

Interactive comment on “Chemical composition, sources, and processes of urban aerosols during summertime in Northwest China: insights from High Resolution Aerosol Mass Spectrometry” by J. Xu et al.

Anonymous Referee #3

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General comments:

This manuscript reports chemical compositions of submicron aerosol particles measured by an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-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 argu-

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ments, 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:

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. 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.

- 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.).

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- 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 Figure 14.
- 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.
- 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, d_{va}) and SMPS (mobility diameter, d_m) is not appropriate. Conversion of d_{va} to d_m requires density and shape factor of aerosol particles. Please refer to AMS literature.
- 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 $C_3H_5O^+$ and $C_4H_9^+$. Although $C_4H_9^+$ is considered as a tracer of fossil fuel combustion particles (e.g. HOA from vehicle), COA is another significant contributor of $C_4H_9^+$ 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 d_{va} . Please specify all major uncertainties of the BC size estimation.
- Diurnal variations of organics: 1) Section 3.3, first paragraph: In addition to primary

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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 on a daily basis, and hence it is strongly recommended to discuss the diurnal variations of those ratios and PMF factor in the same section. The currently discussion on diurnal profiles of organics are scattered 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.

- Section 3.6.2, Page 16208, line 1: Please define $f_{55OOA,sub}$ and $f_{57OOA,sub}$.
- Section 3.6.2, Page 16208, line 5 and line 22-24: It is not clear that 1) the temporal variations of COA and $C_6H_{10}O^+$ 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.
- 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 SO_2 and BC measurements) to support this argument?
- Figure 15a can be removed.

Technical corrections:

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- 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.
- Page 16193, line 21: Please delete “about”.
- 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.”
- Page 16199, line 11: Please change “consistent with” to “similar to”.
- Page 16199, line 16: Please change “contributions” to “fractions”.
- Page 16200, line 4: Please change “Fig. 4a” to “Fig. 4b”.
- Page 16200, line 8: Please change “Fig. 4b” to “Fig. 4a”.
- Page 16204, line 5: Please change “Fig. 9b” to “Fig. 8b”.
- Page 16206, line 28: Please change “Fig. 10e” to “Fig. 10a”.
- Page 16208, line 10: Please change “Fig. 11e” to “Fig. 10a”?
- Page 16208, line 18: Please change “Fig. 11f” to “Fig. 10b”?
- Page 16208, line 25: Please change “Fig. 13b” to “Fig. 11b”?
- Page 16209, line 9: Please change “Fig. 10h” to “Fig. 9h”.
- Page 16209, line 10: Wrong information: “. . .21% of CxHyO2+ ions, 42% of CxHyO2+ ions. . .”
- Page 16210, line 9: Please change “Fig. 10c and d” to “Fig. 10a and b”?
- 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...”

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- 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.
- Figure 3c caption: Please change the caption to “mass concentration of PM1 species”.
- Figure 4 caption: Please change “Fig. 4a” to “Fig. 4b” and vice versa (see previous corrections in Page 16200).
- Figure 9 caption: Please change the caption to “Time series of PMF factor and corresponding tracer species. . .”
- Figure S4 caption: Please change the caption to “5-factor solution performed by PMF on organic high resolution mass spectra.”
- Figure S10: The color of size distribution plot is not labeled.

Reference:

Massoli, P., Fortner, E. C., Canagaratna, M. R., Williams, L. R., Zhang, Q., Sun, Y., Schwab, J. J., Trimborn, A., Onasch, T. B., Demerjian, K. L., Kolb, C. E., Worsnop, D. R. and Jayne, J. T.: Pollution Gradients and Chemical Characterization of Particulate Matter from Vehicular Traffic near Major Roadways: Results from the 2009 Queens College Air Quality Study in NYC, *Aerosol Sci. Technol.*, 46, 1201-1218, 2012.

Lee, A.K.Y., Willis, M.D., Healy, R.M., Onasch, T. and Abbatt, J.P.D. (2014), Single particle characterization using a soot particle aerosol mass spectrometer (SP-AMS), *Atmos. Chem. Phys. Discuss.*, 14, 15323–15361

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 16187, 2014.

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