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Supplement of

Insights into characteristics, sources, and evolution of submicron aerosols during harvest seasons in the Yangtze River delta region, China

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Table S1. Summary of meteorological factors, the mean mass concentrations (μg m^{-3}) of aerosol species and OA components for three case events as marked in Fig. 1.

	Case 1	Case 2	Case 3
Duration	2 h	2 h	3 h
Meteorological factors			
WS ($m\ s^{-1}$)	2.5	1.8	1.5
WD (°)	NW	NW-N	N-NE
RH (%)	71.3	58.6	42.3
T (℃)	24.0	18.9	15.7
Aerosol species			
OA	137.0	121.3	95.0
NO_3	44.3	9.1	9.9
SO_4	16.1	6.5	4.9
NH_4	23.8	7.1	6.7
Chl	12.6	0.5	0.4
BC	19.2	16.9	7.2
K^+	16.5	1.2	1.5
OA components			
HOA + COA	2.1	79.9	58.0
BBOA	25.9	4.1	3.8
OOA-BB	101.6	19.2	16.3
OOA	6.8	2.4	4.5

Table S2. Summary of meteorological factors, the mean mass concentrations (μg m^{-3}) of aerosol species and OA components for L-BB, M-BB, and H-BB as marked in Fig. 4.

	Summer harvest			Autumn harvest		
	L-BB	M-BB	H-BB	L-BB	M-BB	H-BB
Meteorological factors						
WS ($m\ s^{-1}$)	3.0	5.1	3.1	2.5	2.5	2.8
WD (°)	NW - N E, SE	SE	NW	N - NE	NW - N	E - SE
RH (%)	72.0	69.3	73.6	53.4	60.2	54.1
T (℃)	22.3	20.5	23.6	18.0	17.3	18.5
Aerosol species						
OA	13.2	19.0	105.2	15.2	34.8	33.9
NO_3	9.5	7.8	37.9	7.6	11.1	12.6
SO_4	7.7	6.2	23.0	3.9	7.3	6.0
NH_4	6.6	5.1	20.2	5.2	8.6	8.4
Chl	0.3	0.4	9.0	0.5	1.0	1.2
BC	3.1	3.2	14.9	4.5	10.0	8.2
OA components						
HOA + COA	7.2	6.8	6.2	6.1	9.3	6.9
BBOA	3.0	6.7	76.0	3.3	9.9	13.6
OOA-BB	0.6	2.3	19.4	0.8	2.2	2.0
OOA	2.0	2.3	1.6	3.8	10.7	8.6

Table S3. Summary of mass concentrations (μg m^{-3}) of aerosol species for different clusters during the summer and autumn harvest respectively.

	Summer harvest				Autumn harvest			
	NE (25.5%)	EM (16.9%)	SEM (57.4%)	WC (5.1%)	NC (43.8%)	NEM (21.4%)	EM (24.0%)	SC (10.9%)
OA	27.1	16.2	8.9	12.2	38.0	21.0	26.0	16.0
NO_3	19.4	8.7	4.5	10.4	15.3	9.3	12.3	7.2
SO_4	8.2	7.0	5.2	9.8	6.5	5.3	7.2	3.8
NH_4	9.9	5.8	4.0	7.9	9.6	6.7	9.0	5.1
Chl	0.7	0.3	0.2	0.4	1.4	0.7	0.8	0.5
BC	6.1	3.2	1.7	3.0	10.1	6.1	7.7	4.7
HOA + COA	5.2	3.1	1.7	1.2	9.1	7.6	8.5	4.5
BBOA	0.9	1.1	0.9	0.3	3.8	1.2	1.4	1.1
OOA-BB	8.6	4.9	1.4	2.9	18.0	5.4	6.5	4.6
OOA	10.9	7.1	4.5	7.6	7.2	6.8	9.6	5.8

Table S4. Description of PMF solutions for the summer harvest. Note that the 20 different seed runs for the $Q/Q_{expected}$ values showed very little variability (Figure S6). More details are presented in Figure S6 - S10.

No. of factors	Solution Description
2	Too few factors, large residual, MS appear more mixed.
3	Much higher *m/z* 44 in BBOA factor than source spectra of fresh/primary BBOA, which is also mixed with OOA-BB.
4	**HOA + COA, BBOA, OOA-BB, and OOA factors found.**
5	HOA + COA splitting evident and mixing with BBOA. Similar spectra, diurnal cycles and time series.
6 & 7	MS appear more mixed and split. And more similar spectra, diurnal cycles and time series.

Table S5. Description of PMF solutions for the autumn harvest. Note that the 20 different seed runs for the $Q/Q_{expected}$ values also showed very little variability (Figure S11). More details are presented in Figure S11 - S15.

No. of factors	Solution Description
2	Too few factors, large residual, MS appear more mixed.
3	Much higher m/z 44 in BBOA factor than source spectra of fresh/primary BBOA, which is also mixed with OOA-BB.
4	**HOA + COA, BBOA, OOA-BB, and OOA factors found.**
5	HOA + COA splitting evident. Similar spectra, diurnal cycles and time series.
6 & 7	HOA + COA and BBOA splitting evident. And more similar spectra, diurnal cycles and time series.

