Response to anonymous referee #2

We thank reviewer #2 for the thorough and careful review of our manuscript. Below we reply to the reviewer's comments point by point. We list the comments in black, our replies in blue, major changes or additions to the manuscript in red.

Main Comments

1) In page 19410, line 8-9 and in the PMF component spectra it is clear that the observed organic aerosol contains significant levels of m/z 60 and 73. The large body of AMS literature has shown that these ions are typically indicative of influence from biomass burning organic aerosol. These ions have also been used in ACSM studies to show biomass influence (A simple internet search with the keywords "biomass burning ACSM factor", for example, brings up several of the pertinent literature publications). Thus, it is very surprising that the authors do not mention this possibility in the organic aerosol analysis. Why is biomass burning discounted as a source? Some ideas on how the authors can check for the influence of biomass are:

We agree that the presence of m/z 60 and 73 in SV-OOA is unusual. In fact, we tried to separate a biomass burning factor from our PMF analysis but it did not yield satisfactory results. We will give more details in the response to comment #2 further below. We analyzed the source of m/z 60 and 73 but have not included these results in the manuscript. We attribute m/z 60 and 73 in Mong Kok mainly to local cooking activities and long-range transport. The similar diurnal pattern and well matched peaks in the time series data between m/z 60, m/z 73 and COA indicate that cooking emissions contribute part of m/z 60 (Fig 1). Furthermore, LV-OOA tracks very well with the baseline of m/z 60 and m/z 73 as shown in Fig 2, illustrating the partial contribution of long range transport to m/z 60 and m/z 73. Consequently, the sum of LV-OOA and COA shows a better correlation with m/z 60 (Rpr=0.72) and m/z 73 (Rpr=0.78) than each single factor as shown in Table 1. In addition, there are no notable biomass burning sources around the Mong Kok site, which is urban in nature without agricultural or domestic burning practices. Thus a local source of BBOA is very unlikely. Based on above information, we conclude that m/z 60 and 73, marker fragments of BBOA, were mainly imbedded in cooking emissions and transported aerosol rather than a distinct source.

Page 19410, line 4, *addition and modification to main manuscript*:

We note that m/z 60 and 73, important makers of BBOA mass spectra (Aiken et al., 2009; Cubision et al., 2011; Huang et al., 2011), were resolved not only in COA but also in SV-OOA. Their presence in SV-OOA is not the result of artifacts from the PMF analysis, but were attributed to the following reasons, with more details shown in the supplement (sect.3-5). Firstly, when PMF was run using only nighttime data (between 0:00 and 6:00), i.e. when there is little COA (Fig. S6), these two ions still persist with similar fractional intensities in SV-OOA as at other times. Secondly, increasing the number of PMF factors and adjusting the fpeak value did not yield a distinct satisfactory BBOA factor. Thirdly, the time series of m/z 60 and 73 show weak correlation with other burning tracers (EC_residual, CO_residual), with Rpr of about 0.2 and 0.4 respectively, but track well with SV-OOA, with Rpr of 0.92 and 0.93 respectively (Figure 3, Table1).

In terms of the possible sources of m/z 60 and 73, we observe that these two ions showed matching peaks with the COA diurnal profile and good correlations with the sum of the time series of COA and LV-OOA, with Rpr of 0.72 and 0.78 respectively. Furthermore, the ratio of the integrated signal at m/z 60 to the total signal in the organic component mass spectrum is 0.48%, which is just slightly higher than the baseline level (0.3%±0.06%) observed in environments without biomass burning influence and with SOA dominance in ambient OA (Cubision et al.,2011). This indicates that these two ions at Mong Kok were mainly imbedded in cooking emissions and background aerosol due to transport rather than in a distinct source with further details shown in the supplement (sect.6).

Section 4: SV-OOA vs BBOA, *addition to supplemental materials*:

In the 4-factor solution, the mass spectra of resolved SV-OOA contain significant fractions of m/z 60 and m/z 73, which are important makers of BBOA mass spectra (Aiken et al., 2009; Cubision et al., 2011; Huang et al., 2011). The resolved SV-OOA spectrum in our study correlates well with both standard SV-OOA and standard BBOA spectra with Rpr of 0.87 and 0.94, respectively (Figure 4). To assess whether a distinct BBOA factor could be resolved, we increased the number of PMF factors from 4 to 6. With a 5-factor solution, the existing SV-OOA factor was split into factor 1 and factor 4 as shown in figure 5. However, the mass spectrum of factor 1 is very different from that of BBOA, and the time series of factors 1 and 4 show a high correlation with Rpr of 0.8, indicating a common source rather than two different components. For the 6-factor solution, the existing SV-OOA factor was split into factor 1 and factor 4,and the HOA factor was divided into factor 3 and factor 6 (figure 6). The mass spectra of factor 1 and factor 4 are very different from that of standard BBOA with a Ruc of 0.56 and 0.68, respectively. Thus, we are not able to separate a distinct BBOA factor from the existing SV-OOA based on the unit mass resolution data, which is consistent with previous studies conducted in Hong Kong (Li et al., 2011, 2015; Lee et al., 2011, 2015).

Section 5: SV-OOA vs COA, *addition to supplemental materials*:

Apart from SV-OOA, m/z 60 and m/z 73 are also present in the mass spectrum of COA, and they share a similar diurnal pattern with COA (Figure1). To examine the possibility that PMF erroneously assigned these two ions to COA instead of SV-OOA, PMF was run using only nighttime data (between 0:00 and 6:00), when there is only residual COA present (Figure S6). We found that these two ions still persist, with similar fractional intensities in SV-OOA as at other times and have thus been correctly assigned.

On the other hand, the diurnal pattern of SV-OOA also shows matching peaks with COA. To further examine the relationship of the COA factor and SV-OOA factor, PMF was run with 4, 5 and 6 factor solutions respectively. The correlations between COA and SV-OOA like factors decrease (Table 2) as the number of PMF factor increases. However, the mass spectral correlations (Ruc) between resolved COA and standard COA decrease from 0.84 to 0.77, and the correlation of the sum of SV-OOA like factors and standard SV-OOA reduces from 0.87 to 0.79, indicating a reduction of the quality of PMF solution. In addition, the average COA loading decreases from

3.6 to 2.7 µg/m^3 rather than increase, which suggests that increasing the number of PMF factors does not separate possible COA related components from the SV-OOA like factors.

The effects of varying fpeak on the correlations between SV-OOA and the COA time series from this 4-factor solution were also analyzed. As shown in Tables 3 and4, when the fpeak value changes from 0 to -0.2 and -0.4, the time series correlation of SV-OOA and COA just decreases slightly, but the mass spectra correlations between resolved COA, SV-OOA and their standards decrease appreciably. When the fpeak value increases from 0 to 0.4, the similarity of COA and SV-OOA times series increase dramatically, and except for LV-OOA the mass spectra correlations between resolved OA factor and their standard profiles all decrease. We acknowledge the limitation and uncertainty of PMF analysis on resolving factors, but considering the resulting mass spectra, the time series data of the loadings and the correlations with standard mass spectra, we consider that the four-factor solution with an fpeak value of 0 is optimal in our case.

