

*Review of Xu et al. “Wintertime organic and inorganic aerosols in Lanzhou, China...”*

**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.

**General comments**

**Introduction:** There are two sections of the introduction which should be better referenced. These sections are listed below.

- Lines 75 – 77: references are needed that describe the implementation of the new air quality improvement strategies mentioned in the text.
- 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.

**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?

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.

**$^{14}\text{C}$  analysis:** Only four 24 h samples were collected. Therefore, the conclusions drawn from the  $^{14}\text{C}$  analysis may not be representative and are highly dependent on the choice

of days sampled. The  $^{14}\text{C}$  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  $^{14}\text{C}$  data compare to the rest of the measurement period.

**Correlation of OOA and Odd-Oxygen ( $\text{O}_\text{X}$ ):** There are several aspects of this analysis (on lines 800 to 822) that are seriously flawed. First, only the correlation between LV-OOA and  $\text{O}_\text{X}$  has been plotted in the manuscript. It is necessary to include similar plots of SV-OOA versus  $\text{O}_\text{X}$  as well as (SV-OOA + LV-OOA) versus  $\text{O}_\text{X}$ . 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  $\text{O}_\text{X}$  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].

### **Specific Comments**

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

**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.

**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.

**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.

**Lines 274 – 276:** I'm not very familiar with TEOM instruments, but it seems strange that the instrument was operated at 40°C 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.

**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.

**Lines 478 – 479:** To my eye, it appears that the concentration of NO<sub>2</sub> stays the same or even increases slightly after 14:00 rather than decreasing.

**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.

**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.

**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.

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

**Lines 736 – 740:** This sentence is very speculative. It should either be deleted or supported by measurements of actinic flux and OH radicals.

**Lines 748 – 751:** Why would emissions of COA decrease during the holiday? Presumably people still cook during the Chinese New Year.

**Line 771:** The values for SOC in the text are different from those displayed in Figure 12.

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

**Figures 1 & 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.

**Figure 3:** It would be very interesting to include in this figure the rBC size distribution measured using the SP2.

**Figure s1a:** The text in the map is very small and difficult to read.

### **References**

Ortega, A. M., D. A. Day, M. J. Cubison, W. H. Brune, D. Bon, J. A. de Gouw, and J. L. Jimenez (2013), Secondary organic aerosol formation and primary organic aerosol oxidation from biomass-burning smoke in a flow reactor during FLAME-3, *Atmos. Chem. Phys.*, *13*(22), 11551-11571, doi:10.5194/acp-13-11551-2013.

Robinson, A. L., N. M. Donahue, M. K. Shrivastava, E. A. Weitkamp, A. M. Sage, A. P. Grieshop, T. E. Lane, J. R. Pierce, and S. N. Pandis (2007), Rethinking organic aerosols: Semivolatile emissions and photochemical aging, *Science*, *315*(5816), 1259-1262, doi:10.1126/science.1133061.