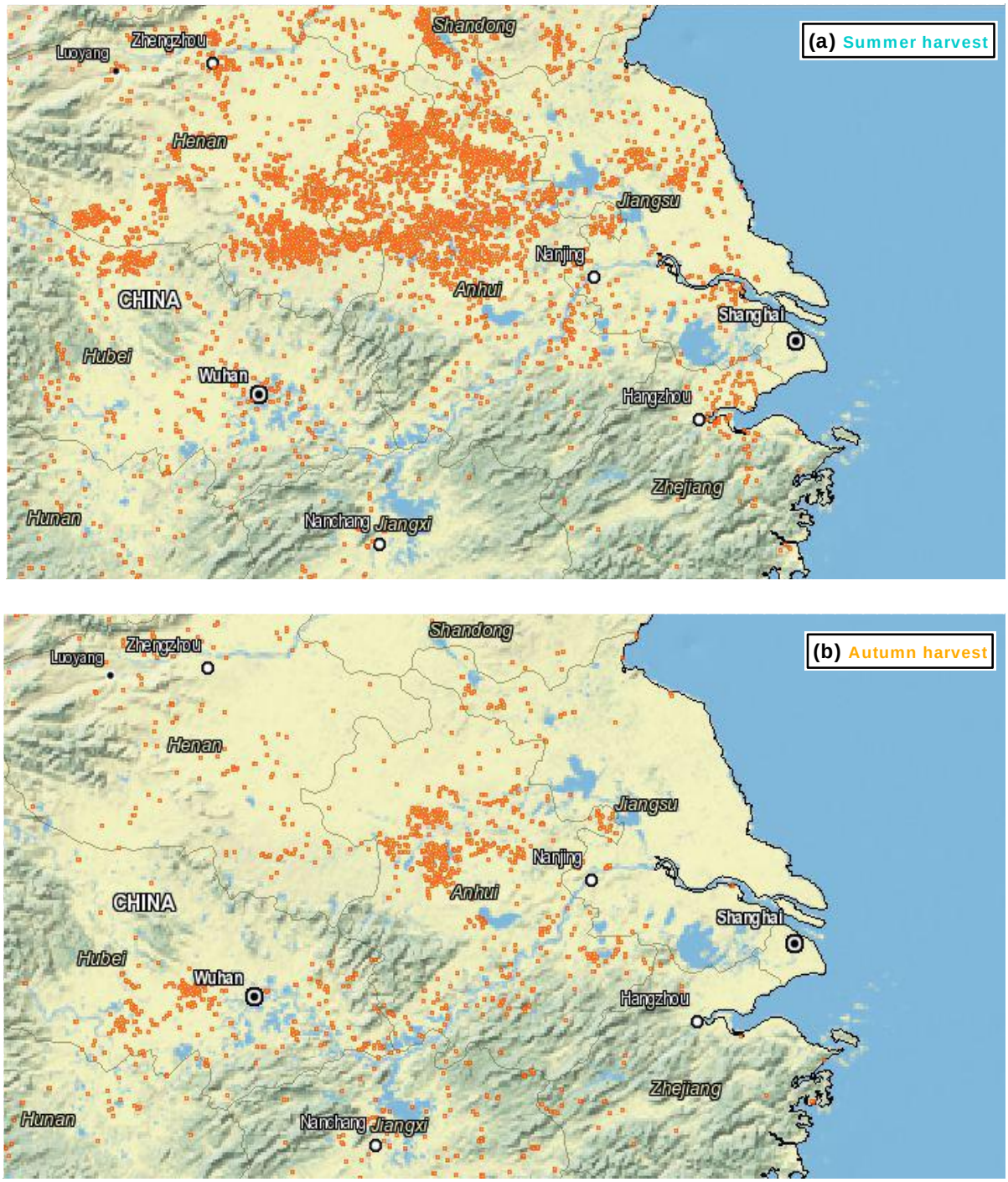

Figure S1. View of total agricultural fire locations (red dots) over YRD region detected by the remote sensing retrieval of Moderate Resolution Imaging Spectroradiometer (MODIS) mounted on NASA's Terra and Aqua satellites in 1 km pixel (a) from June 1 to 15, 2013 and (b) from October 15 to 30, 2013, respectively.

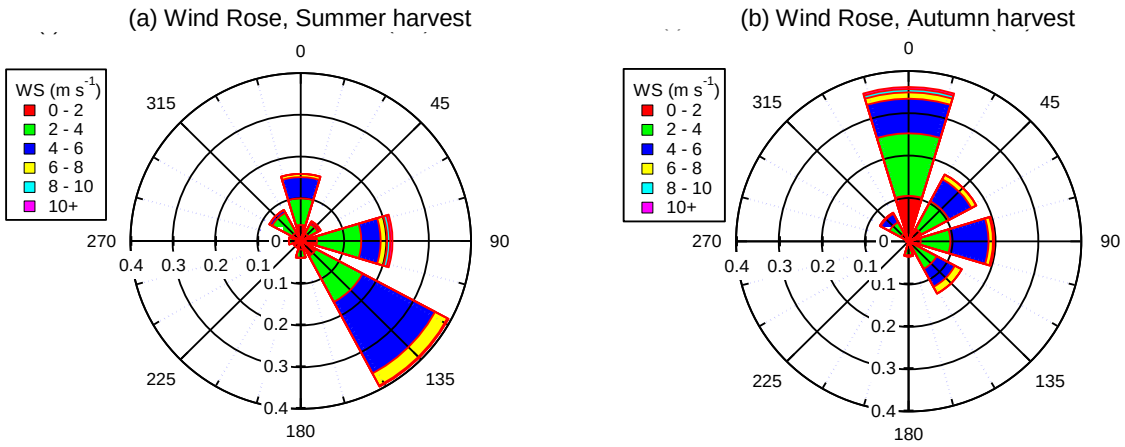

Figure S2. Frequency distributions of hourly averaged wind direction and speed during the harvest seasons.

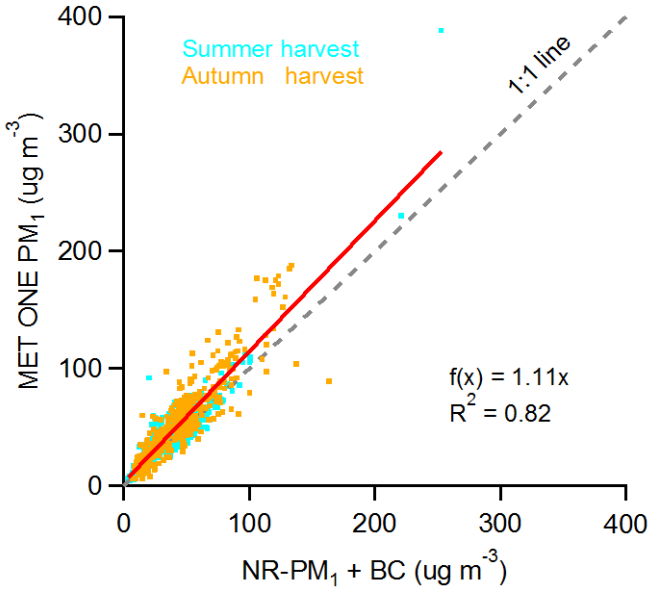

Figure S3. Correlation plot of the MET ONE PM_1 mass loadings vs. the sum of NR-PM_1 and BC mass loadings.

