-4 /(nSO2-4 +nSO2) and fN = nNO2-3 /(nNO2-3+nNO2). {The data of} SO2 and NO2 are {the} average values {of} *from the* two EPA-China stations in Lanzhou. The cross in each box is the mean value, the line is the median value, the box extends from 25 to 75% percentile, and the sticks are the 10 and 90% percentiles."*

Done.

Figure 7: Replace "distribution" with "distributions"

Done.

Figure 9: The relationships between factors and species are already shown in Table 1, so these timelines are redundant. Please keep the PMF factor mass spectra, though!

Done.

Figure 13: This information is covered in Table 1 and the text, so this figure could be omitted for brevity.

Done.

Figure 14: There is a lot of redundant information here. A) and b) could be omitted; c), d), and Table 1 give the same information.

Figure 14 a–d are the diagnostics plots for our BC decomposition method and we think these plots are necessary to keep for the readers to evaluate our method. We also believe this Figure is a good supplement to Table 1. Thus we would like to keep this Figure.

References:

Hagler, G. S. W., Baldauf, R. W., Thoma, E. D., Long, T. R., Snow, R. F., Kinsey, J. S., : : : Gullett, B. K. (2009). Ultrafine particles near a major roadway in Raleigh, North Carolina: Downwind attenuation and correlation with traffic-related pollutants. Atmospheric Environment, 43(6), 1229–1234. doi:10.1016/j.atmosenv.2008.11.024

Liu, P. S. K., Deng, R., Smith, K. A., Williams, L. R., Jayne, J. T., Canagaratna, M. R., : : : Deshler, T. (2007). Transmission Efficiency of an Aerodynamic Focusing Lens System: Comparison of Model Calculations and Laboratory Measurements for the Aerodyne Aerosol Mass Spectrometer. Aerosol Science and Technology, 41(8), 721–733. doi:10.1080/02786820701422278

Zhang, Q., Worsnop, D. R., Canagaratna, M. R., & Jimenez, J. L. (2005). Hydrocarbonlike and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols. Atmospheric Chemistry and Physics, 5(12), 3289–3311. doi:10.5194/acp-5-3289-2005.

Response to Reviewer #2

This paper presents the PMF analysis of AMS data taken in Lanzhou, China. The procedures employed are pretty standard and there are no really surprising results here, however this is the first paper presenting data of this type in this region, so it is still reasonably novel in that regard. The analysis is largely sound and the paper is well written and presented, so I recommend publication subject to minor comments.

We thank the reviewer for his/her positive comments.

Comments:

General: *When presenting an acronym, the authors use bold on the key letters. This is not standard practice (capitalisation is normally employed) and makes the article look a little informal, so I would recommend writing them in standard weight text.*

Maybe this is an ACPD print issue. We will mention it when resubmitting the manuscript.

16204, Line 15: The nominal chemical formula is essentially repeating the ratios already stated and is potentially misleading because the AMS cannot deliver data on carbon number, so I recommend that it be removed.

The nominal chemical formula shows the average elemental composition of the organic aerosol and the molar ratios among elements in a straightforward way. It was reported in a number of peer-reviewed papers. In response to the reviewer's comment, we have revised the text and it now reads "On average, the OA had an O/C of 0.33, H/C of 1.49, and N/C of 0.011 (Fig. 8b), which yields an average organic mass-to-carbon (OM/OC) ratio of 1.58 and thus has a nominal chemical formula of $C_1H_{1.49}O_{0.33}N_{0.011}$. Note that the nominal formula should not be used to infer the average molecular weight or carbon number in the organic aerosol studied here. "

16208, Line 22: The narrower distribution could also be because the particles are more spherical, or vaporise faster.

The possible reason of more spherical particles is now mentioned.

16211, Line 12: This fit should have been performed with an additional parameter not multiplied by a PMF factor, to represent the average BC mass not accounted for by the PMF factors. Without this, the other factors will not be accurate. Also, were the parameters constrained to be non-negative?

All the parameters during the fitting have been constrained to be non-negative. The BC mass that is not accounted by the PMF factors has been evaluated by the residual between the measured and reconstructed BC mass concentration.

Fig. S2: This is not very clear. Is a colour version available?

The resolution of this Figure has been improved.

Running title: I would revise this to make specific reference to Lanzhou

Thanks. Done.