Section 6: the source of m/z 60 and 73, *addition to supplemental materials*:

The fractions of signal at m/z 60 and m/z 73 to the total signal of SV-OOA like factors are persistent and remain above 1% as the number of PMF factor increases from 4 to 6 as shown in table 2. In terms of the source of these two ions, the similar diurnal pattern and well matched peaks in the time series data between m/z 60, m/z 73 and COA indicate that cooking emissions contribute part of m/z 60 and m/z 73 (Fig 1). Furthermore, LV-OOA tracks very well with the baseline of m/z 60 and m/z 73 as shown in Figure 2, illustrating the partial of contribution of long-range transport to m/z 60 and m/z 73. The sum of LV-OOA and COA show a better correlation with m/z 60 (Rpr=0.72) and m/z 73 (Rpr=0.78) than each single factor as shown in Table 1, supporting above hypothesis that transport aerosol and local cooking emissions are both sources of m/z 60 and 73 at Mong Kok. In addition, the comparison between m/z 60, 73 and plumes of EC and CO do not correlate well with NOx represented by EC_resiudal and CO_residual (Fig. 3). m/z 60 showed weak correlation with the residual of EC (Rpr=0.37) and CO (Rpr=0.21). A similar weak relation is apparent for m/z 73. Therefore, biomass burning influence around the Mong Kok site is highly unlikely. Also, the ratio of the signal at m/z 60 to the total signal in the OA mass spectrum (0.48%) in this study is just slightly higher than the baseline level (0.3%±0.06%) observed in environments without biomass burning influence and SOA dominance in ambient OA (Cubision et al.,2011). Based on the above analysis, we conclude that m/z 60 and 73, usually marker fragments of BBOA, were mainly imbedded in cooking emissions and transport aerosol rather than a distinct source.
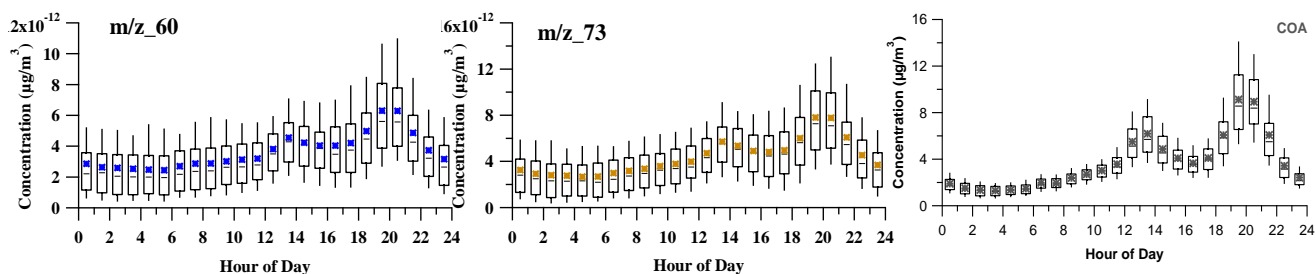

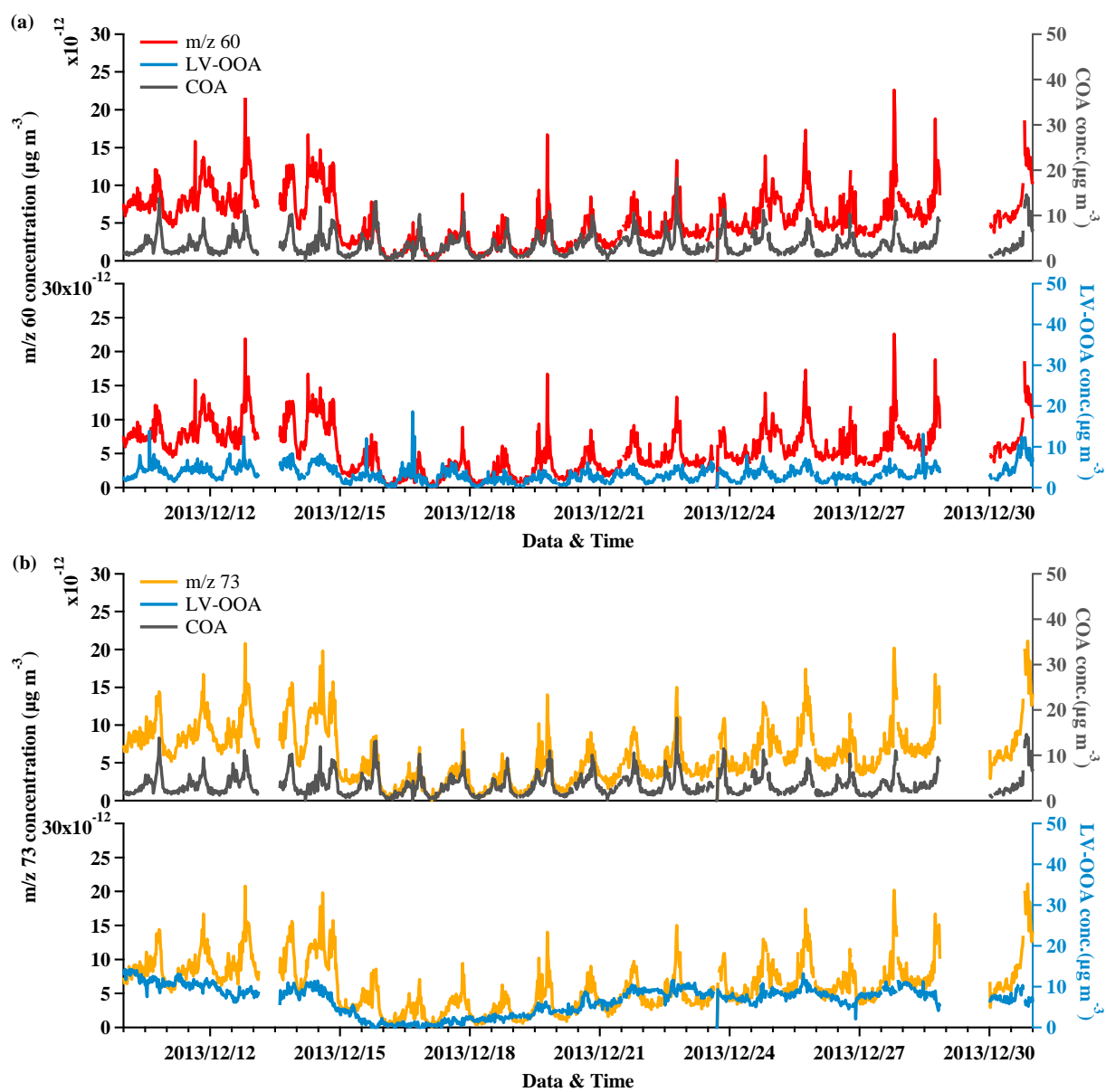Fig 1. Diurnal pattern of m/z 60, m/z 73 and COA

Fig 2. Temporal variation of m/z 60, m/z 73, LV-OOA and COA, excerpt from December, 2013.