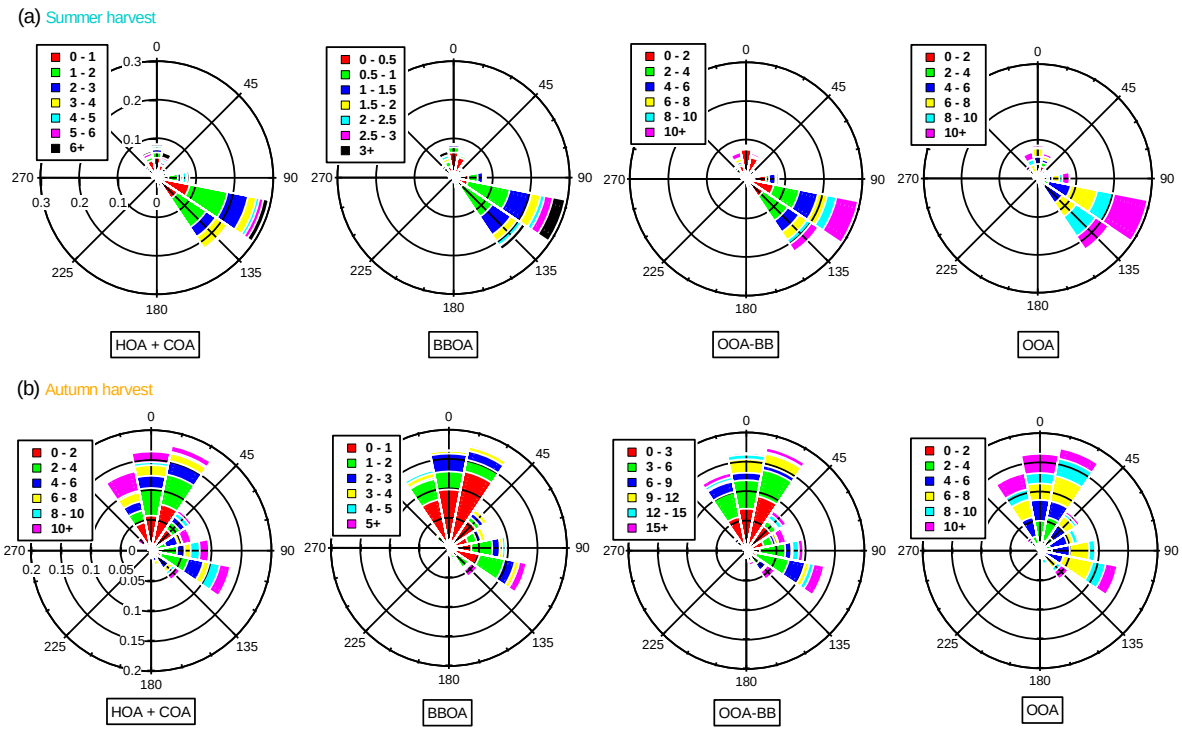

Figure S4. Wind rose plots of the frequency distribution of hourly averaged wind direction and the OA components (HOA + COA, BBOA, OOA-BB, and OOA) concentration during the harvest seasons. Colored by the OA components concentrations (μg m^{-3}). Overall, WD was from the southeast for ~40% and the east for ~20% of the summer harvest, and from the north for ~35% and the east for ~20% of the autumn harvest.

