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Comment on acp-2022-142

Anonymous Referee #2

Referee comment on "Measurement report: Large contribution of biomass burning and aqueous-phase processes to the wintertime secondary organic aerosol formation in Xi'an, Northwest China" by Jing Duan et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2022-142-RC2>, 2022

Article review

Title: Measurement report: Large contribution of biomass burning and aqueous-phase processes to the wintertime secondary organic aerosol formation in Xi'an, Northwest China

Authors: Jing Duan, Ru-Jin Huang, Yifang Gu, Chunshui Lin, Haobin Zhong, Wei Xu, Quan Liu, Yan You, Jurgita Ovadnevaite, Darius Ceburnis, Thorsten Hoffmann, and Colin O'Dowd

General Comments

Duan and coauthors discuss the sources and processes contributing to organic aerosol (OA) composition in a recent Xi'an, China winter. Along with a general presentation of the aerosol characteristics, the observations were contrasted with the group's prior work in summer and in winter several years prior. A particular focus is on the aged biomass burning and aqueous processing contributions to the aerosol. The connection between various variables during two periods with high SIA concentrations is used to demonstrate both new and established observations about the fraction of the OA that was formed via aqueous processing.

My main suggestion is to add additional context and interpretation based on prior literature, both in the introduction and within the Results and Discussion. In some cases,

proper attribution of ideas to prior work must also be added. The organization of the paper is good, but consolidating the many small figures and re-organizing the content in the final section of the Results and Discussion would improve the paper substantially. Some additions to the methods section, updates to grammar, and clarifications of results presented are needed.

Overall, the quality of the content and organization is quite good, and I recommend this paper for publication with minor revisions.

Specific Comments

1. Major Content Suggestions:

1a. Additional clarity is needed to distinguish the new findings in this work from those of prior studies, and to give attribution to those works.

- Lines 123-132: Please cite relevant papers for the sources of each PMF factor.
- Lines 169-192: Please cite relevant papers that contain context for the variables discussed here, such as NO_3/SO_4 , SOR and NOR.
- Lines 224-250: Please cite relevant papers that contain context for OOA-BB and OOA-aq.

2. More background, context, and interpretation is needed.

2a. Since a main focus of the paper is the roles of SIA, ALWC, aqueous processing, and BB in Xi'an aerosol composition, please include a summary of recent papers on these subjects from the Xi'an region and elsewhere in the introduction section.

2b. In many paragraphs of the Results and Discussion, values are presented with little interpretation, making the section difficult to follow and leaving the reader wondering about the overall message. Please add specific interpretation of the many values presented, add context for whether the findings are new to this study, cite prior studies that have discussed similar findings, and discuss what these findings imply about aerosol regionally or generally. Examples: lines 169-192 and lines 224-250.

- Lines 181-182 state that, "...high RH and liquid phase condition may drive the large production of SIA." Further discussion and attribution for this idea is needed. In addition, there is an inherent dependence of ALWC on SIA because their concentrations are used to calculate ALWC. The explanations for Figs. 4 and 5 also require more discussion of this topic. Please further discuss the prior work demonstrating causality between SIA and heightened ALWC, and also heightened OOA-aq, and new implications of this study.

2c. Van Krevelen analysis can be useful to summarize complex organic composition, but not to determine the specific molecular pathways in long datasets. Factors such as mixing of aerosol populations can affect O/C and H/C, and integrated composition over long time periods cannot show what's happening at the molecular level. Please instead focus on the shorter-term and non-molecular Van Krevelen analyses, or perhaps cautiously discuss the similarities between your results and those of lab studies, as in Heald et al., 2010.

3. Structural suggestions:

3a. There are many figures, and some present duplicate information. Many of the figures are barely discussed in the text (fig. 6(a), for example). I suggest consolidating or moving some additional figures to the Supplemental.

3b. Section 3.4: The organization of this section is confusing. The content seems to be a mixture of continued discussions about the BB influence and SIA period source/process characterization, then Van Krevelen analysis for SIA periods, then back to general Van Krevelen periods. I suggest reorganizing the material, renaming the section, and perhaps fitting relevant pieces into 3.1 and 3.2.

3c. Some material in lines 117 through 140 (Methods, Data Analysis section) is Results material rather than Methods, and should be moved.

4. Methods questions:

4a. Were averaged ratios such as O/C calculated following Aiken et al., 2008? (https://cires1.colorado.edu/jimenez-group/wiki/index.php/FAQ_for_AMS_Data_Users#How_do_I_average_OA_elemental_composition_variables_.28O:C.2C_H:C_OA:OC_and_OSc.29.3F)

4b. Please discuss (in the supplemental is fine) the process for selecting the solution with the six PMF factors chosen.