Response to Reviewer #3

General comments:

This manuscript reports chemical compositions of submicron aerosol particles measured by an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HRToF- AMS) at Lanzhou, northwest China in summer. The sampling site located at an urban area of Lanzhou in which

aerosol compositions were strongly influenced by human activities, photochemistry, and regional transports. The data analysis is performed comprehensively and their observations are explained with reasonably arguments, even though some of them have to be further elaborated (see specific comments). The manuscript attempts to understand size distribution and sources of black carbon based on positive matrix factorization (PMF) analysis of AMS organics and black carbon measurements by multi angle absorption photometers (MAAP). Since a newly developed Aerodyne soot particle AMS (SP-AMS) have been deployed in a few urban and on-road studies, it is suggested to highlight the major uncertainties of their estimation compared to direct black carbon characterizations by SP-AMS (see specific comments). The manuscript fit the scope of Atmospheric Chemistry and Physics but the authors should proof read and check all the figure number, captions and labeling carefully in main text and supplementary material (see technical corrections). 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:

- ***Introduction:*** *1) Page 16189, first and second paragraph: The total length of the two paragraphs can be shortened. The authors summarize the current understanding of air pollution in Lanzhou. The second paragraph is however repeating the details of the first paragraph, which is already a good summary for general readers.*

The second paragraph has been removed.

2) Page 16191, second paragraph: Some of the technical contents regarding the design and working principle of Aerodyne HR-ToF-AMS should be moved to Section 2.1.2.

This content has been moved to Section 2.1.2 and shortened accordingly.

- Section 2.1.2, Page 16195, line 3-5: Particulate-free ambient air (or "filter period") provides information to adjust the fragmentation table of AMS data analysis software. One of the major reasons is to correct the contribution of gaseous CO2 to organic fragments at m/z 44 (CO2+) as discussed in Section 2.2.1. On-road engine emission can be a major source of anthropogenic CO2 in urban and can be varied significantly depending on traffic condition. It is generally recommended to perform "filter period" frequently but this work only did twice at the end of the campaign. Please evaluate the potential uncertainties to the final results (e.g. f44, O/C, total organics, etc.).

We agree with the reviewer that conducting frequent filtered air measurements and IE calibrations during the period of field study is useful and we will try to do so in the future. However, for this study, we conducted IE calibration twice on different days and obtained very similar IE_{NO3}/AB (i.e., IE_{NO3} normalized by air beam signal recorded at the time of the IE calibration). This indicates that the AMS performance was stable during this study and more frequent IE calibration was not necessary.

In terms of applying variable gas phase-CO_2 correction, we do not have ambient CO_2 measurement data nearby our site. We therefore estimate the uncertainty in $f{CO_2}^+$ based on typical variation of CO_2 mixing ratio in other urban areas. According to Grimmond et al. (2002), urban CO_2 concentration usually varies in the range of 350–500 ppm, which corresponds to organic-equivalent concentration of ${CO_2}^+$ 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 (Allan et al., 2004) and subtracted from measured CO_2^+ signal to determine Org_{CO2} (or Org44). Given that the average Org_{CO2} of this study is ~ 1.1 org-eq μg m^{-3}, we estimate that applying a constant gas phase CO_2 subtraction may introduce ~ -3 – 5% uncertainty in fCO_2^+, which is very small. This point is now mentioned in the updated manuscript.

- Page 16198, line 5-6: It seems that cp is a fitting parameter without clear physical meaning as this parameter depends on both time variation and mass loading of each chemical component. Rather, products of tp and cp can be used to evaluate the sources of BC (but not degree of mixing with each component) as shown in Section 3.7 and Figure14.

Agree. c_p is only a fitting parameter. t_p and c_p can only be used to evaluate the sources of BC rather than the mixing state. This sentence has been rewritten as following:
"Where t_{BC} denotes the time series of BC mass concentration, t_p denotes the time series of inorganic species or OA factors p, and c_p denotes the corresponding fitting parameter."

- Page 16199, line 5-7: Figure 3a shows that there were only a few moderate rain events within the sampling period. Without providing other precipitation records, it is suggested to delete this argument, as other evidences are sufficient enough to explain their observation.

Agree. We have rewritten this sentence.