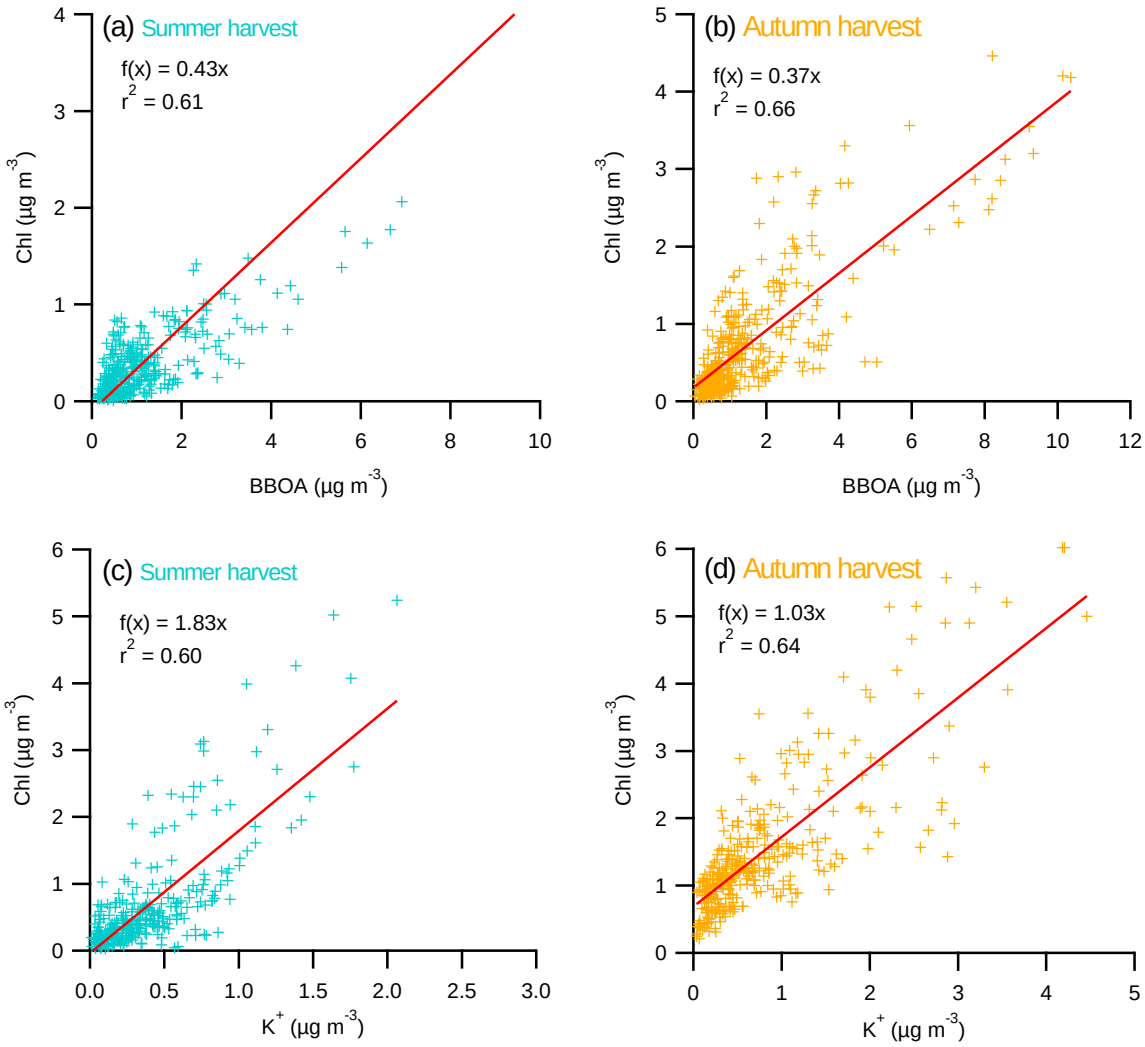

Figure S5. Correlation plots of chloride (Chl) vs. BBOA and Chl vs. K^+ during the summer and autumn harvest respectively.

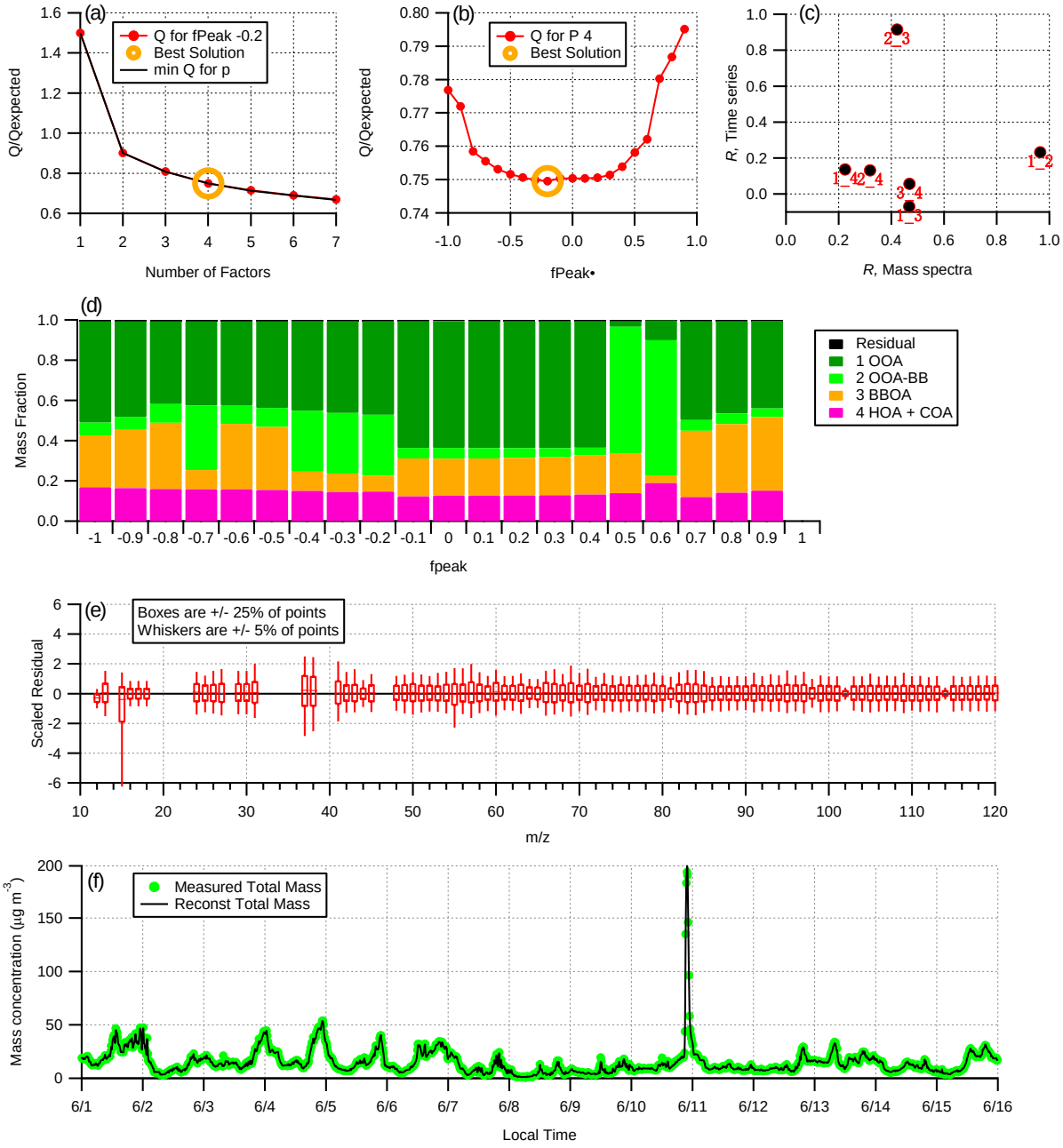

Figure S6. Summary of the evaluation of the PMF analysis of the unified organic aerosols dataset during the summer harvest. (a) $Q/Q_{expected}$ as a function of number of factors (*p*) selected for PMF modeling; (b) $Q/Q_{expected}$ as a function of fPeak; (c) correlations of mass spectra and time series among PMF factors; (d) mass fraction of each factor vs. fPeak values for the 4-factor solution. The residual fraction for different fpeak values is ~0.4%; the numbers refer to the OA factors in (d); (e) the box and whiskers plot showing the distributions of scaled residuals for each fragment ion; (f) time series of the measured mass concentration and the reconstructed mass.