Table 1. Correlations of the time series data between m/z 60, 73 and SV-OOA, LV-OOA, COA, sum of LV-OOA and COA, NO3, NH4 and SO4.

Pearson R	SV-OOA	LV-OOA	COA	LV-OOA+COA	NO3	NH4	SO4
m/z60	0.92	0.55	0.49	0.72	0.66	0.59	0.42
m/z73	0.93	0.54	0.58	0.78	0.64	0.57	0.41

\# The time trends of m/z 60 and m/z 73 can be analyzed and compared to each other as well as external burning tracers such as EC and CO. Plumes of EC and CO that do not correlate with NOx can be used as possibly indication of biomass burning influence

We added the comparison between m/z 60, 73 and plumes of EC and CO that do not correlate with NOx represented by EC_resiudal and CO_residual (Fig. 3). EC_residual and CO_residual are defined as the residual of the equation: EC(or CO) =a* NOx. m/z 60 showed weak correlation with the residual of EC (Rpr=0.37) and CO (Rpr=0.21). The same applies to m/z 73. Therefore, influence of biomass burning at MK is quite unlikely.

Please refer to "**Page 19410, line 4,** *addition and modification*" on the response to main comment 1).

"The time series of m/z 60 and 73 show weak correlation with burning tracers (EC_residual, CO_residual), with Rpr of about 0.2 and 0.4 respectively."
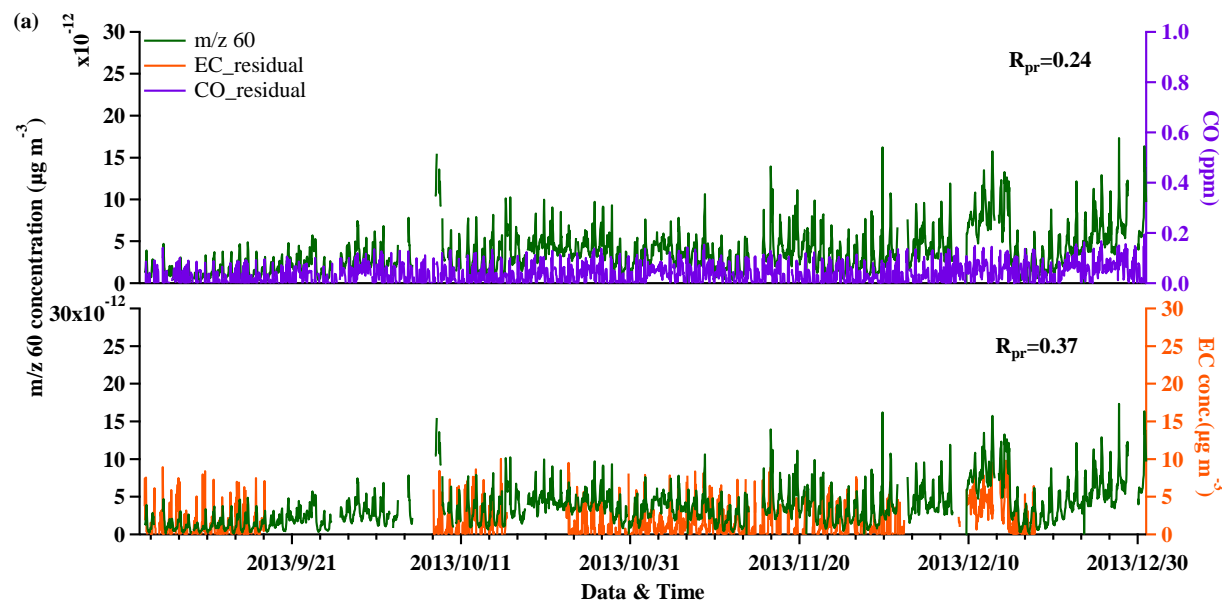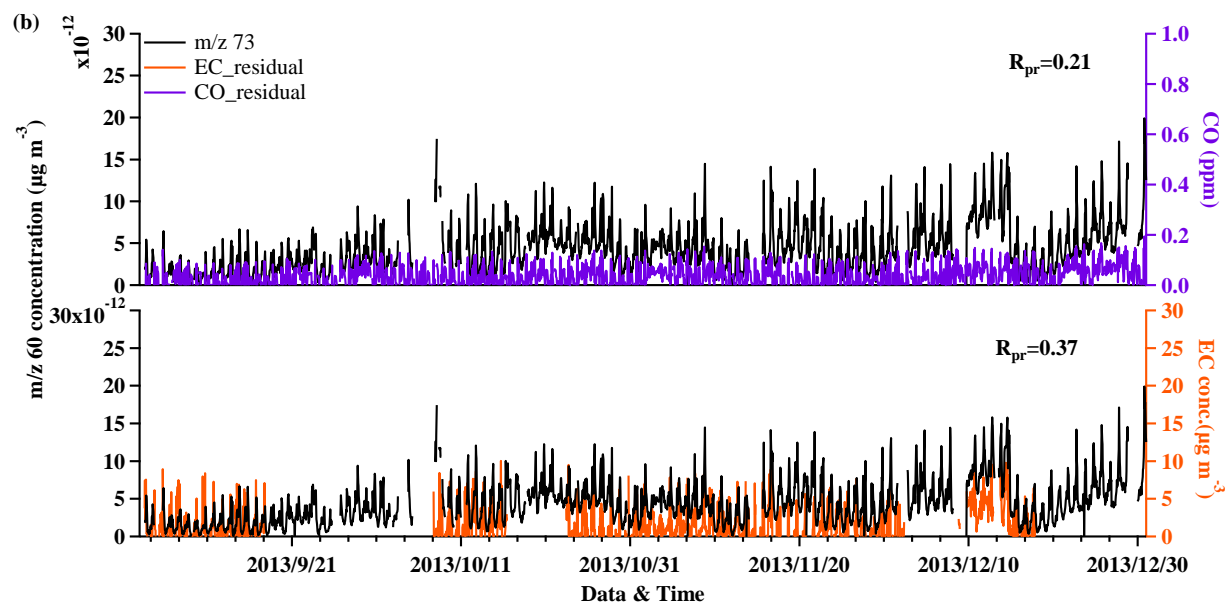
Fig. 3 Time series of m/z 60, m/z 73, EC_residual and CO_residual. Note that EC_residual and CO_residual are defined as the residual of the equation: EC(or CO) =a* NOx.

