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Seasonal variations in the highly time-resolved aerosol composition, sources and chemical processes of background submicron particles in the North China Plain

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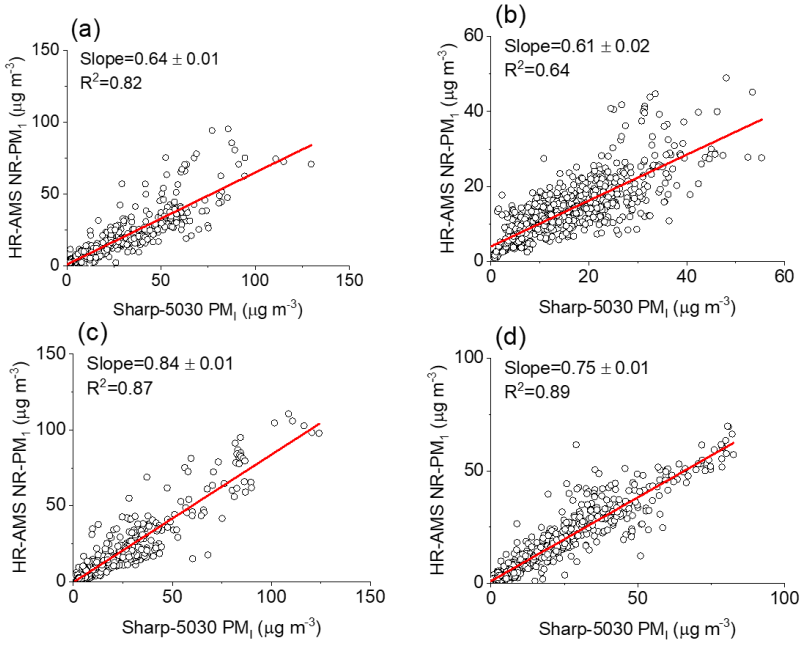

Figure S1. Scatter plots of the mass concentration of NR-PM_1 vs. total PM_1 measured by Sharp-5030 in **(a)** spring, **(b)** summer, **(c)** autumn and **(d)** winter.

Determination of the PMF and ME-2 solution

Spring:

Factor number from one to eight were selected to run in the PMF model. For the spring observation, there was no POA factor appeared in the two- to four- factor solution. A POA factor appeared in the five-factor solution and diagnostic plots of the PMF analysis were shown in Fig. S2. The mass profile of the POA factor had some similarity with that of HOA and CCOA. The correlation coefficient between POA and NO_x was 0.58, and that between POA and chloride was 0.78, suggesting a significant contribution of coal combustion and traffic-related sources to the POA factor in Xinglong. Previous studies found that HOA and CCOA showed remarkably similar mass spectrum patterns when *m/z* was below 120 (Sun et al., 2016; Sun et al., 2018), which was sometimes difficult to be separated by PMF analysis, so FFOA could be considered as a combined factor of HOA and CCOA (Sun et al., 2018). Therefore, the POA factor was identified as FFOA.

As shown in Fig. S4, in the five-factor solution, factor 1, factor 2 and factor 4 had similar mass spectra, time series and O/C ratios (0.87-0.96). It was unclear if the three OOA components represent distinct sources or chemical types. Therefore, it was over split by one OOA factor. Another OOA factor (factor 5) had different mass spectra and lower O/C ratio, suggesting different formation mechanism. Therefore, we constrained the FFOA profiles separated by the five-factor solution of PMF

analysis in spring. As a result, three OA factors, including FFOA, LO-OOA and MO-OOA, were identified with ME-2 analysis in spring. The mass spectra, time series, and diurnal variations of ME-2 result were shown in Fig. S7.

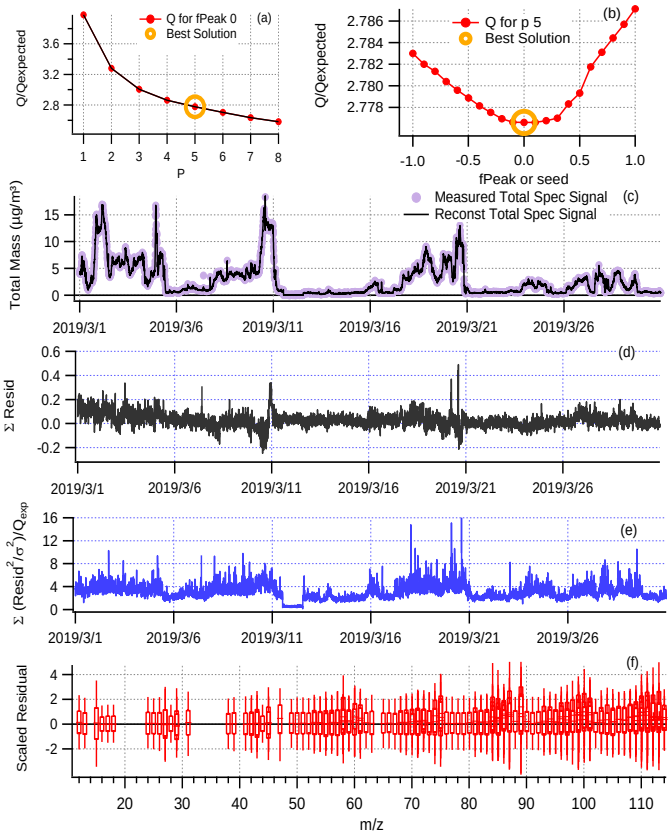

Figure S2. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the spring observation.