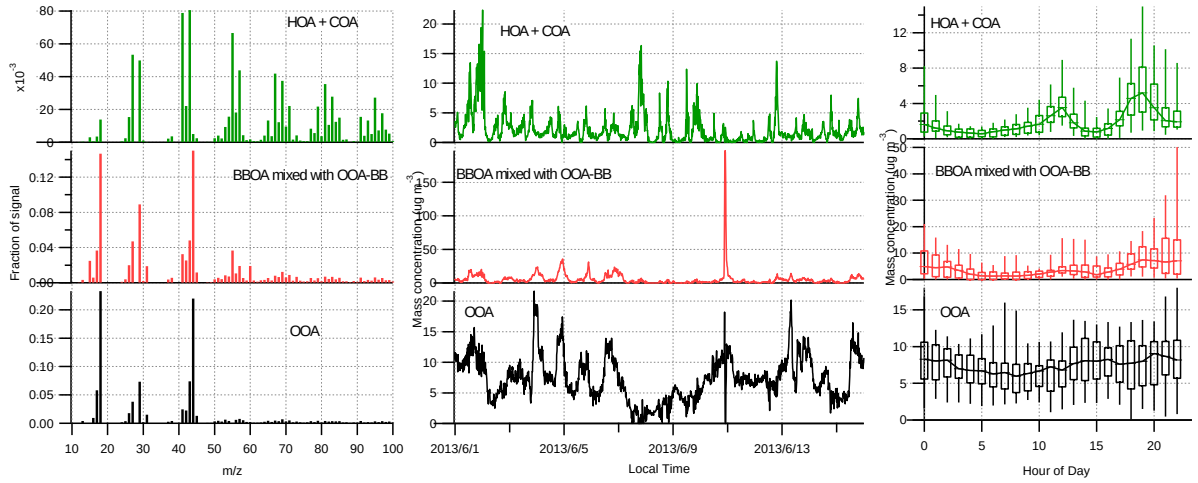

Figure S7. Mass spectra, time series and diurnal plots associated with the 3 factors solution for the summer harvest.

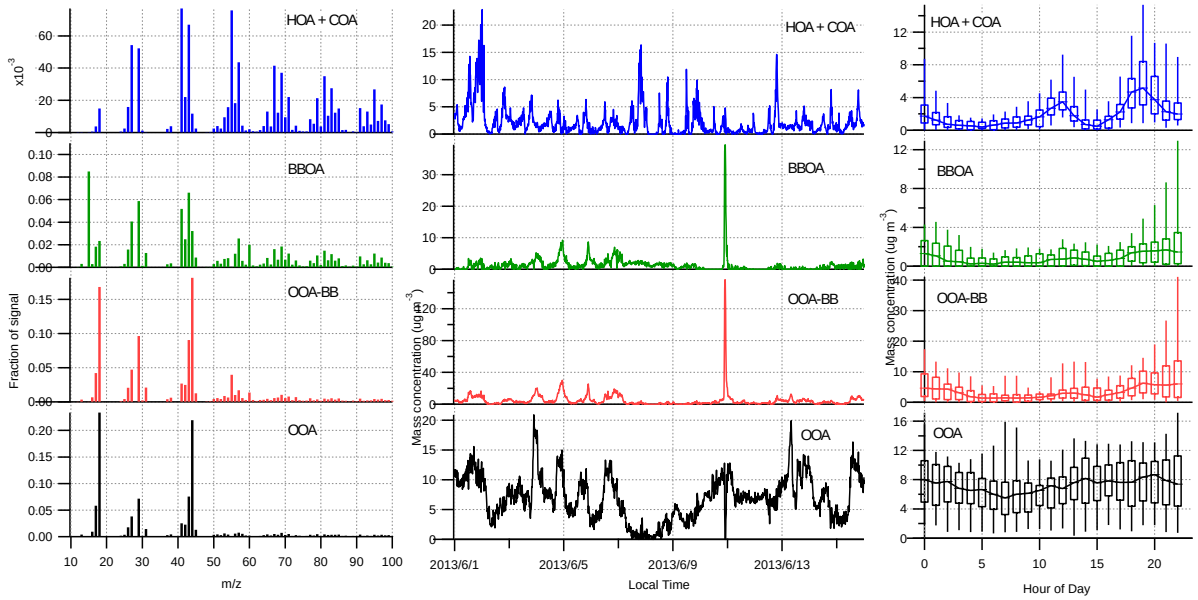

Figure S8. Mass spectra, time series and diurnal plots associated with the 4 factors solution for the summer harvest.