\# A manuscript by Cubison et al. has reported f60 values in various air masses with and without biomass influence. The observations from Hong Kong can be compared to those values.

We agree and will expand on the OOA part as detailed below.

Please refer to "**Page 19410, line 4,** *addition and modification"* on the response to main comment 1).

Furthermore, the ratio of the integrated signal at m/z 60 to the total signal in the organic component mass spectrum is 0.48%, which is just slightly higher than the baseline level (0.3%±0.06%) observed in environments without biomass burning influence and SOA dominance in ambient OA (Cubision et al.,2011)

\# Comparisons of the observed SV-OOA spectrum to "standard" BBOA spectra in spectral databases

We compared the SV-OOA spectrum with the standard ones as shown in the graph below. The resolved SV-OOA spectrum in our study correlates well with both standard SV-OOA and standard BBOA spectra with Rpr of 0.87 and 0.94, respectively. As discussed previously, the BBOA signature ions are mainly attributed to long-range transport and cooking activities rather than local biomass burning. Transport of biomass burning derived pollutants from the PRD region is possible, but would appear as processed OA, i.e. either in form of SV-OOA or LV-OOA. In addition, increasing the number of PMF factors from 3 to 6 (see details below) did not yield a separate BBOA factor. This is consistent with previous studies in Hong Kong (Li et al., 2013, 2015; Lee et

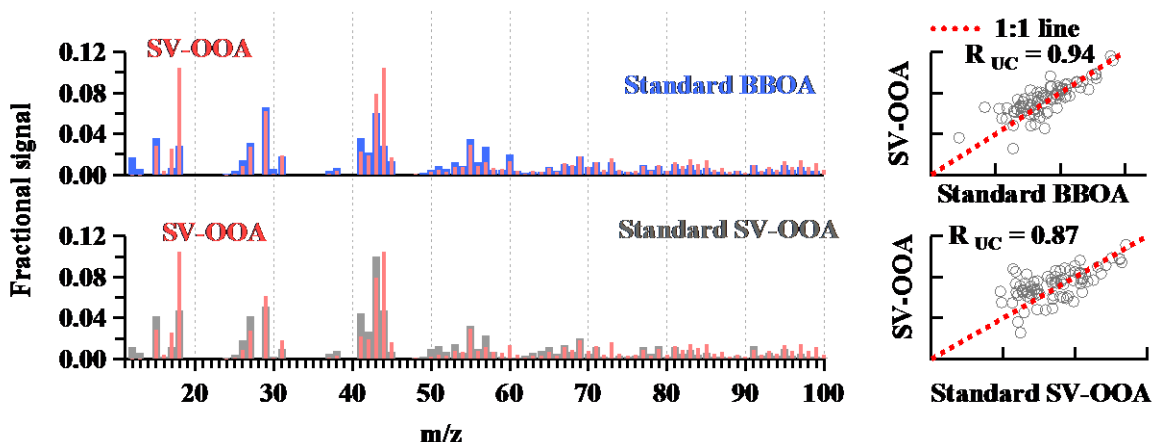al., 2013, 2015) which also could not resolve BBOA.

Fig 4. Mass spectra of resolved SV-OOA (pink) in our study and standard mass spectra of BBOA available on the AMS MS database (Ulbrich, I. M., Lechner, M., and Jimenez, J. L., AMS Spectral Database)

Since the SV-OOA component concentration is largely influenced by regional continental transport, and appears to be particularly important in high concentration pollution events, it may be possible to see whether there is any correlation between SV-OOA and other data regarding fires in the region.

We do not have data regarding fires in and beyond the PRD region.

The residuals in the PMF analysis of m/z 60 and m/z 73 can be investigated to see if they perhaps get a lot smaller at a larger number of factors and if perhaps a clean biomass burning factor splits from the existing SV-OOA component at larger factor numbers.

With a 5-factor solution, the existing SV-OOA factor was split into factor 1 and factor 4 as shown in figure 5. However, the mass spectrum of factor 1 is very different from that of BBOA, and the time series of factors 1 and 4 show high correlation with Rpr of 0.8, indicating a common source rather than two different components. For the 6-factor solution, the existing SV-OOA factor was split into factor 1 and factor 4,and HOA was divided into factor 3 and factor 6 (Figure 6)., The mass spectra of factor 1 and factor 4 are very different from that of standard BBOA with a Ruc of 0.56 and 0.68, respectively. Thus, we are not able to separate a distinct BBOA factor from the existing SV-OOA. In addition, the fractions of signal at m/z 60 and m/z 73 to the total signal of SV-OOA like factors do not reduce much as the number of PMF factors increases from 4 to 6 as shown in Table 2.
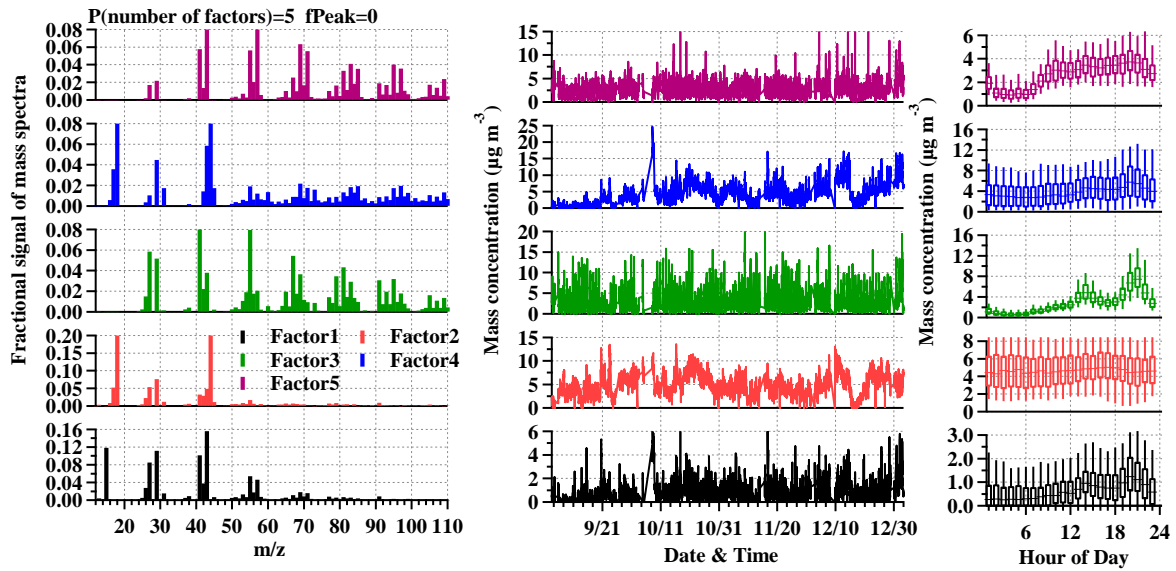
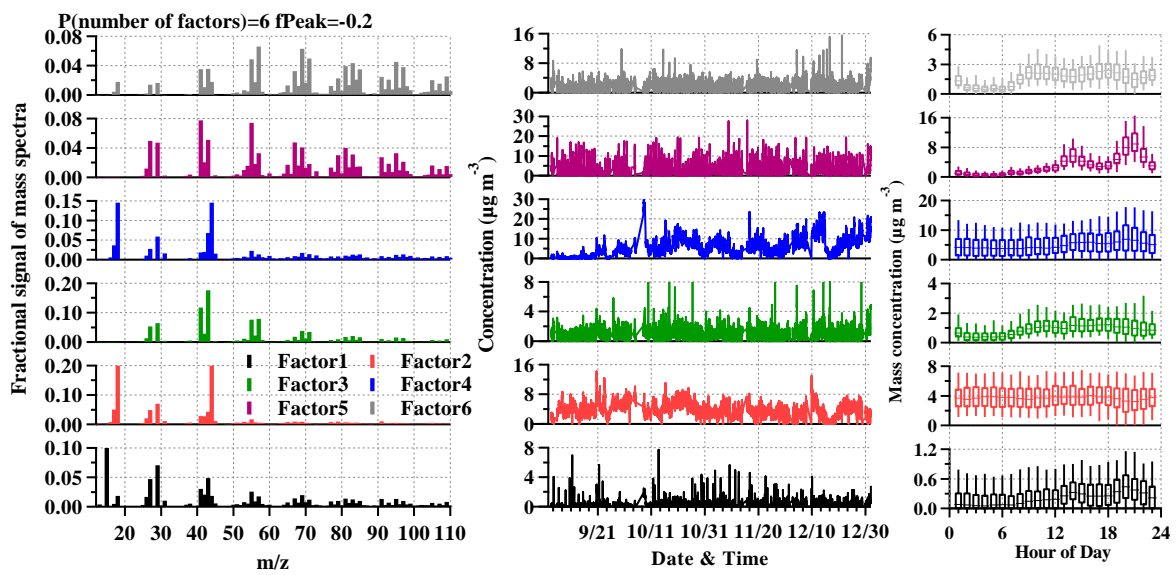Figure 5. Mass spectra, time series and diurnal pattern for 5 factors with fPeak=0.