4c. Please define the equations described by NOR and SOR.

4d. Please add the calculation for odd oxygen here (difficult to find definition for "Ox" within the introduction).

4e. Please add the gas analyzer models.

4f. Please mention the wind speed and direction measurement methods.

4g. Please discuss the precise metrics used to define the beginnings and ends of SIA_P1 and SIA_P2.

4h. Please highlight the assumptions and considerations made to account for changes in volatility or other losses during sampling at high humidity. For example:

- Were tests with wetted standard particles performed to determine whether the humidity impacted the concentrations?
- Was RH considered in composition-dependent collection efficiency calculations?

4i. If a soot particle long time-of-flight aerosol mass spectrometer (SP-LToF-AMS) was used, why were the elemental carbon concentrations not discussed?

5. Additional content questions:

5a. Were the different PMF factors identified here versus in summer Xi'an expected? What does this imply about these factors and atmospheric chemistry of the region? For example, Sun et al., 2016 found a substantial summertime agricultural burning source at Xianghe, but there is no BB factor in Duan et al., 2021.

5b. Ammonium is included in the SIA concentrations but is never discussed separately even though it is emitted separately from nitrate and sulfate. Have ammonium concentrations or the competition by nitrate and sulfate for ammonium changed? How do they relate to the organic aerosol (OA) and OA fraction concentrations? Also see Zhang et al., 2021 (DOI: 10.1021/acs.estlett.0c00756).

6. Minor comments:

- Some grammar should be updated. In particular, the word "promoted" is used in a confusing way in many places.
- Introduction lines 49-50: I suggest adding more recent references to support the statement that SOA is, "...becoming a critical concern for air pollution research".
- Line 69: "...Research on aerosol composition and SOA formation mechanisms are still limited" would be well qualified with "in the region".
- Introduction, line 60: Are there recent papers to support the statement that aqueous-phase chemistry is missing in SOA simulations? I suggest also saying here that aqueous SOA is difficult to identify.
- Introduction, line 75: Please add a reference for the "13th five-year energy conservation and emission reduction plan".
- Throughout the Results and Discussion: Values are presented that are apparently averages, and some with an error calculation ($X \pm Y$). Please clarify that these values are the arithmetic means and standard deviations of the per-minute samples over the campaign or specified sub-period.
- Throughout the Results and Discussion: Concentrations of the OA fractions should be included to support the trends in emissions (for example, at lines 200-214). A change in contribution does not necessarily demonstrate an absolute change in abundance.
- Results and Discussion, lines 173 and 176: The OA/NR-PM₅ percentage is presented differently as 65 % and 66 % at these lines, respectively.
- Results and Discussion, line 183: Please clarify the equations used to calculate the, "...increase ratio of sulfate contribution from reference days to SIA_P1" ... "and to SI_P2...".
- Figure 4: The data in (a) are already plotted in a prior plot. Suggest adding ALWC to Fig. 1 and removing 4(b).
- Results and Discussion, lines 272-274: these ions are not listed in Tan et al., 2009.
- Fig. 3(b): Are there additional studies using high resolution AMS data in China that could be added to demonstrate work done outside of your research group?
- Fig. 4(b) and Fig. 5: These patterns are surprising and very stark. Would you please confirm that the different slope groupings are not the result of the ALWC ISORROPIA modeling? (Wu et al., 2018 also used ISORROPIA). Additionally, please define "S" in the figure caption.
- Results and Discussion, lines 305-307 ("This may suggest that aq-OOA..."): This is an interesting statement, but I'm not sure I understand how this conclusion was drawn. Please further explain the logic.
- Fig. 6(b): Pie charts do not have labels. Also, when characterizing aqueous processes, the f_{44} vs f_{43} space has been used (for example, Lee et al., 2011; DOI: 10.1029/2011GL047439), and might be more relevant for aq-OOA discussion here.
- Results and Discussion, lines 345-354: Do the meteorological parameters or mesoscale meteorological observations help explain the differences between SIA_P1 and SIA_P2?
- Supplement: Figure S5 (b) is confusing. What data are included in the aq-OOA profile resolved in summer? It seems like they are data from Duan et al., 2021, but it would not be possible to pair summer with winter concentrations. Please explain.
- Author contributions: Several authors are simply listed as having. "...commented on and discussed the manuscript". Please ensure that the integrity of co-authorship is preserved (https://www.springer.com/journal/10874/submission-guidelines#Instructions%20for%20Authors_Authorship%20principles).