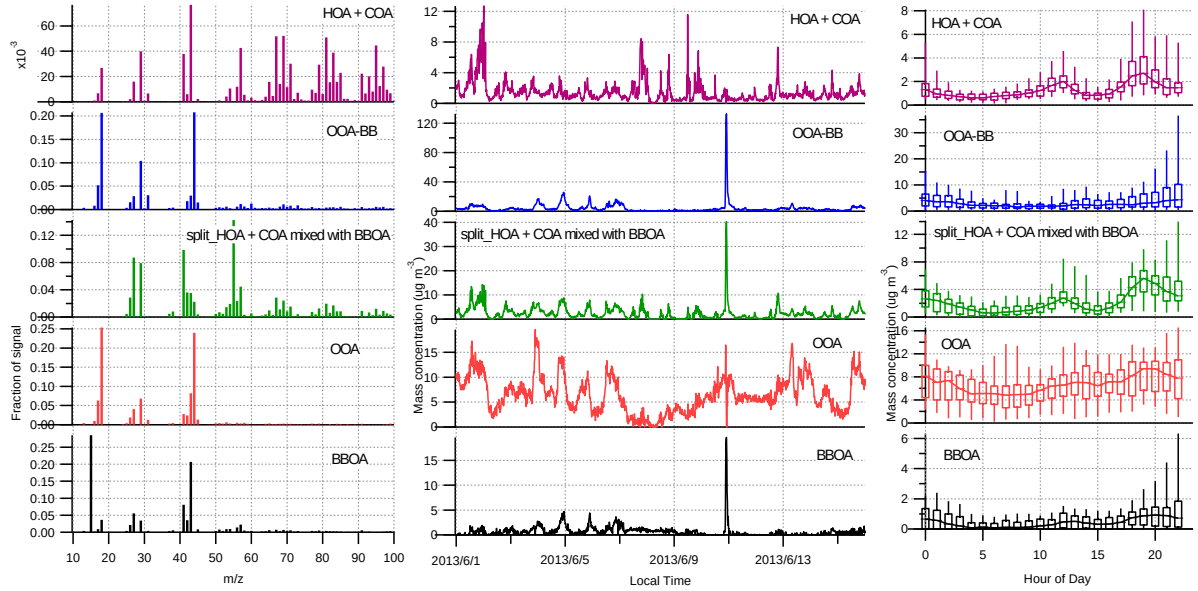

Figure S9. Mass spectra, time series and diurnal plots associated with the 5 factors solution for the summer harvest.

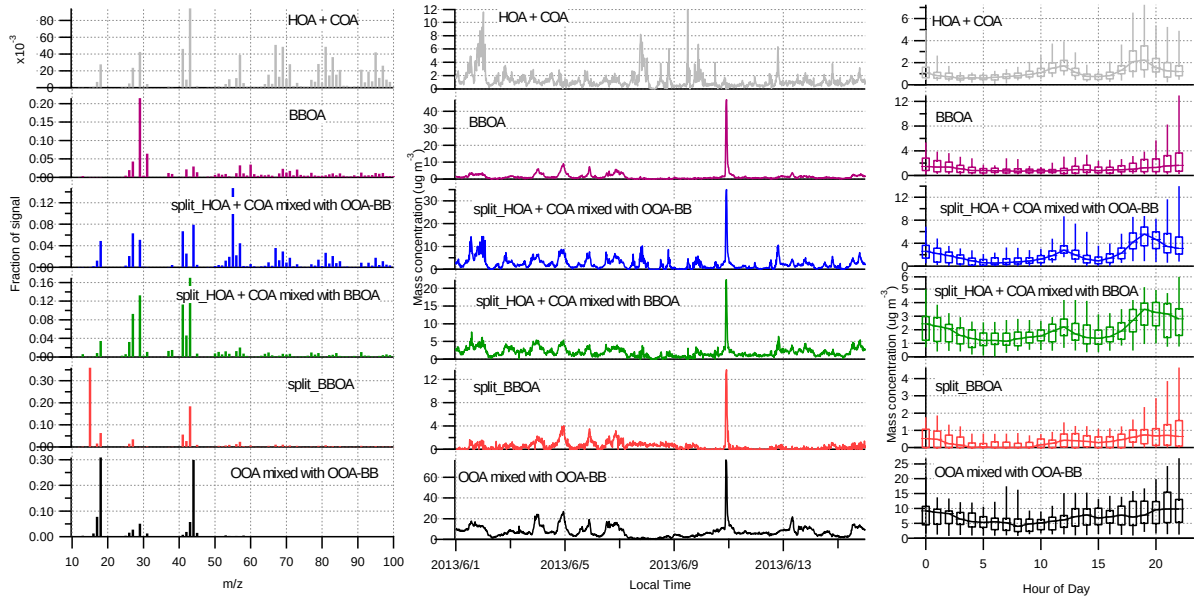

Figure S10. Mass spectra, time series and diurnal plots associated with the 6 factors solution for the summer harvest.