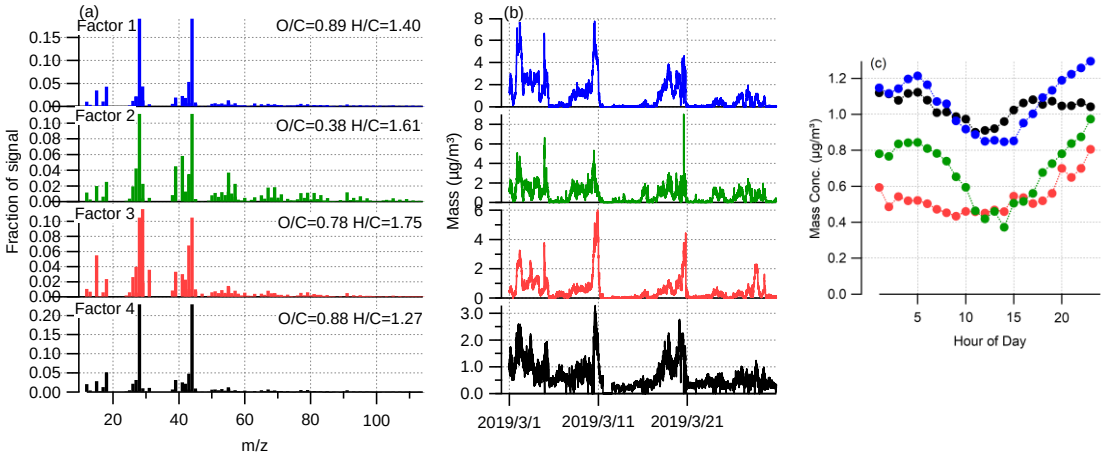

Figure S3. The mass spectra, time series, and diurnal variations of four-factor solution of PMF analysis for the spring observation.

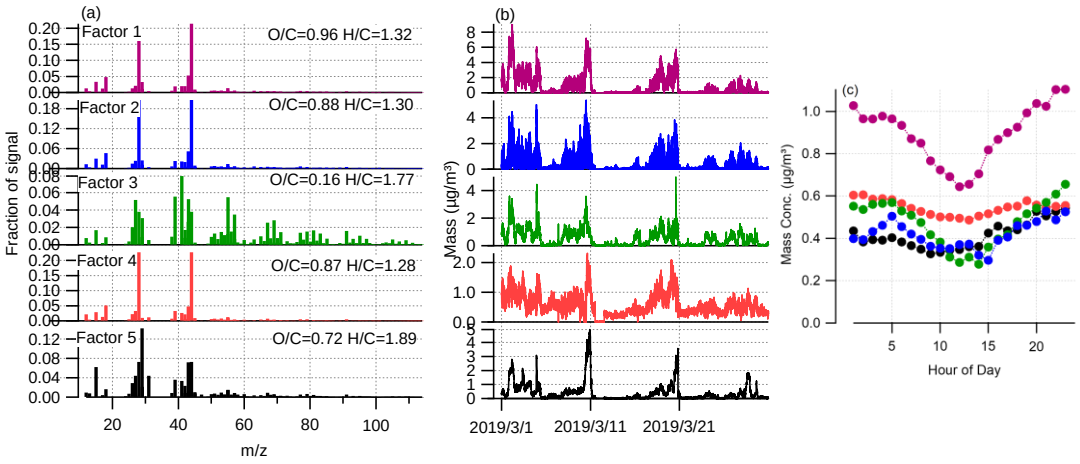

Figure S4. The mass spectra, time series, and diurnal variations of five-factor solution of PMF analysis for the spring observation.

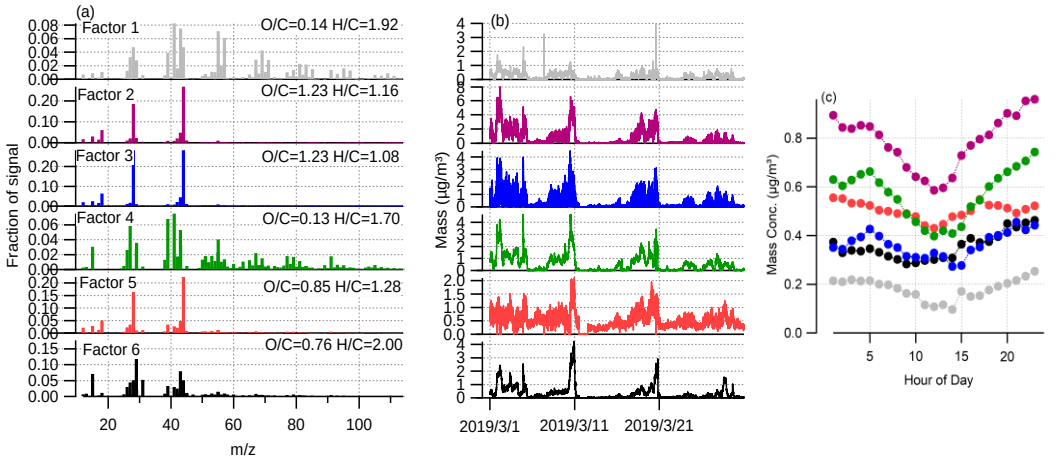

Figure S5. The mass spectra, time series, and diurnal variations of six-factor solution of PMF analysis for the spring observation.

