Supplementary Information

Characterization of submicron aerosols during a month of serious pollution in Beijing, 2013

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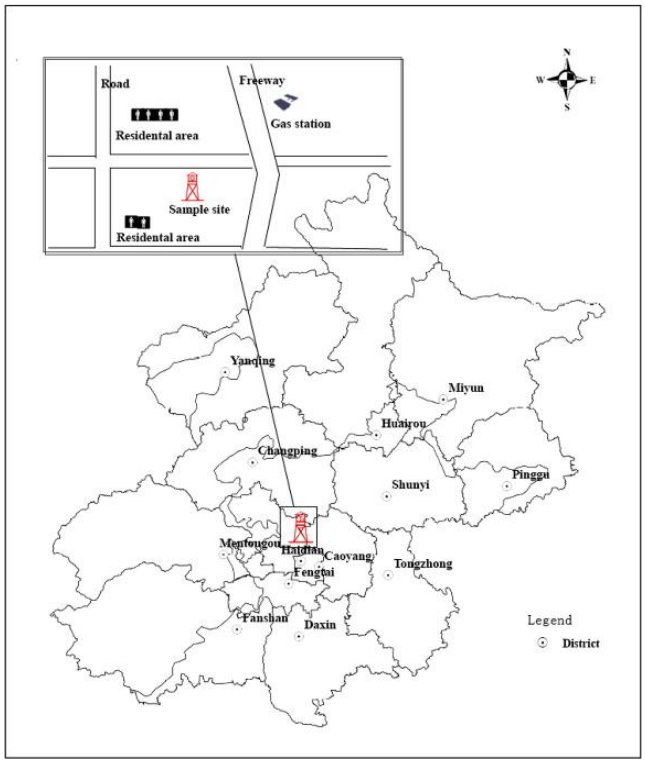