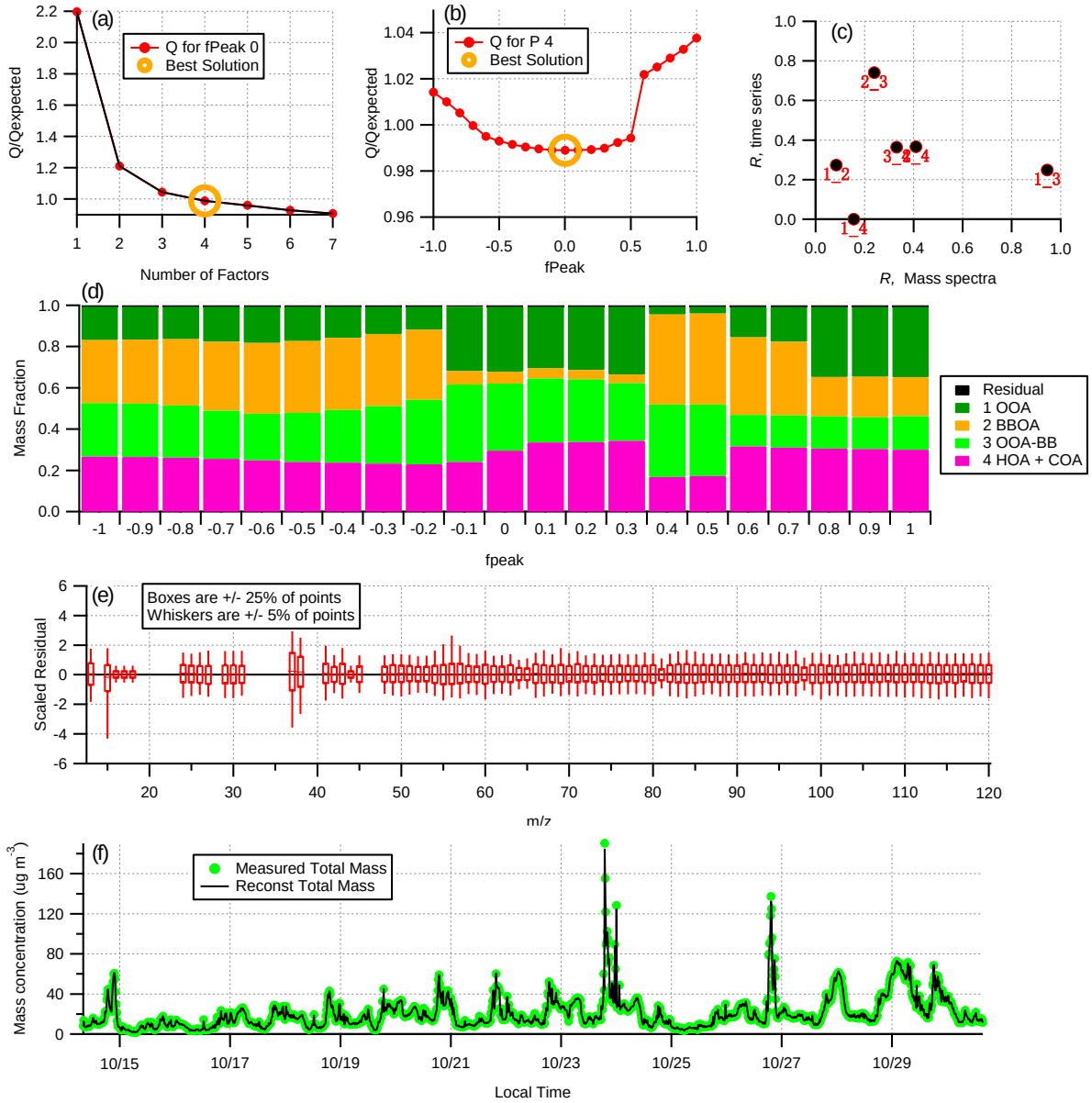

Figure S11. Summary of the evaluation of the PMF analysis of the unified organic aerosols dataset during the autumn harvest. (a) $Q/Q_{expected}$ as a function of number of factors (p) selected for PMF modeling; (b) $Q/Q_{expected}$ as a function of fPeak; (c) correlations of mass spectra and time series among PMF factors; (d) mass fraction of each factor vs. fPeak values for the 4-factor solution. The residual fraction for different fpeak values is ~0.5%; the numbers refer to the OA factors in (d); (e) the box and whiskers plot showing the distributions of scaled residuals for each fragment ion; (f) time series of the measured mass concentration and the reconstructed mass.

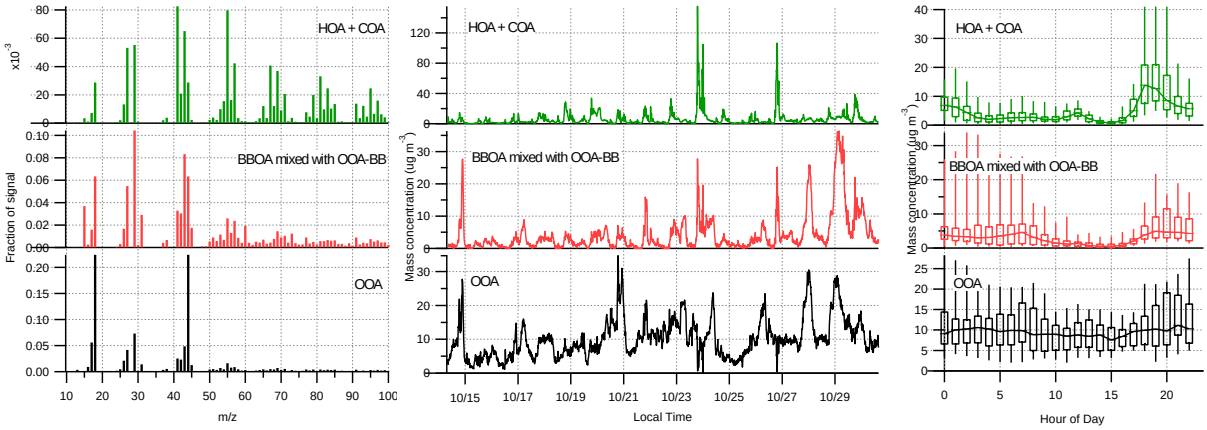

Figure S12. Mass spectra, time series and diurnal plots associated with the 3 factors solution for the autumn harvest.

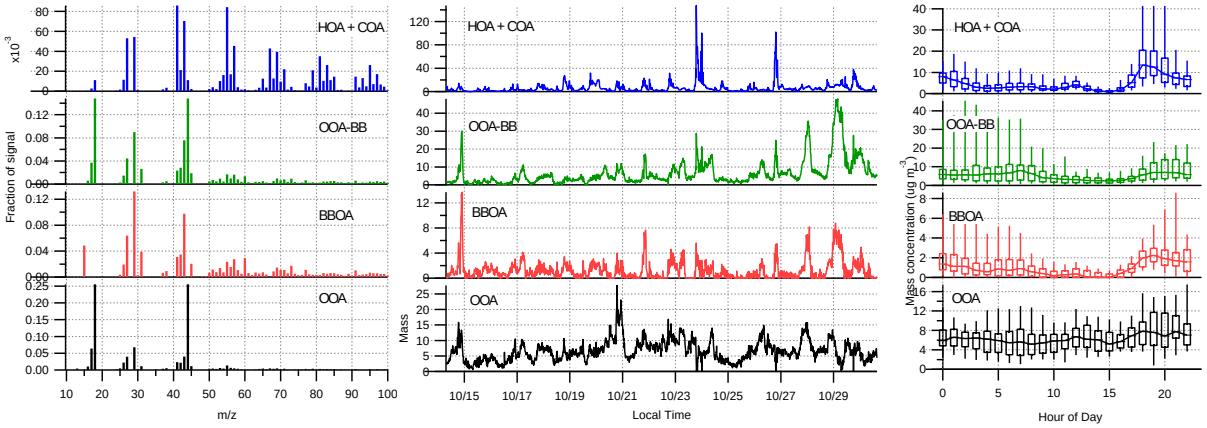

Figure S13. Mass spectra, time series and diurnal plots associated with the 4 factors solution for the autumn harvest.