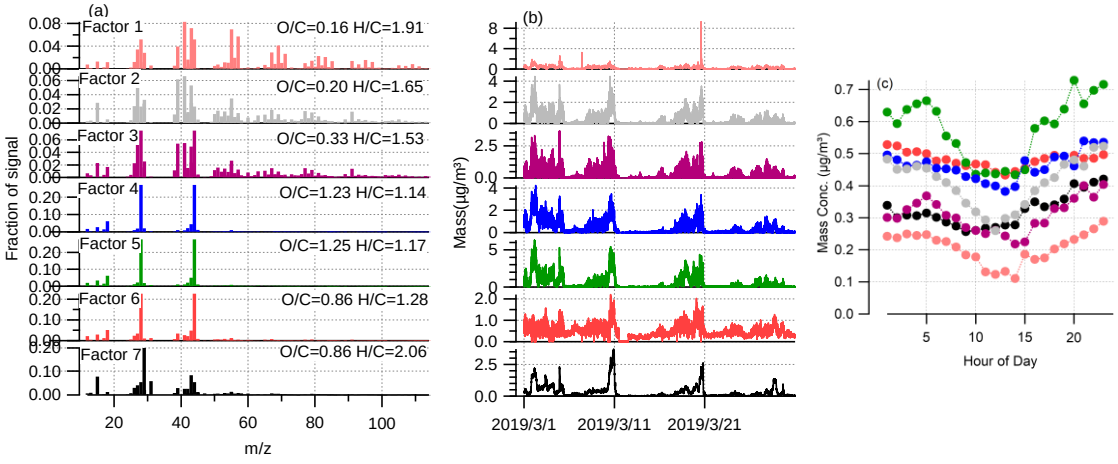

Figure S6. The mass spectra, time series, and diurnal variations of seven-factor solution of PMF analysis for the spring observation.

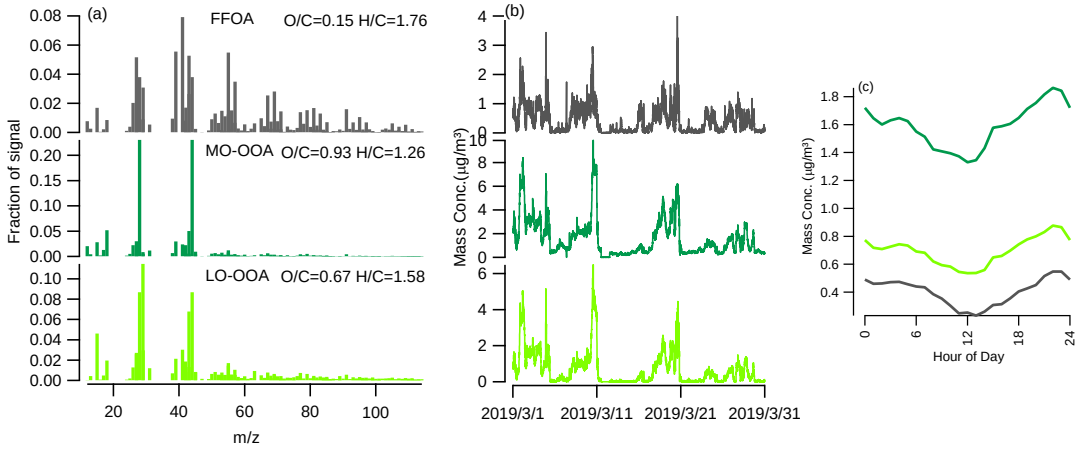

Figure S7. The mass spectra, time series, and diurnal variations of ME-2 analysis for the spring observation.

Summer:

For the summer observation, the two-factor, f_{peak}=0 solution was selected as the optimum solution. The diagnostic plots of the PMF analysis were shown in Fig. S8. The two OA factors are more oxidized (MO-OOA) and less oxidized OOA (LO-OOA). The mass spectrum, time series and diurnal variations of OA factors were different. The O/C of the two factors were 0.58 and 0.93, respectively. No POA factor appeared in the two- to nine-factor solutions and OOA was over spilt in the three- to nine-factor solutions. The detailed information on how to select the optimum PMF solution can be found in Figure S9-S12 and Table S1.

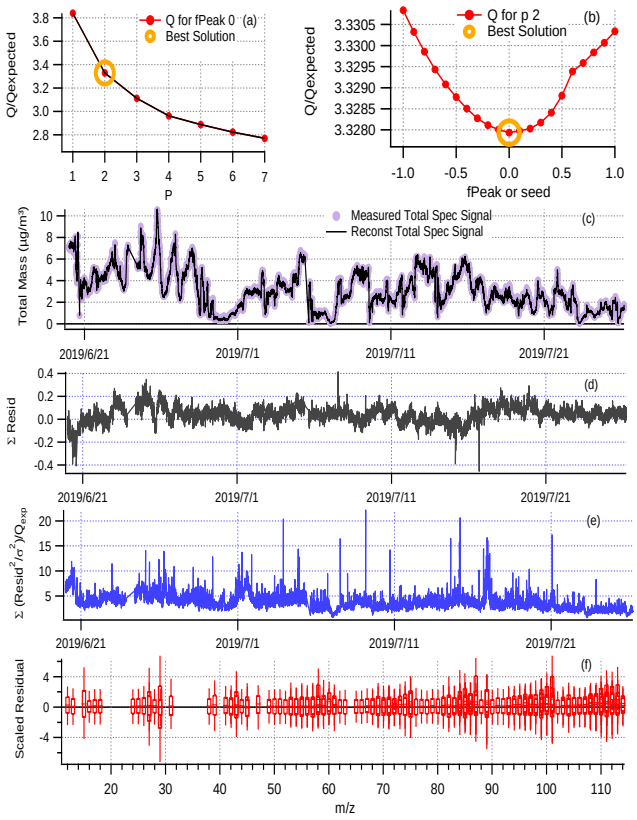

Figure S8. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the summer observation.