Fig. S-1. The location of the monitoring site in Beijing

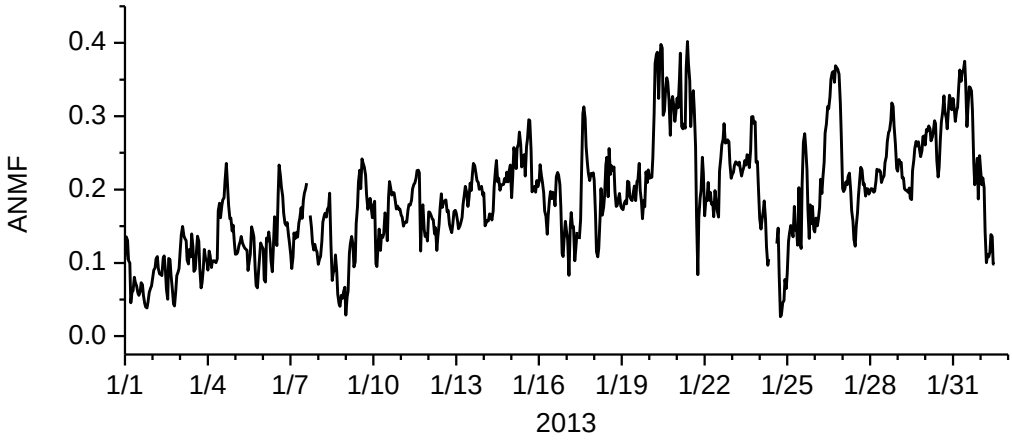
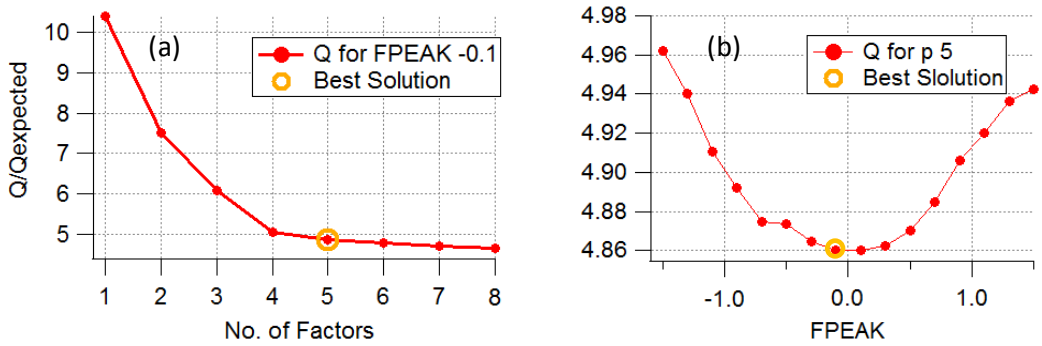
