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.

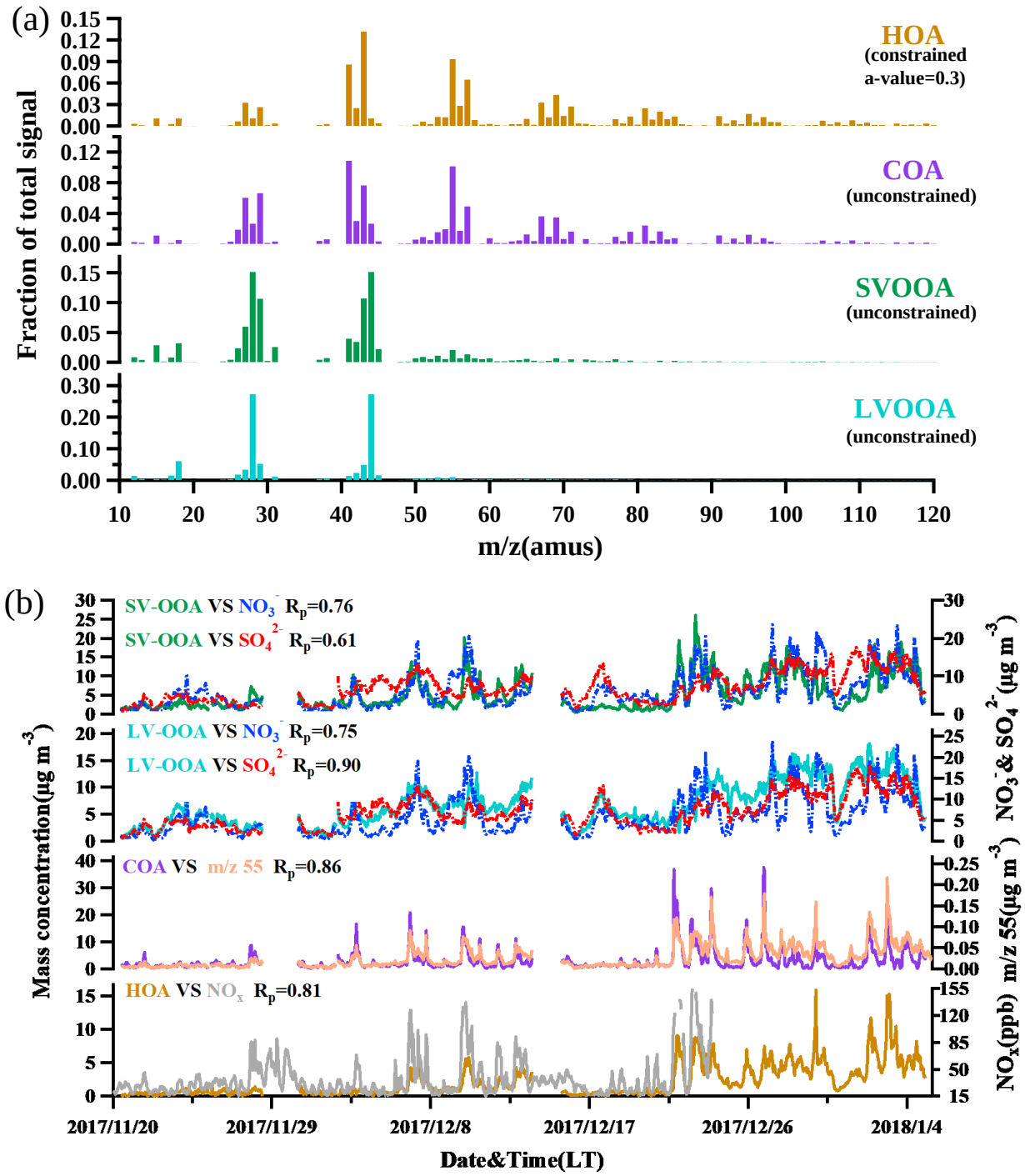

Figure 4. The mass spectra and time series of the four OA components (HOA, COA, SVOOA, and LVOOA).

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."

