Response to the referee’s comments

We would like to thank the reviewer for valuable comments and suggestions. We have addressed all raised issues in the revision accordingly. Please kindly find our following point-by-point responses. The reviewer’s comments in black and our responses in blue. Any amendments in the revised manuscript are highlighted in red.

Response to Reviewer #2

This manuscript reports a ToF-ACSM measurement study of sub-micron particles conducted during winter time in Guangzhou, South China. PMF with ME-2 algorithm was applied on the dataset to identify the major sources of organic aerosols (OA). Discussions are made on concentrations, compositions, and sources ambient PM1, highlighting the important roles of SOA. Additionally, the relationship with SOA and peroxy radicals was examined to reveal the different mechanisms responsible for SOA formation between non-pollution period and pollution Eps. The manuscript is well written and provides some interesting results for understanding ambient primary and secondary organic aerosol sources and processes. I would recommend the publication of this manuscript in Atmospheric Chemistry and Physics after the authors address the following comments.

[A]: Thank you for the comments and valuable suggestions. We have changed accordingly. Please find our point-by-point responses below.

Comments:

1. The resolution and font sizes need to be improved? (e.g., Fig. 1, Fig. 4b and Fig. 5).

[A]: We have increased the resolution and font sizes of the figures.

2. Please check the subscript in the texts and figures.

[A]: We have doubly checked the entire manuscript to ensure no typos with the subscript.

3. Page 7, Line 191-194: It would be good to add a more accurate discussion of the calculated composition dependent CE values (e.g., range, highest frequency and uncertainty of CE values).

[A]: We have added several sentences to discuss the calculated composition dependent CE values in the revision (lines 217-220, pages 7-8):
“The results showed that only about 1% of samples (78 of 6623) had CE values larger than 0.45 (others are 0.45), with the largest value being 0.578. Hence the influence induced by fluctuation of the CE values is negligible and we chose a CE value of 0.45 for the ACSM measurements in this study.”

4. Page 8, Line 237-241: Recently, a large number of AMS/ACSM studies have been conducted in China in recent years. Is it possible to add more references and discuss with more results?

[A]: We thank the reviewer for updating us on this information. We have included 11 additional publications, with a comprehensive summary being added in the revision (Table 1, in the revised manuscript or below). In addition, we have also modified in Figure 3 by including more measurement data for Beijing and Lanzhou in the revision.

Here we only compare measurements during winter season, corresponding to our measurement periods. Our survey shows that SOA formation is significantly influenced under different underlying surfaces (urban, suburban, and country). Nevertheless, additional survey adds more measurement data into the NR-PM$_1$ pool. However, our original conclusion of increasing the fraction of SOA in OA from north to south still holds. We have modified the paragraph that describes NR-PM$_1$ measurements in China in the revision (line 267-269, page 9). “Furthermore, Table 1 shows that the SOA fraction is generally enhanced from winter to summer for a specific site in China. In addition, Table 1 also revealed that SOA formation is significantly influenced under different underlying surfaces (urban, suburban, and country).”

5. Page 15, Line 455-477: I suggest the authors put these parts in introduction and highlight the differences of your results from previous ones.

[A]: We thank the reviewer for valuable suggestions. We have made some modifications and moved these sentences to introduction section in the revision (lines 140-159, page 5).

In addition, we have added one sentence to highlight differences of our results from previous studies in the revision (lines 177-178, page 6).

“Possible mechanisms for wintertime SOA formation were explored through introducing RO$_2^*$ as a proxy for gas-phase oxidation capacity during both daytime and nighttime.”

6. Page 28, Fig. 4: The factors of HOA and COA were resolved using the constrain mode (a-value), but SVOOA and LVOOA were identified using the PMF free mode. So, to be more directly clear for readers,
the authors may consider adding the corresponding label in each mass spectrum of POA factors (e.g., constrained or a specific a-value) and SOA factors (e.g., unconstrained or free).

[A]: According to the reviewer’s suggestions, we have added the corresponding labels in Figure 4.