- Section 3.2: 1) Collection efficiency (CE) of AMS depends on phase state of aerosol particles, which is a function of aerosol compositions. To compare AMS and SMPS data, the authors should address the approach that they used to determine the CE correction factor (e.g. compositional dependent correction). 2) Line 15-17: Direct comparison of particle cut-size between AMS (vacuum aerodynamic diameter, dva) and SMPS (mobility diameter, dm) is not appropriate. Conversion of dva to dm requires density and shape factor of aerosol particles. Please refer to AMS literature.

The CE value used in the manuscript is 0.5 which has been widely used and justified in many field studies. The reason for the large discrepancy between AMS mass vs. SMPS volume has been attributed to the interference of non-spherical particles and using a fixed CE value. Please refer to the response to reviewer #1 at the 4th specific comment.

- Section 3.4, Page 16203, line 1-3: Please briefly describe the approach and assumptions used to determine the size distribution of BC. Ion signals at m/z 57 can be due to C3H5O+ and C4H9+. Although C4H9+ is considered as a tracer of fossil fuel combustion particles (e.g. HOA from vehicle), COA is another significant contributor of C4H9+ as shown in Figure 9e and other AMS measurements. Massoli et al. (2012) observed a large difference between the size distributions of BC and HOA (using m/z 57 as a HOA tracer), especially within the range of accumulation mode particles, measured by SP-AMS in New York City. Furthermore, Lee et al. (2014) recently illustrates that coating thickness of HOA on BC particles may vary significantly with particle dva. Please specify all major uncertainties of the BC size estimation.

The uncertainties of the estimated BC size distribution are added in the updated manuscript (Section 3.4) as following:

"Note that the size distribution could be overestimated due to the contribution of $C_3H_5O^+$ (22.3%) and other source for $C_4H_9^+$ such as cooking activities (33.4%, see section 3.6.2) which has been proven by other study (Lee et al., 2014)".

- Diurnal variations of organics: 1) Section 3.3, first paragraph: In addition to primary emissions, the high concentration of organics observed from 10:00 to 13:00 can be partly due to formation of oxygenated organic aerosol (both SV-OOA and LV-OOA) via photochemistry as demonstrated in Figure 10. 2) Section 3.5, Page 16204-16205: The diurnal profiles of O/C and H/C highlight a relative importance of certain primary emissions and secondary processing in a daily basis, and hence it is strongly recommend to discuss the diurnal variations of those ratios and PMF factor in the same section. The currently discussion on diurnal profiles of organics are scatter in the manuscript. The authors may reorganize the text (Section 3.5 and 3.6) and combine Figure 8c and Figure 10 to make the discussion clear.

Solar radiation is still relatively weak between 8 - 10 am BJT, which is 7 - 9 am local time. Thus, photooxidation is unlikely an important factor for the sharp increase of SV-OOA during that time period (Fig. R1b). The mechanism of the morning SV-OOA increase is likely similar to that of nitrate which is related with the downward mixing of residual layer.

The description of diurnal variation of OA has been integrated in Section 3.6 as shown following:
"Diurnal changes of the mass fraction of HOA (Fig. 10d) presented an afternoon minimum (less than 5% of OA mass from 12:00 to 18:00), and a morning maximum (more than 20% of OA mass from 0:00 to 8:00), which was due to the photooxidation consumption of fresh aerosol and emissions from traffic activities including late night diesel truck traffic and morning rush hours. This feature can further be supported by the diurnal variations of the elemental ratios (O/C and H/C) of OA (Fig. 10a) that H/C peaked at morning rush hour and evening rush hour/dinnertime, while O/C was particularly elevated in the afternoon."

- Section 3.6.2, Page 16208, line 1: Please define f55OOA,sub and f57OOA,sub.

Done.

- Section 3.6.2, Page 16208, line 5 and line 22-24: It is not clear that 1) the temporal variations of COA and C6H10O+ are expected to be well correlated, and 2) the authors compare the size distribution of COA and m/z 55. Are they good COA tracers? Please clarify. Also, it seems that HOA has a larger contribution to m/z 55 than OOA (Figure S11d). Indeed, m/z 55 and 57 had similar size distributions.

We agree that they are indeed not tracers for COA. But it has been found in some studies (e.g., Sun et al., 2011;Ge et al., 2012) that COA show most significant correlation with a few $C_xH_yO_1^+$ ions (e.g., $C_5H_8O^+$, $C_6H_{10}O^+$, and $C_7H_{12}O^+$) and can usually use these ions to justify the COA factor. One feature for COA factor is a high signal at *m/z* 55 in mass spectrum and a high ratio of *m/z* 55/57 which can be used as a diagnostic indicator for COA factor (Mohr et al., 2012). The fraction of COA at *m/z* 55 is higher than that of HOA (44% *vs.* 30%) in Lanzhou study.

