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Atmospheric Chemistry and Physics Discussions

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 #1

Received and published: 26 June 2014

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 devel-

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oping pollution mitigation strategies. While generally sound, there are a few technical points that should be addressed.

Recommendation:

Publish after revisions

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

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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.

Specific Comments:

Page- and Line-referenced comments on content.

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.

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.

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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.

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).

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?

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.

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

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

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rather emphasize that PMF factors are static representations of average composition patterns.

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.

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

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. . ."

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

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

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

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. . ."

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Page 16189, Line 6: Replace “quick” with “rapid”

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. . .”

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

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

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

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).

Page 16190, Line 15: subject reference

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

Page 16190, Line 25: “. . .has *been* reduced. . .”

Page 16190, Line 25: Replace “fast” with “rapid”

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

Methods Comments:

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

Page 16194, Line 16: Replace “briefly” with “brief”

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Page 16194, Line 20: “∼6000 *m/Δm* in this study” [include units]

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

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.

Results/Discussion Comments:

Page 16198, Line 21: “The concentrations of all PM1 species varied dynamically during this study, with. . .” [insert “,” between ‘study’ and ‘with’]

Page 16200, Line 25: Replace “mixing down” with “downward mixing”

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).”

Page 16202, Line 27: Replace “maintained” with “remained”, and replace “till” with “until or ‘til”

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

Page 16206, Line 15: Replace “reasonable” with “reasonably”

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

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.

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

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

Page 16212, Line 20: Replace “period” with “periods”

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

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

Page 16213, Line 15: “. . .of *the* urban boundary layer. . .”

Table 2 could be omitted for brevity.

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.”

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

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

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.”

Figure 7: Replace “distribution” with “distributions”

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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!

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

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.

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). Hydrocarbon-like 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

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