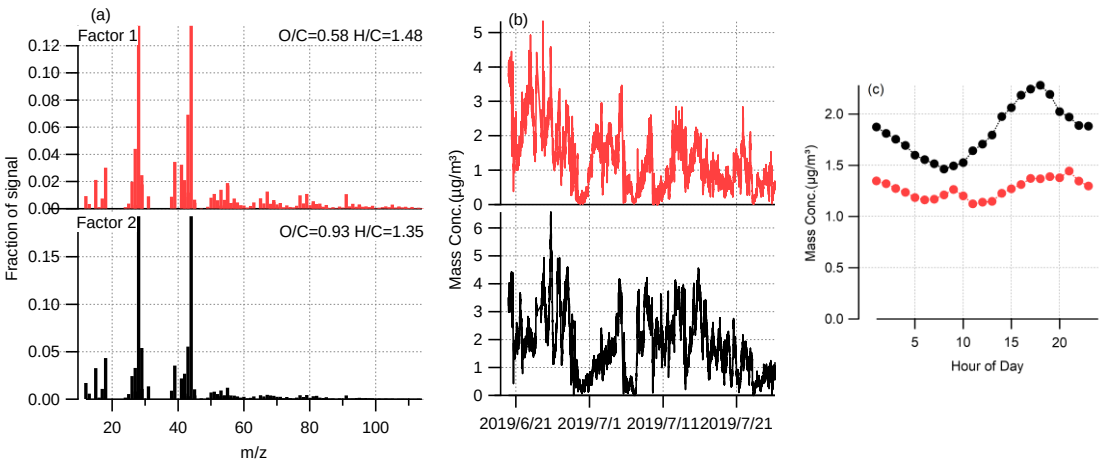

Figure S9. The mass spectra, time series, and diurnal variations of two-factor solution of PMF analysis for the summer observation.

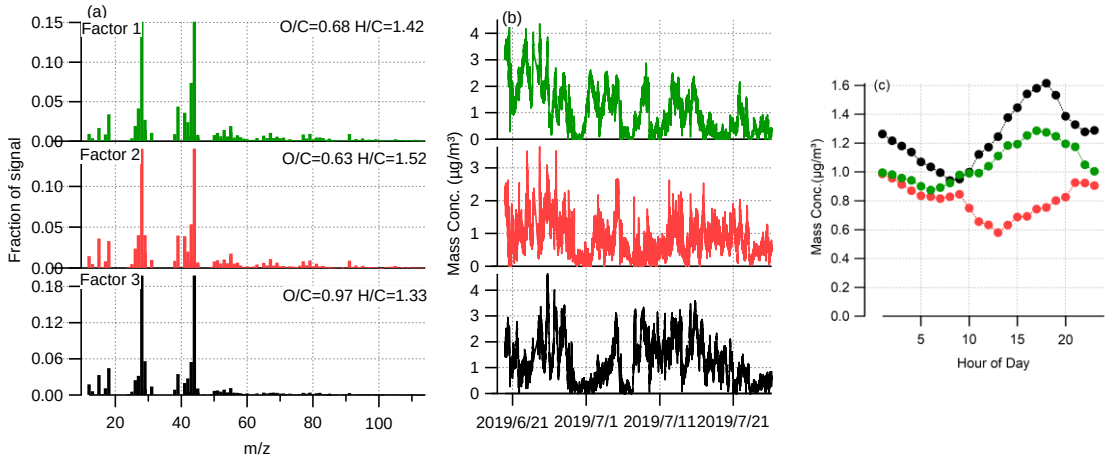

Figure S10. The mass spectra, time series, and diurnal variations of three-factor solution of PMF analysis for the summer observation.

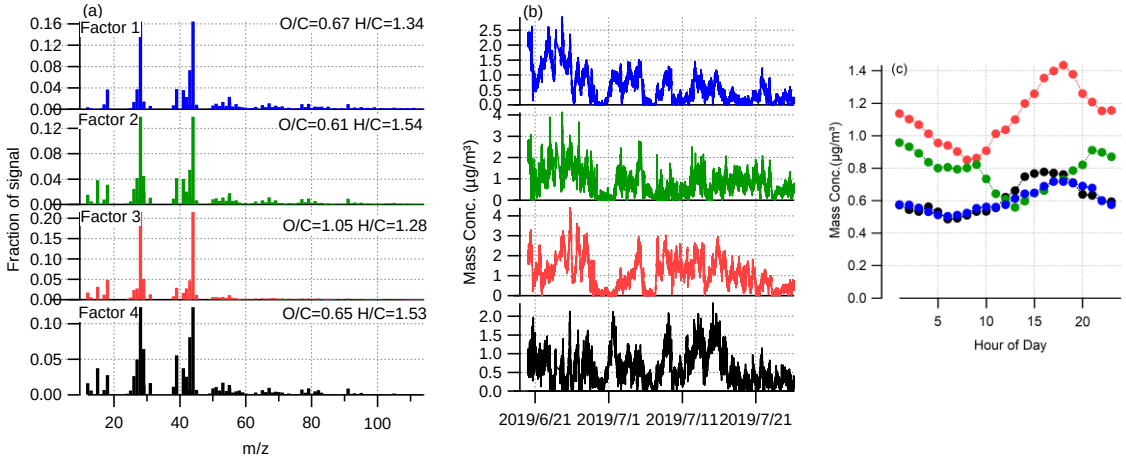

Figure S11. The mass spectra, time series, and diurnal variations of four-factor solution of PMF analysis for the summer observation.