- Nature of SV-OOA, Section 3.6.3: At the early part of SV-OOA discussion (Page 16209, line 14-16), the authors mention that the SV-OOA identified in this study correlated well with the reference UMR spectra of SV-OOA previously reported by Ng et al. (2011a). In the later part (Page 16210, line 10-14), the authors further discuss the nature of SV-OOA that the mass

spectrum of SV-OOA is similar to that of coal combustion from chamber study. In general, SV-OOA is considered as secondary species with lower O/C ratio than LV-OOA as described in this manuscript but coal combustion organic aerosol is POA. Please clearly clarify the source and nature of "SV-OOA". In particular, if the "SV-OOA" originates from coal combustion sources, is there any evidence (such as SO2 and BC measurements) to support this argument?

We found that the time series of SV-OOA in the Lanzhou study correlate well with both $C_xH_yO_1^+$ and the $C_xH_{x-2}^+$ ions. The source of SV-OOA in Lanzhou is likely different than the SV-OOA factors derived from many other urban studies. The mass spectrum of coal combustion shows the important ions at m/z 69, 91, 115 (Hu et al., 2013), which all appear in our SV-OOA MS. The emission of coal combustion can also consist of secondary species as shown by the high signal at *m*/*z* 44 in the mass spectrum (Dall'Osto et al., 2013).

- Figure 15a can be removed.

We think it is necessary to keep the time for two data comparison. We would like to keep this Figure.

Technical corrections:

- Page 16193, line 3: The sampling period reported here (Jul 11-Aug 7) is different to that reported in abstract (Jul 12 - Aug 7). Please correct.

It should be Jul. 11 – Aug. 7. Revised.

- Page 16193, line 21: Please delete "about".

Done.

- Page 16197, line 2: The meaning is not clear. I suggest to change the sentence to "... while the 5-factor solution shows the splitting of a SV-OOA factor from the 4-factor solution."

Done.

- Page 16199, line 11: Please change "consistent with" to "similar to".

Done.

- Page 16199, line 16: Please change "contributions" to "fractions".

Done.

- Page 16200, line 4: Please change "Fig. 4a" to "Fig. 4b".

The Figure has been revised.

- Page 16200, line 8: Please change "Fig. 4b" to "Fig. 4a".

The Figure has been revised.

- Page 16204, line 5: Please change "Fig. 9b" to "Fig. 8b".

Done.

- Page 16206, line 28: Please change "Fig. 10e" to "Fig. 10a".

Done.

- Page 16208, line 10: Please change "Fig. 11e" to "Fig. 10a"?

Done.

- Page 16208, line 18: Please change "Fig. 11f" to "Fig. 10b"?

Done.

- Page 16208, line 25: Please change "Fig. 13b" to "Fig. 11b"?

Done.

- Page 16209, line 9: Please change "Fig. 10h" to "Fig. 9h".

Done.

- Page 16209, line 10: Wrong information: ": : :21% of CxHyO2+ ions, 42% of CxHyO2+ ions: : :"

Corrected.

- Page 16210, line 9: Please change "Fig. 10c and d" to "Fig. 10a and b"?

Done.

- Figure 1 caption: Please change the caption to "Summary of AMS measurements in China. In each site, total mass concentration, mass fractions (pie chart), and OA components based on PMF analysis (column) are shown..."

Done.

- Figure 2a: Both maps can be deleted. The location map is repeated information of Figure 1. The digital elevation model map is not discussed in the text.

The digital Figure is used to show the valley terrain of Lanzhou which can impact the aerosol evolution and chemical process. We want to emphasize this point as descripted in the beginning of the introduction.

- Figure 3c caption: Please change the caption to "mass concentration of PM1 species".

Done.

- Figure 4 caption: Please change "Fig. 4a" to "Fig. 4b" and vice versa (see previous corrections in Page 16200).

Done.

- Figure 9 caption: Please change the caption to "Time series of PMF factor and corresponding tracer species: : :"

Done.

- Figure S4 caption: Please change the caption to "5-factor solution performed by PMF on organic high resolution mass spectra."

Done.

- Figure S10: The color of size distribution plot is not labeled.

Done.

Reference:

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