![Figure 4](image)

**Figure 4.** The mass spectra and time series of the four OA components (HOA, COA, SVOOA, and LVOOA).

7. Page 29, Fig. 5: The diurnal profile of NOx appears to be bi-modal, yet no morning traffic feature is visible in the HOA diurnal plot during pollution Eps. Have the authors looked for the variation of HOA mass fraction during early rush hour? More explanation about the diurnal profile of HOA would be good.
[A]: We thank the reviewer for pointing this out. Though it is small, there was a small HOA peak at about 8:00 in Fig. 5 in the manuscript. In fact, strong traffic emissions from heavy duty vehicles during midnight to 6:00 in the early morning weaken the morning rush hour peak. Similar features were frequently reported in previous studies (Sun et al., 2013; Qin et al., 2017; Huang et al., 2019). The rapidly rising boundary layer after 7:00 would be possibly another reason for diluting PM accumulation from rush hour traffic. Besides, if we discuss the variation of HOA mass fraction, a clear morning peak was observed (Fig. S12, see it in supplementary or below). Thus, the diurnal feature of HOA during pollution Eps is reasonable. We have added a sentence in the revision for clarification (lines 298-299, page 10): “Besides, effects of emissions from heavy duty vehicles and the rapidly rising boundary layer after 7:00 would also account for the insignificant peak of HOA during morning rush hour.”

![Figure S12. The diurnal variations of 4 OA components mass fractions for pollution EPs.](image)

8. Section 3.5.2: It would be interesting to see how the SOA changes during different conditions. The authors may consider adding the correlations between SVOOA and LVOOA with RO2*, perhaps in the supplement.

[A]: We have plotted dependence of SVOOA/LVOOA concentrations on RO2* concentration during non-pollution periods and pollution periods in Figure S15 (see it in supplementary or below). It is shown that better correlations between LVOOA and RO2* than between SVOOA and RO2*. In addition, the slope from LVOOA vs RO2* is higher than that from SVOOA vs RO2*, implying transformation of SVOOA to LVOOA. In contrast, neither LVOOA nor SVOOA were well correlated to RO2*, possibly due to strong heterogeneous/multiphase reactions during pollution EPs as discussed in the text.
We have added several sentences in the revision to reflect the correlations between SVOOA/LVOOA concentrations and RO$_2^*$ concentration (lines 491-495, page 17).

“In addition, correlations between SVOOA/LVOOA and RO$_2$ were explored by plotting dependence of SVOOA/LVOOA concentrations on RO$_2^*$ concentration during non-pollution periods and pollution periods (Fig. S15). The results show that better correlations and larger slope for LVOOA vs RO$_2^*$ than for SVOOA vs RO$_2^*$ during non-pollution periods. In contrast, neither LVOOA nor SVOOA were correlated to RO$_2^*$ during pollution EPs.”

**Figure S15.** Scatter plots between RO$_2^*$ and SVOOA/LVOOA for non-pollution periods and pollution EPs.
<table>
<thead>
<tr>
<th>Location</th>
<th>Date</th>
<th>Region</th>
<th>NR-PM$_1$ (μg m$^{-3}$)</th>
<th>OA (%)</th>
<th>SOA/OA (%)</th>
<th>Ref.</th>
</tr>
</thead>
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<tr>
<td>Guangzhou</td>
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<td>49</td>
<td>70</td>
<td>This study</td>
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<td>Winter, 2014</td>
<td>Pearl River Delta region</td>
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<td>50.5</td>
<td>61</td>
<td>Qin et al., 2017</td>
</tr>
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<td>Winter, 2009</td>
<td>Pearl River Delta region</td>
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<td>46.2</td>
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<td>Huang et al. 2011</td>
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<td>66</td>
<td>Zhang et al., 2017</td>
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<td>Huang et al., 2012</td>
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<tr>
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<td>Winter</td>
<td>Beijing-Tianjin-Hebei region</td>
<td>70</td>
<td>52</td>
<td>43</td>
<td>[1]</td>
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$^{[1]}$: The three quantities for winter Beijing are the averaged values over six studies (Sun et al., 2013; Sun et al., 2014; Jiang et al., 2015; Wang et al., 2015; Hu et al., 2016a; Sun et al., 2016).
References


Sun, Y., Du, W., Fu, P., Wang, Q., Li, J., Ge, X., Zhang, Q., Zhu, C., Ren, L., Xu, W., Zhao, J., Han, T., Worsnop, D. R., and Wang, Z.: Primary and secondary aerosols in Beijing in winter: sources,