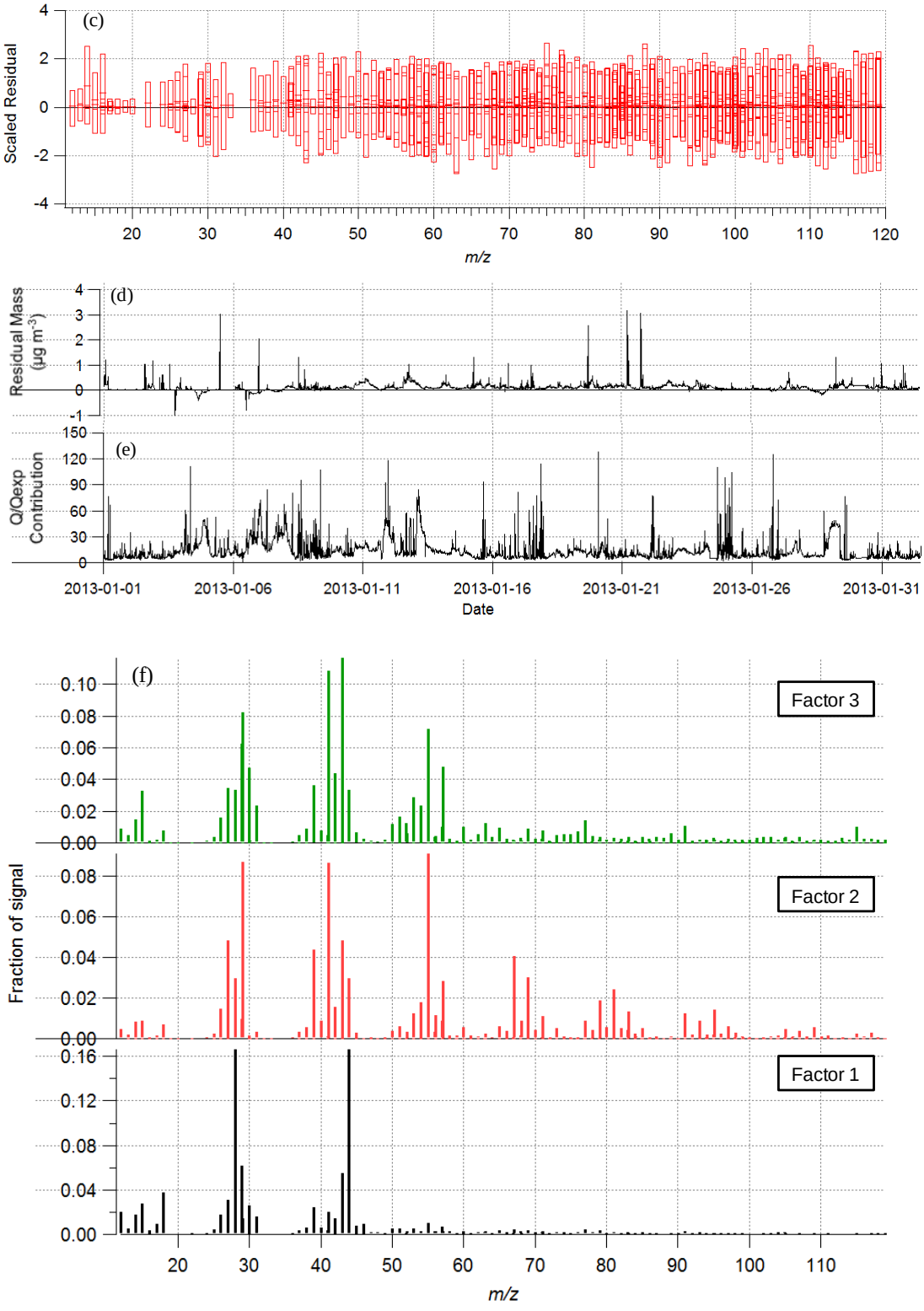
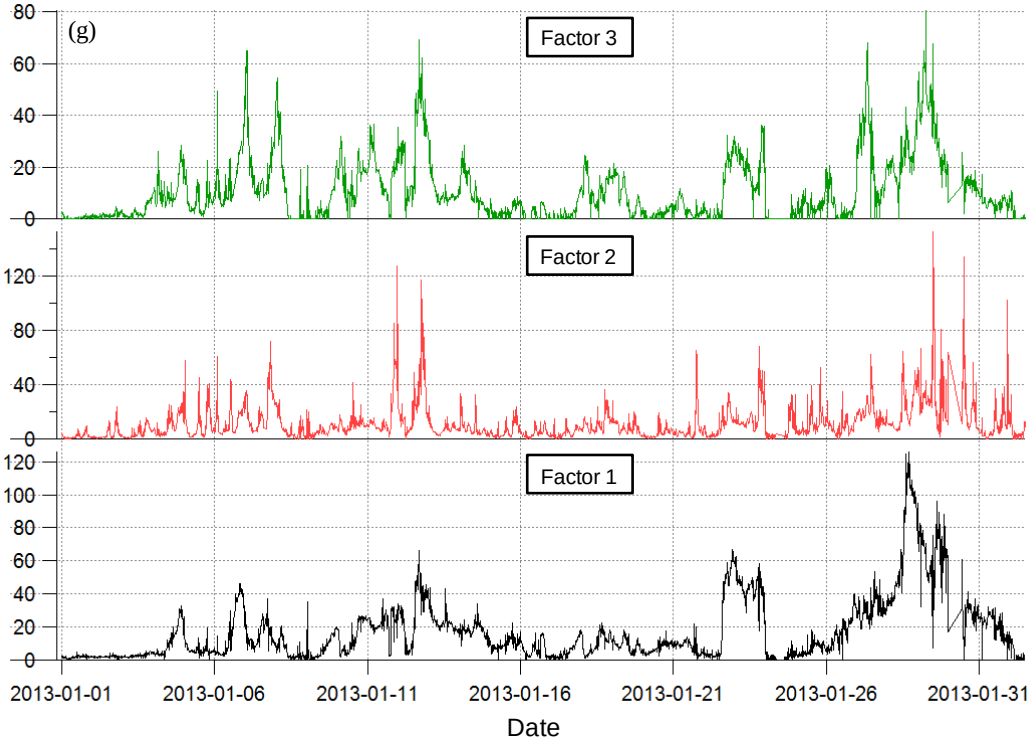
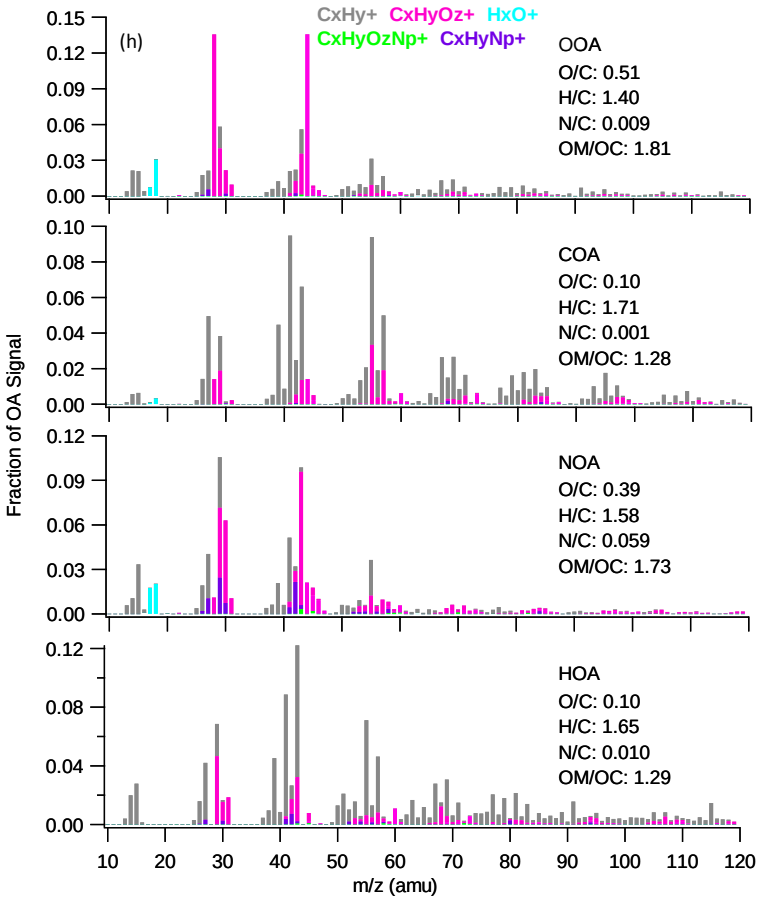