Figure 6. Mass spectra, time series and diurnal pattern for 6 factor solution with fPeak=-0.2.

Table 2. Correlation of resolved OA factor mass spectra and their standard profiles, time series correlation between COA and SV-OOA like factors, fractional signal at m/z 60, 73 to total SV-OOA signal, and the average mass concentration of OA factors for the 4-factor, 5-factor and 6-factor solutions.

	Organic factors	4 factor	5 factor	6 factor
Ruc for mass spectra	COA with standard	0.84	0.76	0.77
	SV-OOA with standard	0.87	0.85	0.79
	HOA with standard	0.93	0.93	0.98
	LV-OOA with standard	0.97	0.97	0.97
Rpr for Time series	COA with SV-OOA like factor 1	0.45	0.36	0.26
	COA with SV-OOA like factor 2		0.39	0.22
	COA with SV-OOA like factor 1+ factor 2		0.37	0.28
Fraction in SV-OOA	m/z 60	1.4%	1.3%	1%
	m/z 73	1.6%	1.5%	1.1%
Concentration ($\mu g/m^3$)	Average COA	3.6	2.6	2.7
	Average SV-OOA	3.1	4.4	5.0
	Average HOA	2.7	2.5	2.3
	Average LV-OOA	5.7	5.2	5.0

2) It is clear from the observed SV-OOA time trends (particularly peaks at 12 pm and 6 pm) that the SV-OOA factor contains some influence of the COA factor. Unless the SV-OOA species are secondary species formed from the cooking process (or other co-located SOA source), there is no other simple reason why SV-OOA should contain this diurnal trend. The authors do not clearly address how they tried to deal with this mixing. This is important considering that the SV-OOA is a significant fraction of the OA. Some questions pertinent to this are:

Did the authors try to go to a much larger number of factors and investigate what happened to the COA loadings and correlation with the SV-OOA-like factor?

Table 2 above summarizes the results of possible 4, 5 and 6 factor solutions from the PMF analysis. When the number of PMF factors increases from 4 to 6, the correlations between COA and SV-OOA like factors show a decrease (Table 2). However, the mass spectral correlations (Ruc) between resolved COA and standard COA decrease from 0.84 to 0.77, and the correlation of the sum of SV-OOA like factors and standard SV-OOA reduces from 0.87 to 0.79. In addition, the average COA loading decreases from 3.6 to 2.7 $\mu g/m^3$ rather than showing an expected increase, which suggests that increasing the number of PMF factors does not separate possible COA related components from the SV-OOA like factors.

We acknowledge the limitation and uncertainty of PMF analysis on resolving factor completely, but considering the resulting mass spectra, the time series data of the loadings and the correlations with standard mass spectra, we consider the four-factor solution as optimal in our case.

Did the authors investigate the effect of fpeak on the time series correlation between SV-OOA and COA. What do the results look like at the fpeak setting where this correlation is minimized?

According to the above analysis, we have identified the 4-factor solution is the most appropriate for our study. The effects of fpeak on the correlations between SV-OOA and COA time series are thus presented for the 4 factor solution. As shown in Tables 3 and 4, when the fpeak value changes from 0 to -0.2 and -0.4 subsequently, the time series correlation of SV-OOA and COA decreases slightly, but the mass spectral correlations between resolved COA, SV-OOA and their standards decrease appreciably. When the fpeak value increases from 0 to 0.4, the similarity of COA and SV-OOA times series increase dramatically, and except for LV-OOA, the mass spectra correlations between the resolved OA factors and their standard profiles all decrease. Thus, an fpeak value of 0 for the 4-factor case considered as the most appropriate solution for the PMF analysis.

Table 3. Time series correlation between resolved SV-OOA and COA for the 4-factor solution for different fpeak values.

Rpr	-0.4	-0.2	0	0.2	0.4
SV-OOA and COA	0.40	0.43	0.45	0.64	0.63

Table 4. Correlation between our resolved PMF factors from the 4-factor solution with standard mass spectra for different fpeak values.

Ruc\fpeak	-0.4	-0.2	0	0.2	0.4
HOA vs STD	0.94	0.94	0.92	0.90	0.88
COA vs STD	0.77	0.77	0.84	0.76	0.75
LV-OOA vs STD	0.98	0.98	0.97	0.97	0.97
SV-OOA vs STD	0.83	0.85	0.87	0.78	0.84

Even if they are unable to use it for this manuscript, the authors should at least mention that ME-2 based analyses like possible with the SOFI tool could be a means of dealing with this.
We have expanded on the Experimental section as detailed below.