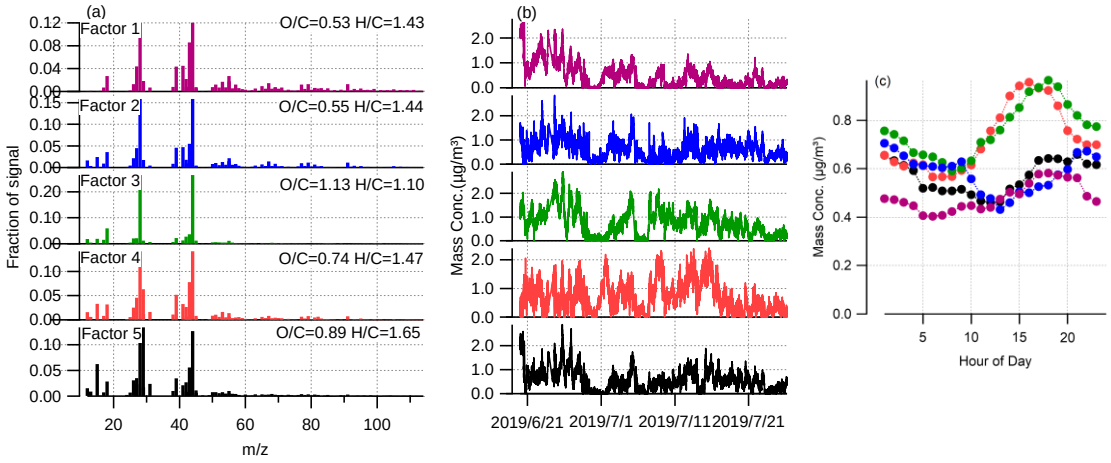

Figure S12. The mass spectra, time series, and diurnal variations of five-factor solution of PMF analysis for the summer observation.

Table S1 Descriptions of PMF solutions for the summer observation in Beijing.

Factor number	f_{peak}	Q/Q_{exp}	Solution Description
1	0	3.83	Too few factors, large residuals at time periods and key *m/z*'s
2	**0**	**3.32**	**Optimum solution for the PMF analysis (MO-OOA and LO-OOA). The mass spectrum, time series and diurnal variations of OA factors were different. The O/C of the two factors were 0.58 and 0.93, respectively.**
3-9	0	3.11	Factor split. Take 3 factor number solution as an example, factor 2 was similar to the factor 2 which resolved in the two-factor solution with similar mass spectrum, time series, diurnal variation and O/C ratios. Factor 1 and factor 3 were likely over split with similar time series and different mass spectrum. However, it was difficult to explain if they represent distinct sources or chemical types.

Autumn:

The solution of the PMF analysis for the autumn observation was similar with that in spring. A POA factor appeared until the seven-factor solution and OOA was over-split. The POA factor was also identified as FFOA. The diagnostic plots of the PMF analysis were shown in Fig. S13. We constrained the POA profile separated by the seven-factor solution of PMF analysis by ME-2 analysis. As a result, three OA factors, including FFOA, LO-OOA and MO-OOA, were identified with ME-2 analysis in autumn. The mass spectra, time series, and diurnal variations of ME-2 result were shown in Fig. S16.

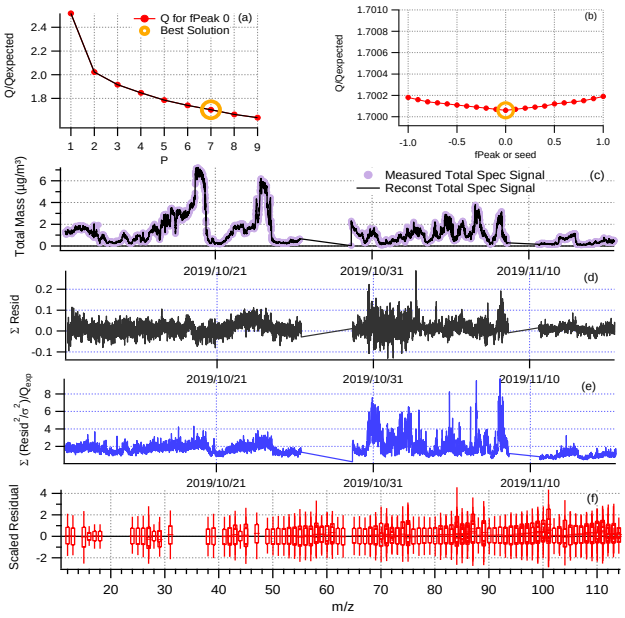

Figure S13. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the autumn observation.