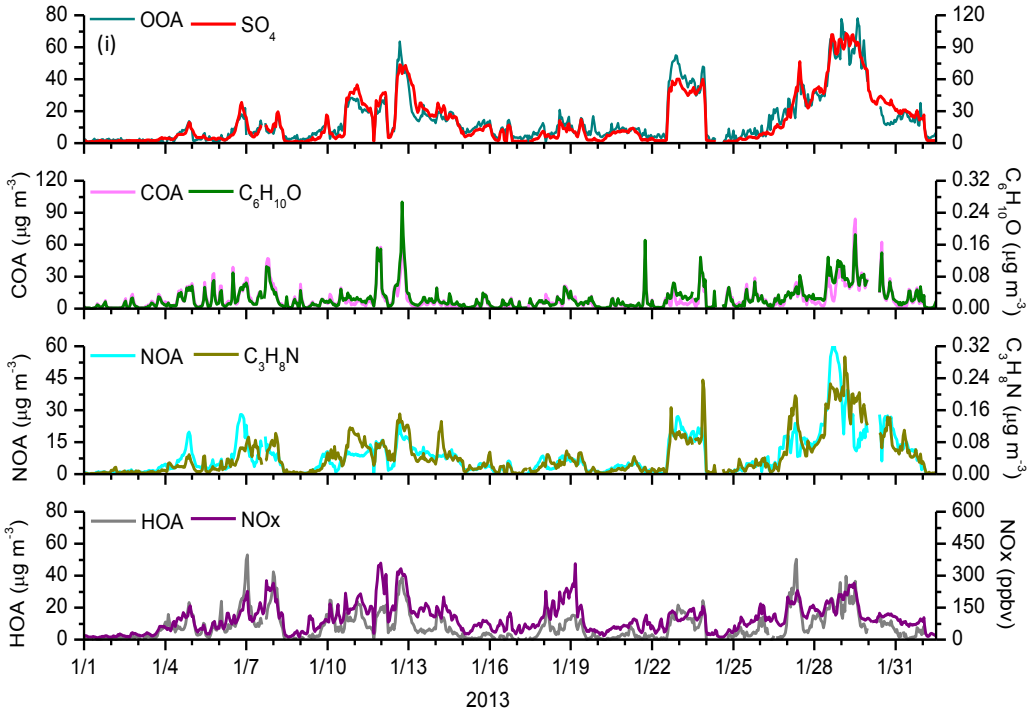
Fig. S-2. The time series of ANMF (ANMF=(80/62×NO_3)/(NH_4 + SO_4 + NO_3 + Chl + Org))