Page 19409, line 19, *addition to main manuscript***:**

"ME-2 analysis with the SOFI tool as applied in several studies may yield additional insights but has not been applied in this study due to its ongoing development (Canonaco et al., 2013; Minguillón et al., 2015)."

Relevant methods similar to those used by Aiken et al. to evaluate biomass burning in Mexico city (http://www.atmos-chem-phys.net/10/5315/2010/acp-10-5315-2010.pdf) could be attempted.

Aiken et al. used satellite data derived fire counts and FLEXPART modeling, both of which are not available for this study.

3) One weakness of this manuscript is that it reads like a report of AMS/ACSM measurements at yet another field site. It would be useful for the authors to provide as much inter-comparison with other previous measurements as possible to provide a larger con-text within which we can understand these measurements. For example:

The authors mention that transport from PRD can be a source of some of the observed aerosol at the Hong Kong site. How do the loading and composition of the aerosol particles observed at the current site differ from those previously observed in the PRD region? Is it possible, for example, that BBOA from the PRD is a source of the observed m/z 60 and m/z 73 in the ACSM spectra at this site?

These measurements were conducted in the winter and fall. How do the results (absolute concentrations and relative compositions) differ with previous studies at the same site or similar site that were conducted at the same or other seasons?

We will expand on the introduction as detailed below.

Page 19408, line 10, *addition to main manuscript***:**

Recently, a high resolution aerosol mass spectrometer (HR-ToF-AMS) was applied at an urban site in the Shenzhen metropolitan area and a rural site in PRD region during October and November (He et al., 2011; Huang et al., 2011). They found that organic concentration dominates followed by sulfate which is similar to this study, but the fraction of sulfate at the rural site is larger than that of the urban site. Four OA components were identified in urban site including HOA, BBOA, LV-OOA and SV-OOA, but only three OA factors without HOA were resolved in rural site. They both reported an important contribution from BBOA with about 24% of total OA.

We have previously deployed HR-ToF-AMS at the supersite of the Hong Kong University of Science and Technology (HKUST) to determine typical variations in submicron species concentrations, overall composition, size distributions, PMF-resolved organic factors and degree of oxygenation. The supersite measurements provided valuable insights into characteristics of mainly of secondary components of submicron particulate matter, with dominance of sulfate and oxygenated organic aerosol species observed [Lee et al., 2013;Li et al., 2013, 2015]. Subsequent work was conducted at a downtown location (Mong Kok) in Hong Kong, next to the roadside, in spring 2013 to assess important primary aerosol sources in the inner-city to identify contributions of long-range transport to roadside pollution, and to establish characteristic concentration trends at different temporal scales. Cooking aerosol was identified as the dominant component in submicron non-refractory organics, followed by traffic-related emissions [Lee et al., 2015].

This work focuses on the characterization of roadside aerosol during the fall and winter seasons, when the influence of transported air mass is greatest and PM pollution in Hong Kong generally more severe. Episodic haze events were found to be mainly driven by secondary aerosol rather than primary emissions, while hourly high PM concentrations were often driven by cooking aerosol. Statistical methods were employed to show that the correlation of COA and HOA to SV-OOA varied under different conditions and period of a day. While HOA showed a stronger relationship to SV-OOA overall, COA can be an important contributor to SV-OOA during meal times.

Other comments

4) Section 3.2: When the various OA components are described, it would be useful to have the brief description of their mass spectra (which is currently in the supplementary) included in the main manuscript to reinforce the key mass spectral features used in the factor assignments. Similarly, the discussion of the COA that is currently in the supplementary could be moved to the main.

We agree and will modify the manuscript accordingly.

Page 19412, line 12, *addition to main manuscript*:

The mass spectrum of HOA is dominated by the $C_nH_{2n-1}^+$ ion series (m/z 27, 41, 55, 69, 83, 97), typical of cycloalkanes or unsaturated hydrocarbon, which account for 27% of total peak intensity in the HOA spectrum. The other prominent group is the $C_nH_{2n+1}^+$ ion series (m/z 29, 43, 57, 71, 85, 99), typical of alkanes and accounting for 26% of the total peak. This mass spectrum is very similar to the standard HOA spectrum with R_{uc} of 0.92, and its fractions of $C_nH_{2n-1}^+$ and $C_nH_{2n+1}^+$ (27%, 26%) are consistent with standard ones (=28%, 27%) (Ng et al., 2011). This HOA spectrum is also consistent with that resolved by HR-ToF-AMS at the HKUST Supersite on the dominance of saturated CxHy-type ions, most notably at *m/z* 43 and 57 (Lee et al., 2013).

Page 19413, line 19, *addition to main manuscript*:

The most prominent ions of the resolved COA profile at MK were *m/z* 41 (mainly C_2HO^+, $C_3H_5^+$) and *m/z* 55 (mainly $C_3H_3O^+$, $C_4H_7^+$). Ratios of *m/z* 41/43 =1.8 and *m/z* 55/57=2.2, which are distinctly larger than that of HOA at 0.73 and 0.76 respectively (Figure 4); such ratios have been widely reported for COA in AMS and ACSM studies.For example, Lanz et al. (2010) reported ratios of *m/z* 41/43 and *m/z* 55/57 of 0.5 and 0.4 in HOA, and 1.2 and 1.2 in COA, respectively, while Sun et.al (2013) reported 0.5 for these two ratios in HOA and 2.3 for those in COA, respectively.

Page 19414, line 18, *addition to main manuscript*:

LV-OOA is characterized by the prominent m/z 44 ion (mainly CO_2^+) and minor C_nH_{2n-1} and C_nH_{2n+1} ion series generated by saturated alkanes, alkenes and cycloalkanes.

Page 19414, line 24-25, *modification of main manuscript*:

SV-OOA, which is less oxidized than LV-OOA, is marked by the dominant ions of m/z 43 and m/z 44 mainly contributed by $C_2H_3O^+$ and CO_2^+, The mass spectrum of SV-OOA closely resembles that of 'standard' SV-OOA with a R_{uc} of 0.87 (Fig. 3). Its time series also follows that of nitrate (R_{pr}=0.63, Figure 1), another secondary and semi-volatile species.

5) Page 19415, line 22-19416, line 5: this section is a little long winded. It would be better to cut out a lot of the discussion of the correlation coefficients, which are not really that useful, and instead point out the differences in actual mass concentrations for the different periods that are in the table. While it is useful to have done the HiOx and LOx comparisons, I think it can be summarized in a couple sentences and it is not clear to me that this extended discussion provides any more useful information about the SV-OOA than is possible from the diurnal cycle. So, I would get rid of this aspect of the discussion. It would be better to focus on the types of diagnostics suggested in comment #2 above in the main comment section.