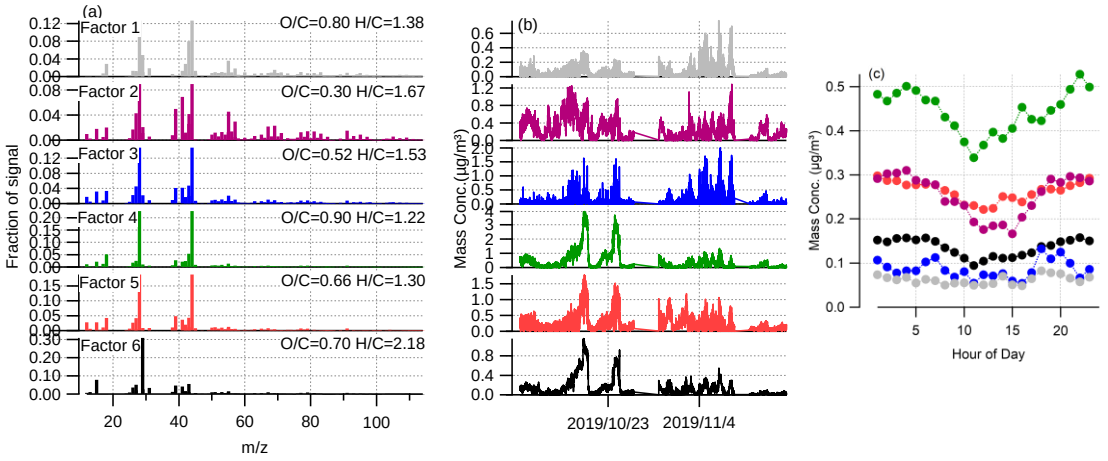

Figure S14. The mass spectra, time series, and diurnal variations of six-factor solution of PMF analysis for the autumn observation.

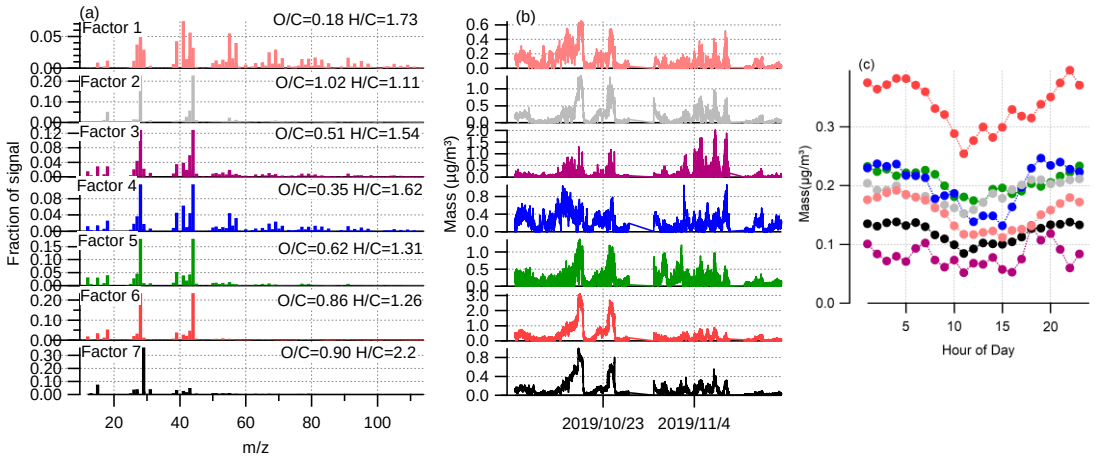

Figure S15. The mass spectra, time series, and diurnal variations of seven-factor solution of PMF analysis for the autumn observation.

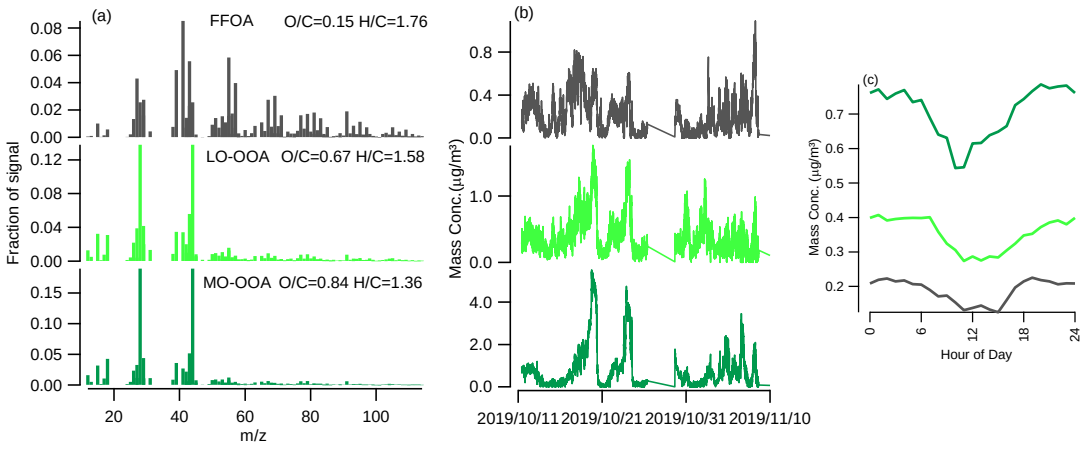

Figure S16. The mass spectra, time series, and diurnal variations of ME-2 analysis for the autumn observation.