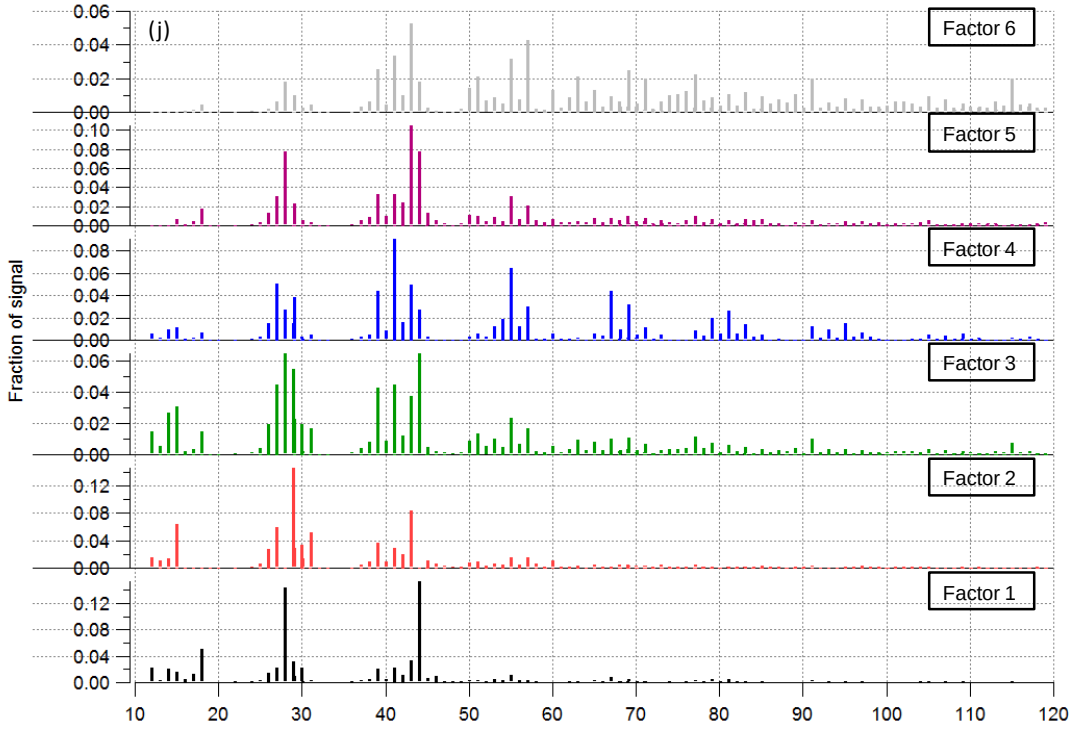



(c)
Scaled Residual
m/z
(d)
Residual Mass (µg m-3)
(e)
Q/Qexp Contribution
2013-01-01
2013-01-06
2013-01-11
2013-01-16
2013-01-21
2013-01-26
2013-01-31
Date
(f)
Factor 3
Factor 2
Factor 1
Fraction of signal
m/z


(g)
Factor 3
Factor 2
Factor 1
80
60
40
20
0
120
100
80
60
40
20
0
2013-01-01
2013-01-06
2013-01-11
2013-01-16
2013-01-21
2013-01-26
2013-01-31
Date


(h)
CxHy+ CxHyOz+ HxO+
CxHyOzNp+ CxHyNp+
OOA
O/C: 0.51
H/C: 1.40
N/C: 0.009
OM/OC: 1.81
COA
O/C: 0.10
H/C: 1.71
N/C: 0.001
OM/OC: 1.28
NOA
O/C: 0.39
H/C: 1.58
N/C: 0.059
OM/OC: 1.73
HOA
O/C: 0.10
H/C: 1.65
N/C: 0.010
OM/OC: 1.29
Fraction of OA Signal
0.15
0.12
0.09
0.06
0.03
0.00
0.10
0.08
0.06
0.04
0.02
0.00
0.12
0.09
0.06
0.03
0.00
0.12
0.08
0.04
0.00
10
20
30
40
50
60
70
80
90
100
110
120
m/z (amu)


(i)
OOA
SO4
COA
C6H10O
NOA
C3H8N
HOA
NOx
COA (μg m-3)
C6H10O (μg m-3)
NOA (μg m-3)
C3H8N (μg m-3)
HOA (μg m-3)
NOx (ppbv)
1/1
1/4
1/7
1/10
1/13
1/16
1/19
1/22
1/25
1/28
1/31
2013


