General comments:

The authors present detailed chemical characteristics of atmospheric submicron particles by performing a wintertime field observation at a suburban site in the Sichuan Basin, southwestern China, using multiple advanced instruments such as HR-ToF-AMS, MAAP, GC-MS/FID, PTR-MS, and TEM-EDX. On the basis of AMS high-resolution mass spectra data, four OA factors were identified by PMF source apportionment analysis. Secondary formation and aging process of organic aerosols were also investigated with different approaches; especially, significant influence contributed by biomass burning was discussed.

I would recommend this paper to be accepted after more in depth discussions are included, and after the following specific comments are addressed.

Specific comments:

1. The **Introduction** section is not logically connected well with the Results and Discussion part, which mainly focuses on particle chemical characterization, secondary OA formation, aging processes of OA, and the possible influence of biomass burning on these properties. Relating to the abovementioned topics, previous studies and corresponding results, especially in this studied region, were expected to be summarized in the introduction instead of simply included in results and discussion (e.g. Sect. 3.3.4). The introduction described much previous work on poor visibility/haze issues; however the connections with the discussion part were not clearly illustrated. The authors did not explain well why high time resolution particle chemical characterization in Sichuan Basin is needed, and why it is necessary to investigate secondary formation in the influence of biomass burning by using high time resolution aerosol mass

spectra here (**paragraph 2, Page 3**).

Throughout the paper: The authors often use the term **biomass burning** organic aerosol (BBOA) to indicate OA contributed by domestic cooking (COA) and residential heating (e.g. Page 3, line 20). On Page 3, line 22-23, biomass burning is actually included in biogenic sources; thus biomass burning could also originate from other open fires such as rice straw burning and forest fires. On Page 4, line 13-15, the authors have pointed out that BBOA and COA are regarded as two different primary sources of OA in some studies, although complete identification among BBOA, COA, and CCOA with PMF analysis is not easy in practice. To avoid confusion during using these different expressions, it would be better to keep a consistent way, and define biomass burning and BBOA of this study clearly in the very beginning.

I would suggest the authors to reorganize the Introduction to better connect with the topics discussed in this manuscript.

2. **Introduction**, Page 4, line 3-5

   How should this sentence be understood? Why is primary organic aerosol excluded from the discussion here (only SOA is included)? Is there any difference between OM and OA?

3. **Methodology**, Page 5, line 8

   What is the elevation of the sampling site itself? How did you choose the altitude of the starting point of the backward trajectory calculation?

   Page 5, line 12, *"Therefore, the atmospheric processes are dominated by the isolated meteorology of the basin."*

   Did the backward trajectory analysis really support this idea?

4. Although the chemical composition analysis is focusing on submicron

particles, the actual cut-size of MAAP is $PM_{2.5}$, while that of HR-ToF-AMS is PM_1. How did the authors consider this mismatch of two different size ranges into the mass fraction calculation, which could vary significantly with different proportions of BC accounted for PM_1? Relevant details should be provided in the measurement and data processing descriptions.

5. Page 7, line 1-2

   Why RIE = 4.04 was applied to ammonium? Can you provide any reference or supporting information?

   The recommended value of CE = 0.5 is used. Are there any comparative results or strong supporting evidence to verify its applicability? Middlebrook et al. (2011) have demonstrated the composition-dependence of CE with field measurements, suggesting that CE could be higher when ambient particles are composed of high fraction of ammonium nitrate or strongly acidic sulfate. This phenomenon could be significant especially under high RH conditions. In this sense, these issues may need to be considered, as high RH conditions were frequently observed during the investigation period of this work.

6. **Results and discussion**, Page 9, line 4

   Is this sentence describing inorganic species or organic species?

7. Page 9, line 7

   "However, the humid air caused by the precipitation may favor the aqueous-phase secondary formation and hygroscopic growth of SNA in turn."

   How should the reader understand "and hygroscopic growth of SNA in turn" here? Please provide some necessary supporting information and illustrate the connections clearly.

8. **Sect. 3.1.2**, Page 10, line 1

"It was consistent with the morphology and mixing state of single particles, mostly spherical and in internal mixing state (Fig. 4a-d)."

It is not clear to me if Fig.4 really supports the authors' conclusion. At least, some aggregated soot particles can be clearly seen. Besides, the size resolution of TEM images is 2 μm, much larger than submicron or even ultrafine size ranges.

9. Page 10, line 4

*"… indicating that the aerosols at Ziyang site may be **more aged** than in other areas."* How did you arrive at this conclusion? The higher peak sizes only demonstrate that particles are larger.