Winter:

For the winter observation, the 3-factor, f_{peak}=0 solution was selected as the optimum solution. When OA was separated into four factors, OOA was also split into three factors (Fig. S19). In the four-factor solution, factor 2 was similar to factor 2 which was resolved in the three-factor solution with similar mass spectra, time series, diurnal variation and O/C ratios. However, factor 3 and factor 4 in the four-factor solution had similar O/C ratios, time series and diurnal variation. It was unclear if the two OOA components represent distinct sources or chemical types. When more than five factors, OOA decomposed into three or more factors. Thus, two OOA factors were combined into total OOA for further analysis. The correlation coefficient between the POA factor resolved in the three-factor solution and NO_x was 0.73, and that between POA and chloride was 0.61. Meanwhile, the mass profile of the POA factor showed similarities with that of HOA and CCOA. Therefore, the POA factor was identified as FFOA and the three-factor solution (FFOA, LO-OOA and MO-OOA) from PMF analysis was good enough in winter. The detailed information on how to select the optimum PMF solution can be found in FigureS18-S20 and Table S2.

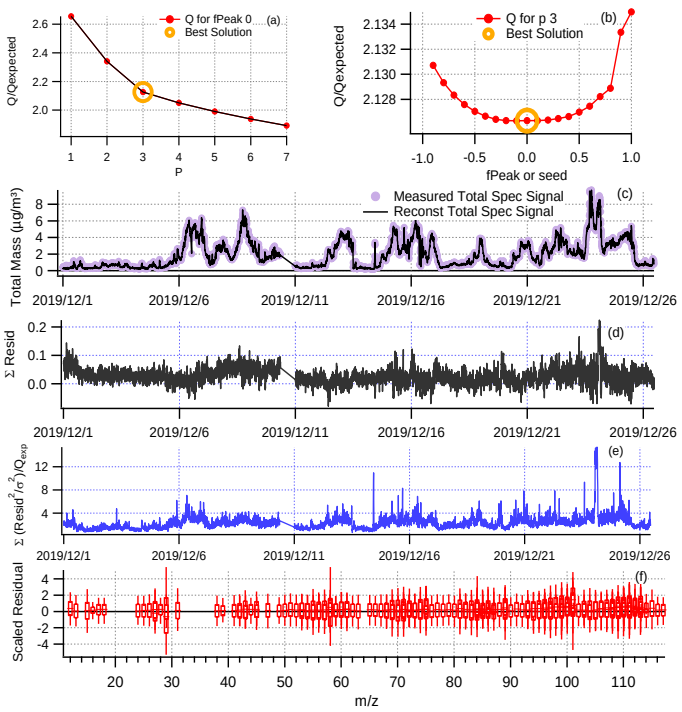

Figure S17. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the winter observation.

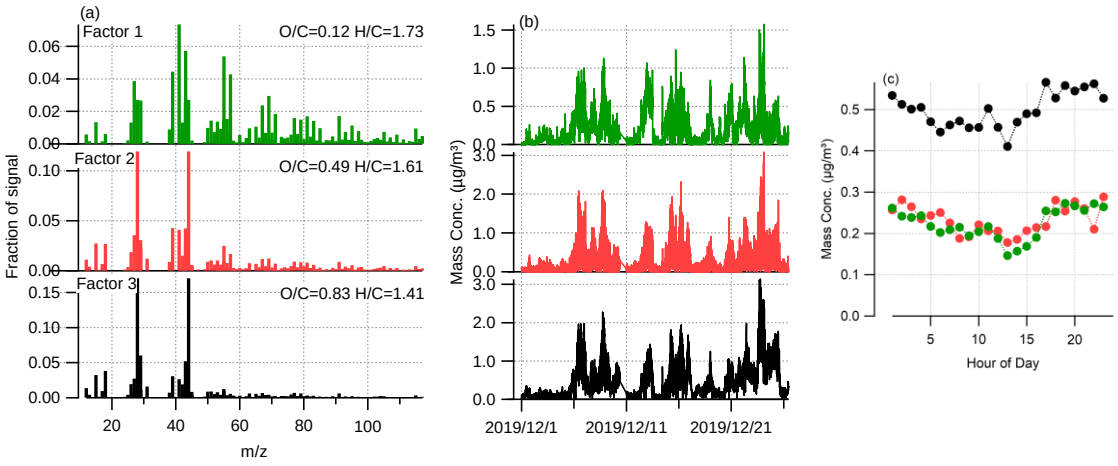

Figure S18. The mass spectra, time series, and diurnal variations of three-factor solution of PMF analysis for the winter observation.

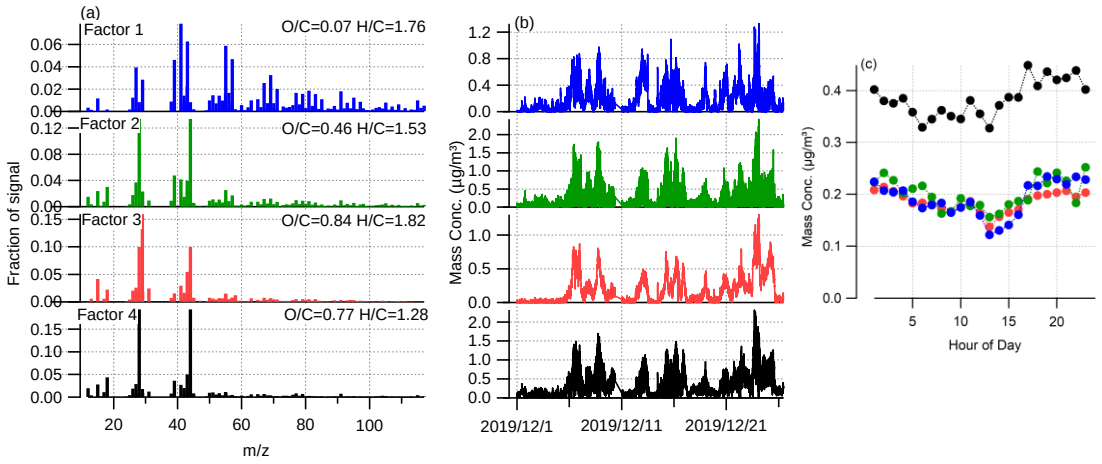

Figure S19. The mass spectra, time series, and diurnal variations of four-factor solution of PMF analysis for the winter observation.