(j)
Factor 6
Factor 5
Factor 4
Factor 3
Factor 2
Factor 1
Fraction of signal

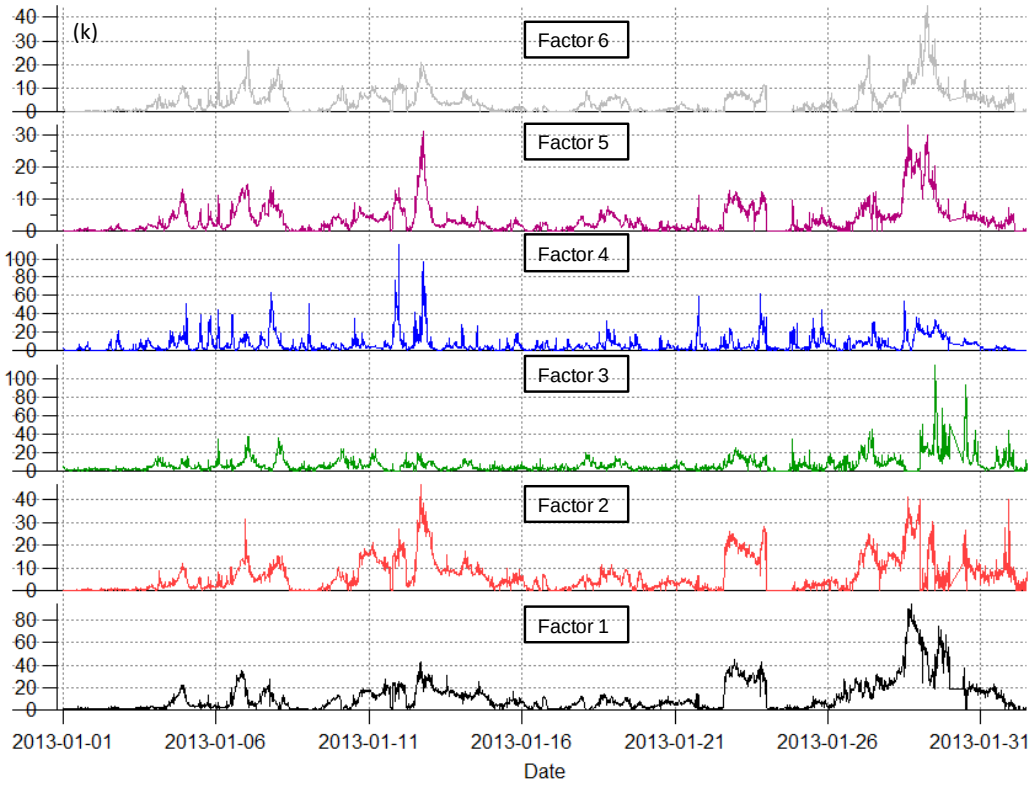


Figure S-3. PMF diagnostic plots: (a) Q/Q expected (Q = the sum of squared scaled residuals over the whole dataset) plotted versus the number of factors used in the PMF solution; (b) Q/Q expected plotted versus the rotational forcing parameter (FPEAK) for solutions with 5 factors; (c) Median (the line within the box) and lower/upper quartiles (boxes) of the scaled residuals per m/z; (d,e) time series of the total residual and Q/Q expected contribution for every point in time during the study; (f) 3-factor profiles (mass spectra); and (g) time series the 3-factor solution (with FPEAK = -0.1); (h) 4-factor profiles and (i) time series for the 4-factor solution (with FPEAK=-0.1); (j) 6-factor profiles and (k) time series for the 6-factor solution (with FPEAK=-0.1)

Discussion S1: Choose the optimal factors number

The PMF analysis based on the HRMS dataset observed in the campaign was performed for 1 to 8 factors. In the PMF analysis, the Q/Q_{exp} values represent the ratios between the actual sum of the squares of the scaled residuals (Q) obtained from the PMF least square fit and the ideal Q (Q_{exp}) obtained if the fit residuals at each point were equal to the noise specified for each data point. The Q/Q_{exp} values of greater than the ideal value of 1 may be indicative of the fact that the input noise values underestimate the true noise because they do not include errors associated with the high‐resolution peak fitting process (He et al., 2011). In a 1 or 2 factor solution, the residual at the key m/z's and time periods were too large. Meanwhile, the Q/Qexp values were too high, 10.4 and 7.5 respectively. In the 3 factor solution, the Q/Qexp decreased (6.1). We identified three components, including oxygenated organic aerosols (OOA), cooking-related (COA) and hydrocarbon-like (HOA), whereas there were many nitrogen-containing fragments in the MS of the OOA (N/C is 0.023) and HOA (N/C is 0.028) and the time trends and diurnal cycles of them mixed with each other. Meanwhile, the *m/z*60 also was distributed in COA and HOA. In the 4 factor solution, the Q/Q_{exp} was 5.0 and a new component was identified. The MS of this new component contained many nitrogen-containing