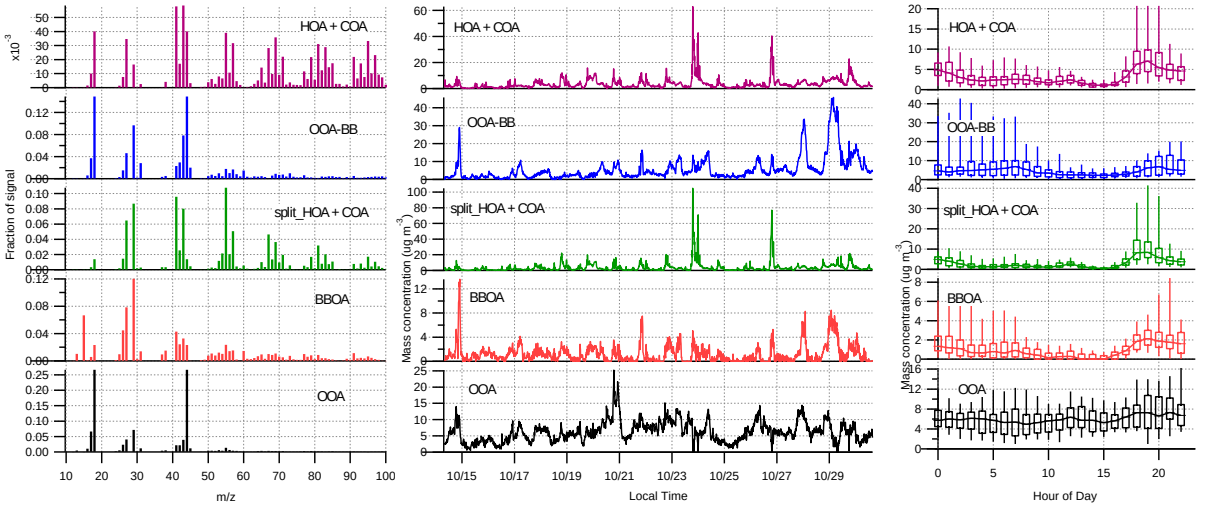

Figure S14. Mass spectra, time series and diurnal plots associated with the 5 factors solution for the autumn harvest.

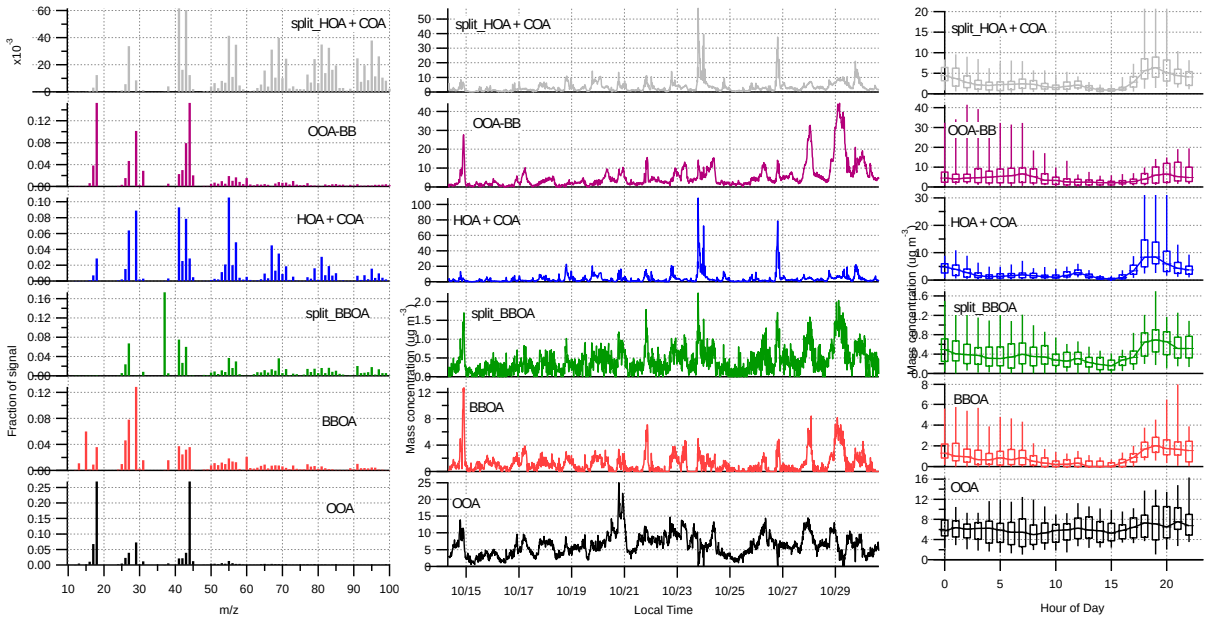

Figure S15. Mass spectra, time series and diurnal plots associated with the 5 factors solution for the autumn harvest.

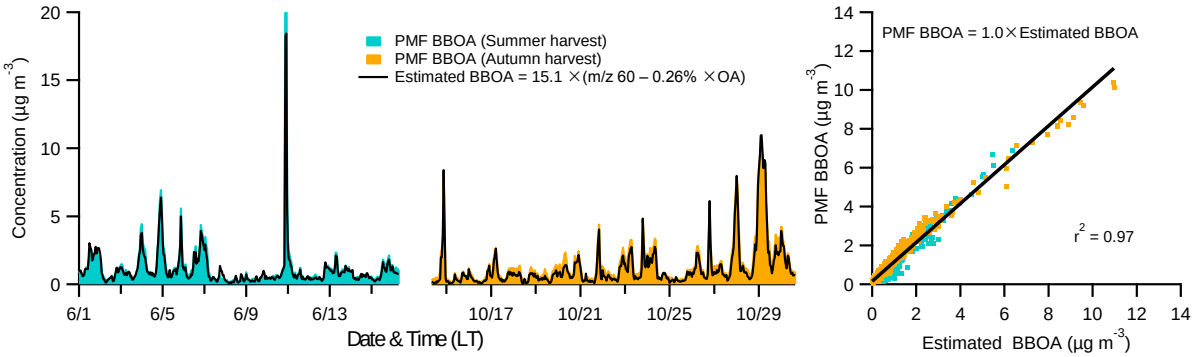

Figure S16. Time series of fresh BBOA identified by PMF (PMF BBOA) and estimated BBOA during the harvest seasons, as well as correlation plot of estimated BBOA vs. PMF BBOA. Note that the highest values for case 1 (Fig. 1c) during the summer harvest have been removed for fitting.