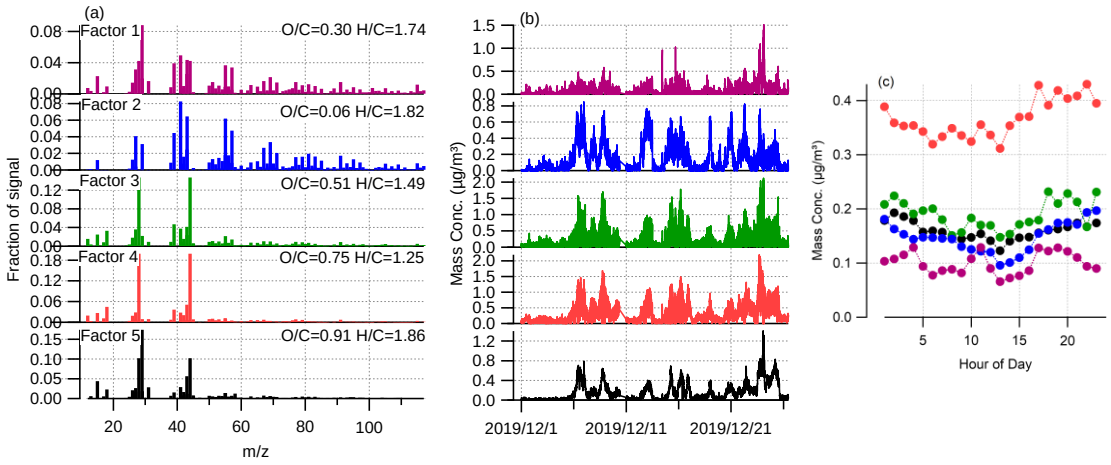

Figure S20. The mass spectra, time series, and diurnal variations of five-factor solution of PMF analysis for the winter observation.

Table S2 Descriptions of PMF solutions for the winter observation in Beijing.

Factor number	f_{peak}	Q/Q_{exp}	Solution Description
1	0	2.65	Too few factors, large residuals at time periods and key *m/z*'s.
2	0	2.34	Too few factors, POA was mixed with OOA.
3	**0**	**2.12**	**Optimum solution for the PMF analysis (FFOA, MO-OOA and LO-OOA). The mass spectrum and time series of the two OOA factors were different. The O/C of the two factors were 0.49 and 0.83, respectively. Thus, two OOA factors were for further analysis. The correlation**

			coefficient between POA and NO_x was 0.73, and that between POA and chloride was 0.61. Meanwhile, the mass profile of the POA factor had similarities with that of HOA and CCOA. Therefore, the POA factor was identified to FFOA.
4-7	0	1.89-1.98	Factor split. Take four factor number solution as an example, factor 2 was similar to the factor 2 which resolved in the two-factor solution with similar mass spectrum, time series, diurnal variation and O/C ratios. Factor 1 and factor 3 were likely over split with similar time series and different mass spectrum. However, it was difficult to explain if they represent distinct sources or chemical types.

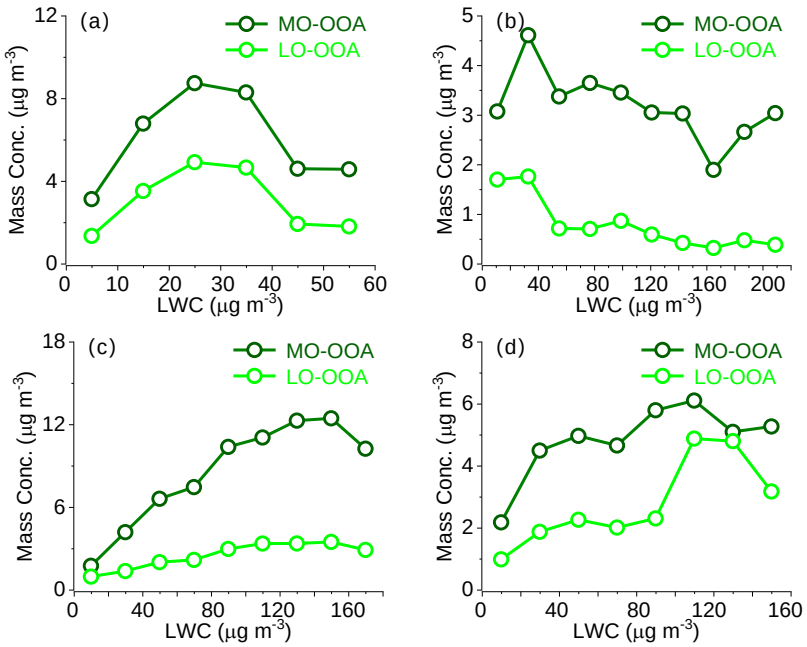

Figure S21. Variations in the mass concentrations of LO-OOA and MO-OOA as a function of LWC in **(a)** spring, **(b)** summer, **(c)** autumn and **(d)** winter.