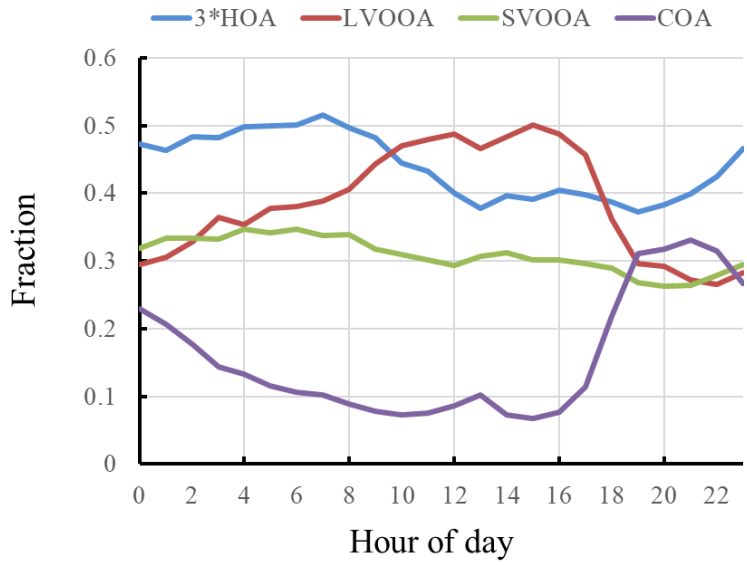

Figure S12. The diurnal variations of 4 OA components mass fractions for pollution EPs.

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 RO_2* 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 RO_2* than between SVOOA and RO_2*. In addition, the slope from LVOOA vs RO_2* is higher than that from SVOOA vs RO_2*, implying transformation of SVOOA to LVOOA. In contrast, neither LVOOA nor SVOOA were well correlated to RO_2*, 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."

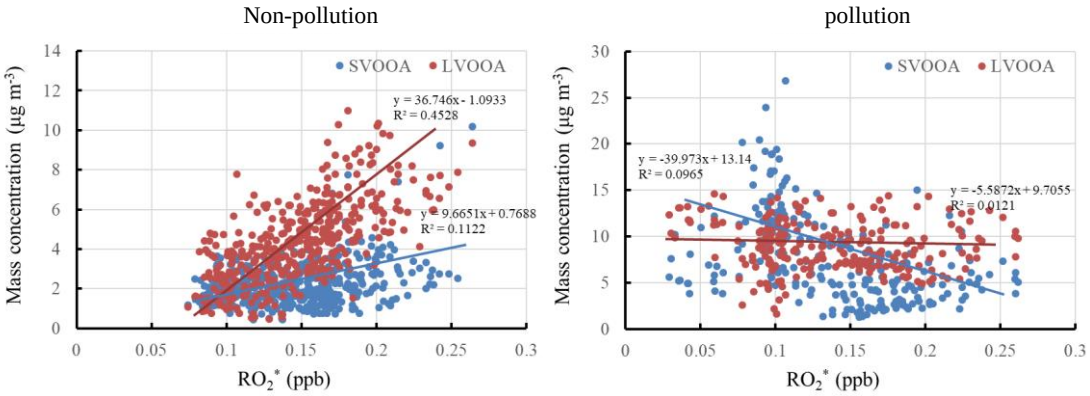

Figure S15. Scatter plots between RO_2* and SVOOA/LVOOA for non-pollution periods and pollution EPs.

Table 1. Summary of reported NR-PM_1 measurements in China.

Location	Date	Region	NR-PM_1 (μg m^{-3})	OA (%)	SOA/OA (%)	Ref.
Guangzhou	Winter, 2017	Pearl River Delta region	35.3	49	70	This study
Panyu	Winter, 2014	Pearl River Delta region	55.4	50.5	61	Qin et al., 2017
Shenzhen	Winter, 2009	Pearl River Delta region	44.5	46.2	46.6	He et al., 2011
Kaiping	Winter, 2008	Pearl River Delta region	33.1	36.3	75.4	Huang et al. 2011
Nanjing	Winter, 2015	Yangtze River Delta region	32.5	36	66	Zhang et al., 2017
Nanjing	Summer, 2013	Yangtze River Delta region	36.8	42	72	Zhang et al., 2015
Shanghai	Summer, 2010	Yangtze River Delta region	27	31	78	Huang et al., 2012
Beijing[1]	Winter	Beijing-Tianjin-Hebei region	70	52	43	**[1]**
Beijing	Summer, 2011	Beijing-Tianjin-Hebei region	80	32	65	Hu et al., 2016a
Shijiazhuang	Winter, 2014	Beijing-Tianjin-Hebei region	178	50	22	Huang et al. 2019
Handan	Winter, 2015	Beijing-Tianjin-Hebei region	178	47	17	Li et al., 2017
Lanzhou	Winter, 2014	Northwest of China	57.3	55	37	Xu et al., 2016
Lanzhou	Summer, 2012	Northwest of China	24	53	59	Xu et al., 2014
Hong Kong	Winter	South of China	20.7	45.5	69	Sun et al., 2016
Hong Kong	Summer, 2011	South of China	15.6	26	82	Li et al., 2015
Ziyang	Winter, 2012	Southwest of China	60	40	71.2	Hu et al., 2016

[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).

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