We will make the manuscript more concise on the Table 2 and 3 description as detailed below. However, we consider the discussion of correlation coefficients useful to show their relative importance to SV-OOA under different conditions and help to identify possible reasons for the observed concentration changes.

Page 19416, line 5 to Page 19417, line 5, *modification of main manuscript*:

The average concentrations of HOA and SV-OOA under HTemp are obviously lower than under LTemp for each period but the concentration of COA and LV-OOA varies little across different temperatures (Table2). Combining the stronger correlations between HOA and SV-OOA than between COA and SV-OOA, a stronger and closer temperature dependence of HOA and SV-OOA was revealed. In addition, the regression coefficients of HOA and COA during each period under HTemp (Table 2) are much smaller than under LTemp, reflecting a weakening of their relationship with SV-OOA as temperature increases.

Consistent with the discussion of Fig 7, the concentrations of HOA, SV-OOA and LV-OOA except for COA under HiOx are greatly higher than those under LOx for each period. Besides, HOA shows an increase correlation with SV-OOA under HiOx due to the more intensive oxidation of HOA precursor to SV-OOA. However, LV-OOA shows a reverse trend with smaller coefficients with SV-OOA. It is probable that HiOx conditions favor the conversion of SV-OOA to LV-OOA leading to smaller coefficient of LV-OOA on SV-OOA, although overall most LV-OOA is considered to be from transport.

At last, we also can conclude that HOA overall has a stronger relationship to SV-OOA than COA has, supported by much higher coefficients of HOA than that of COA over all time periods, and temperature and Ox levels. Cooking emissions are not as important to SV-OOA in the BT periods but they can be important during MT periods, indicated by the lowest concentration and correlation with SV-OOA during BT but highest concentration during MT periods."

6) Page 19418, page 3- Isn't the diurnal variation in aerosol components like Chloride also driven by the boundary layer? Are their measurements of CO that show how much the dilution is during the day? If so, it would be useful to show the CO diurnal trends as well for reference.

As our sampling site is situated in a typical roadside area with heavy influence of traffic emission, the concentration of CO was influenced greatly by traffic emissions, as shown below. In addition, complex and high building structures around the measurement site (street canyon) complicate the effects of circulation and PBL variation and thus we are not able to directly assess the influence of dilution due to changes in PBL height.
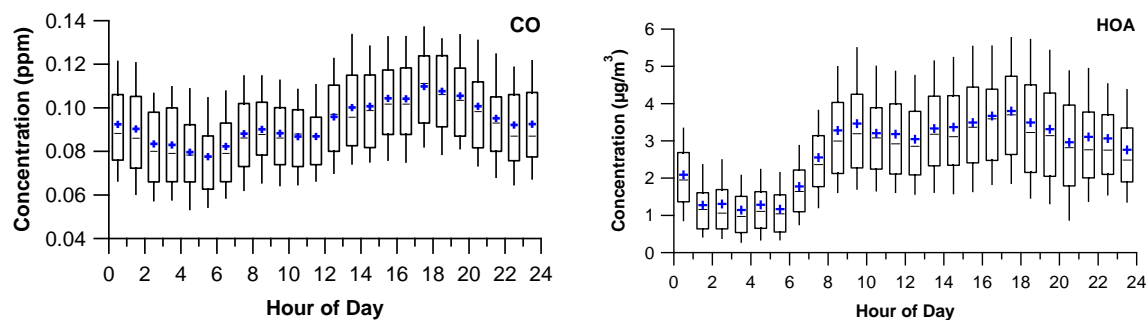

Figure 7. The diurnal pattern of CO and HOA during the whole study.

7) Page 19420- what is the wind direction classification for C2? Is the main difference between C1 and C2 the fact that it rained? If so this should be stated. Also, it would be nice if in the discussion of Table 4, the periods that should be directly compared with each other due to similarity in source regions or other conditions are explicitly stated. Otherwise, the reader has to try and summarize for themselves the results from the analysis of table 5. It would also help if in Table 4, the source region classification (i.e. continental, coastal etc.) of each period was provided.

The wind direction of C2 is mainly northerly, belonging to continental wind. The main differences in meteorological conditions between C2 and C1 are the occurrence of precipitation, much lower temperature and source region shift from coastal to continental region. According to your comment, Table 4 will be expanded as detailed below and the manuscript will be reorganized.

Page 19420, line19, *modification of main manuscript*:

Although the total NR-PM1 of C1 (12.2 μgm^{-3}) and C2 (11.8 μgm^{-3}) are both only 25–30% of that during haze periods, they were driven by different mechanisms. The main differences in meteorological conditions between C1 and C2 are the dominance of continental wind rather than coastal wind, much lower temperature and the existence of precipitation in C2. The low concentration of C1 is mainly attributed to easterly wind bringing less air pollutants and diluting local air pollutants. To a lesser extent, it is influenced by both particle evaporation, especially for SV-OOA, and dilution of local emissions during high temperatures, which might be the reason why HOA, COA and SV-OOA in C1 are lower than in C2 despite the lack of rain. The low mass

loading of C2 was mainly caused by the wet deposition of precipitation. It dramatically reduces the concentration of secondary species such as SO4, NH4, NO3, SV-OOA and LV-OOA, but not primary HOA and COA. Compared to the adjacent period H3, the total organic mass reduces by 68% to an average of 8.1 μgm^{-3} (Table 4). Precipitation effectively removes secondary particles but is less efficient for primary particles that are continuously generated locally.

With similar continental source region as C2, the most severe pollution event H3 occurred during 10–13 December with an average NR-PM1 of 47.7 μgm^{-3}. The persistent northerly wind continually brought air masses from the PRD region into Hong Kong and lead to a marked mass increase of secondary species of SO4, NH4, NO3, LV-OOA and SVOOA. Furthermore, H3 is characterized by the highest mass concentration and relative contribution of nitrate and SV-OOA compared with other haze periods. This is likely due to the average temperature of H3 being 5–6°C lower than that of other haze events.
In addition, although all three haze events have very similar SO4 mass loading, there is a ~ 50% increase in NH4 concentration during the H3 episode, consistent with the increase of nitrate in that period.

The other two haze events are adjacent with influence from both continental and oceanic region in H1 and continental source region in H2. The mixed pattern of source regions during H1 identified as land–sea breeze (Fig. S8) can redistribute PM pollution over the whole PRD region and accumulate air pollutants effectively (Lo et al., 2006; Chan and Yao, 2008; Lee et al., 2013). The pronounced high concentration of LV-OOA and SV-OOA, jointly contributing 70% of total organics, reflects the oxidation of primary emissions in the PRD under such cycles, which is also observed at the suburban HKUST site (Lee et al., 2013). The periodic nitrate peaks in H1 with low concentration in daytime and high concentration in nighttime coincide with temperature changes. During H2 period, the prevailing wind is northwesterly and there is a sharp decrease in relative humidity. It is interesting to note that the dip in RH during H2 coincides with the dip in sulfate, ammonium, nitrate and LV-OOA; this might be caused by decreased aqueous-phase processing, and by decreased gas-particle partitioning associated with water uptake under low RH for secondary aerosol particles (Sun et al., 2013a, b).