10. **Sect. 3.2.1**, Page 11, line 10

"The MS of HOA correlated well with the average MS of HOA factor reported in previous studies, as well as that of COA, BBOA and vehicle emitted OA (Vehicle-OA) factors (Table S3). Thus, it was likely that the HOA factor was a mixture of COA and other primary organic aerosols."

In this case, could it be possible to resolve different factors better by increasing the number of factors for the PMF analysis? It is hard to believe that emissions from the three sources (COA, BBOA, and Vehicle-OA) correlate well all the time. The HR-ToF- AMS simply observes fragments. Is there any possibility that there was a specific source of OA during the observation in that area? m/z 60 exists in the HOA factor. Where does it come from: coal combustion or biomass burning?

11. **Sect. 3.2.2**, Page 12, line 13

*"… presented good correlations with BC and acetaldehyde (Table S4), which were mainly emitted from **primary sources**."*

Can you tell that it is only emitted from biomass burning, or is it also contributed by other types of primary sources?

12. **Sect. 3.2.3**, Page 12, line 18

*"… as the influence of biomass burning is **negligible**."*

Applicability of this assumption depends on characteristics of specific observation site, even though some studies have suggested insignificant influences of biomass burning on OA. The authors have also highlighted that BBOA contributes significantly to their data. Accordingly, this concept may not be justified in this study.

13. Page 13, line 4

*"In this study, LV-OOA **correlated well** with SNA (r=0.66-0.68)"*.

Is the reported ***r* value** considered as an indication of good correlation?

14. Page 13, line 8

*"LV-OOA also showed a similar trend to BC (r=0.75), **maybe because** BC was difficult to diffuse and mixed well in the static air."*

The statement is confusing and ambiguous. How should readers understand it?

15. **Sect. 3.3.4**, Page 19, line 2

*"The increase of OA …, **which** was approximate to that reported at Changdao Island …"*

Does the "which" mean the slope of increased OA or contribution of SOA?

16. Page 19, line 5

"… the average OA/ΔCO ratio decreased with photochemical age, caused by

the decrease of LV-OOA/ΔCO ratio." Is it still valid if the *SV-OOA/ΔCO ratio* increased at the same time?

The following descriptions of the subsequent sentence are unclear. Please clarify them so that the readers can understand it clearly. Namely, how should the readers understand the "**relatively stable** SV-OOA concentrations" resulted from "inhibited evolution from POA to SV-OOA", while "inhibited evolution from POA to LV-OOA resulting in **lower** LV-OOA"?

17. Page 19, line 10

*"SOA dominated OA (56-84%) in both **fresh** and aged plumes…"*

Do you need to define the "fresh" plume in this work to distinguish it from "aged" ones, or provide a certain threshold value in terms of different photochemical ages?

18. Page 19, line 15

"… implying that the photochemical formation of SV-OOA was more efficiently than that of LV-OOA in this campaign."

Is this conclusion applicable only to cases for longer photochemical age? We can find from Fig.11 that the fractions of SV-OOA are not always higher than that for LV-OOA, especially when the photochemical age is less than about 6h.

19. Figure 1

In addition to wind speed, wind direction is also an important indicator of air mass origin or possible influence by transportation. Perhaps you can try to display both wind speed and direction parameters in Fig.1(a) and discuss accordingly.

20. The whole passage is generally well organized; however some important

statistics of chemical information are expected to be presented in the manuscript, instead of the supplementary materials. For example, **Table S1** actually contains many new and interesting primary results obtained from this study. The mass concentrations of BC under different meteorological conditions could also be a good case. The contribution of BC to PM_1 has been included in abstract and conclusion sections, indicating the importance of BC in chemical characteristics of submicron particles. The corresponding results would be more straightforward to readers if shown in the manuscript.

Technical corrections:

1. **Introduction**, Page 2, line 9

   "… has become one of the most polluted regions in China."

   Corresponding references are needed, as well as for the specific values that are not obtained from this study (e.g., Page 3, line 17 and 19).

2. **Methodology**, Page 7, line 13

   "… the diurnal patterns of different factors, etc. (Zhang et al., 2011)".

   Please specify the "etc" clearly.

3. **Sect. 3.2**, Page 10

   *"**SOA** (OOA) dominated in OA as much as 71% …"*

   *"… secondary formation (**SOA**+SNA) …"*

   Please be careful when using SOA and OOA, as OOA is not completely the same as SOA.

4. **Sect. 3.3.4**, Page 19, line 21

   "… and reached saturation frequently (Table S1)."

   Does it mean average RH or RH?

Reference: *Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer using Field Data, Aerosol. Sci. Tech., 46, 258–271, doi:10.1080/02786826.2011.620041, 2011.*