Table 4. Measured and calculated parameters in four chosen periods (C1, H1, H2, H3 and C2)

	Clean period 1 (C1)[a]		Haze period 1 (H1)		Haze period 2 (H2)		Haze period 3 (H3)		Clean period 2 (C2)	
RH (%)	70.8		65.0		36.4		64.8		84.6	
T (°C)	27.6		25.0		23.8		18.7		13.2	
O_x (ppb)	69.6		82.0		99.5		70.4		40.9	
$f44$	0.114		0.118		0.120		0.108		0.057	
Precip(mm)	0		0		0		0		8.9	
Wind	coastal		continental/oceanic		continental		continental		continental	
(µg/m³, %)	Conc.	Perc.	Conc.	Perc.	Conc.	Perc.	Conc.	Perc.	Conc.	Perc.
NR-PM$_1$	12.2		44.1		39.0		47.7		11.6	
Org	6.7	54.4	25.2	57.2	21.1	54.2	25.1	52.6	8.1	69.6
SO$_4$	3.8	31.2	11.8	26.8	12.1	30.9	11.4	23.8	1.5	12.8
NH$_4$	1.2	9.9	4.4	10.1	4.4	11.3	6.5	13.6	1.1	9.4
NO$_3$	0.4	3.5	2.4	5.6	1.3	3.4	4.4	9.2	0.8	7.3
Chl	0.1	1.0	0.2	0.4	0.1	0.2	0.4	0.8	0.1	0.9
HOA	1.2	18.5	3.8	15.1	3.0	14.4	4.2	16.9	2.1	26.2
COA	2.3	34.8	3.7	14.5	3.3	15.5	3.3	13.1	2.6	31.7
LV-OOA	3.0	44.8	11.5	45.4	10.2	48.4	9.9	39.6	1.8	22.0
SV-OOA	0.1	2.0	6.3	25.0	4.5	21.6	7.6	30.4	1.6	20.1

8) Figure 3: The COA and SV-OOA MS correlations with the reference spectra look quite scattered. This is likely indicative of the fact that the SV-OOA has COA mixed into it. It would be useful in supplementary to show the correlation spectra for SV-OOA and COA with markers corresponding to m/z so that the masses that have discrepancies are more easily identified. Another option would be to draw the reference spectra behind each of the component mass spectra in gray so that the comparison can be readily made by the reader.

We have modified Figure 3 in the manuscript by adding reference spectra behind each of the component mass spectra in gray as detailed below.
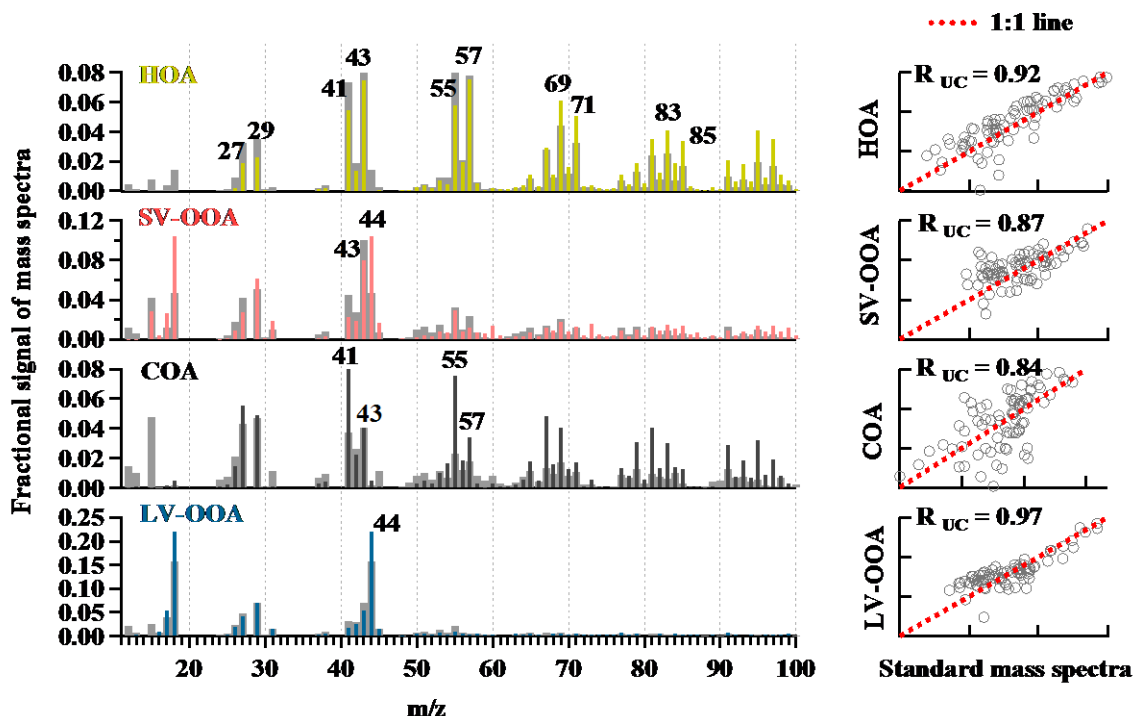
Figure 3. Mass spectra of resolved OA components (HOA, SV-OOA, LV-OOA, COA) with the corresponding standard spectra (in gray) and the correlation with standard mass spectral profiles available on the AMS MS database (Ulbrich, I. M., Lechner, M., and Jimenez, J. L., AMS Spectral Database). The x and y axes in the right-hand graphs are mass spectra of resolved factor and the standard, respectively.

9) It is not clear that Figure 7 adds that much to the discussion. The conclusions from these figures could be stated in words in a few sentences

Figure 7 serves as an illustration of the comprehensive effect of Ox and temperature on the concentrations of the OA components. We agree that these conclusions from Figure 7 could be stated in words, but consider a Figure easier to interpret.

10) Figure captions for Figure 9 and Figure 10. Please clarify what you mean when you say that the data is binned with a range of 7 ug/m3. This is not clear

We will modify the caption of Figure 9 and Figure 10 as detailed below.

Page 19443, caption, *addition to main manuscript*:

All the mass concentrations and fractions of above species were sorted according to the hourly average NR-PM1 mass in ascending order. The solid circles represent the average value for each concentration bin with a width of 7 μg m^{-3} , and the vertical lines represent the standard deviations.