fragments, and the N/C value was 0.059. The N/C values for OOA and HOA then dropped to 0.009 and 0.010, respectively. While, the *m/z*60 still have not been separated from HOA, NOA and COA. Meanwhile, these sources cannot fully comply with the actual situation in Beijing. Because the coal combustion has been proved is a very important OA source in winter in Beijing. Sun et al. (2013) found that the MS of CCOA (coal combustion OA) contain a large number of *m/z*60. Therefore, the 4 factor solution is inappropriate. In the 5 factor solution, although the Q/Q_{exp} only decreased by 0.1, the *m/z*60 was separated now. Then the NOA disappeared and a SV-OOA and a CCOA were indentified. Moreover, the diurnal cycles of five components were distinctive and the element ratios of them were in a reasonable range compare with other studies. Most important, the 5 factors solution is fully comply with the actual situation in Beijing. When the number of factors changed from 6 to 8, there was not an obvious decrease of the Q/Q_{exp} value and some of the split factors had time series and MS that appeared mixed. Thus, the five factor solution was chosen as the optimal solution. The sensitivity of the 5 factor solution to rotation and starting values was explored by varying the FPEAK and seed parameters. Lower Q/Q_{exp} values can indicate a better fit to the data set and thus be used as one criterion for choosing a suitable solution (Ulbrich et al., 2009). With FPEAK varying from –1.5 to 1.5 in increments of 0.2 (seed = 0), the lowest Q/Q_{exp} was obtained at approximately -0.1. Therefore, FPEAK = -0.1 was chosen as the best solution. With a seed value varying from 0 to 250 in increments of 10 (FPEAK=0), the Q/Qexp almost had no change. Based on all of these tests, the four factor, FPEAK=-0.1, seed=0 solution was chosen as the optimal solution for this analysis. For more details on PMF and the interpretation of these plots see Ulbrich et al.(2009).

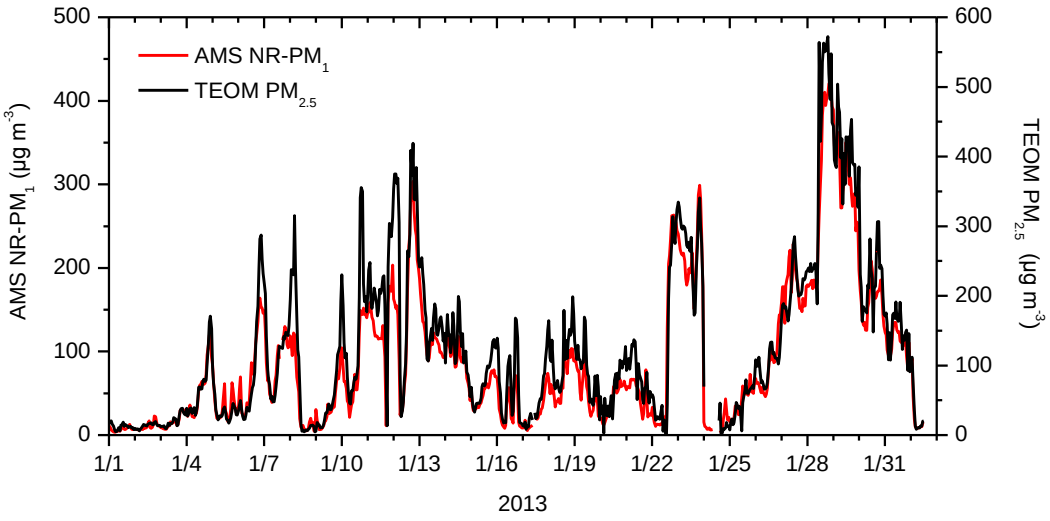


Fig. S-4. The NR-PM_1 and $PM_{2.5}$ mass concentrations measured by AMS and TEOM, respectively

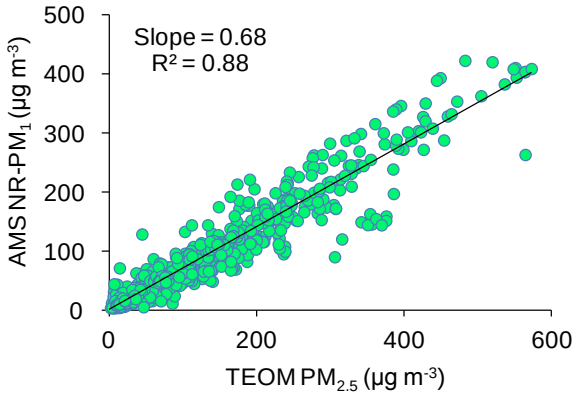


Fig. S-5 The scatter plot of $PM_{2.5}$(TEOM) versus NR-PM_1(AMS)

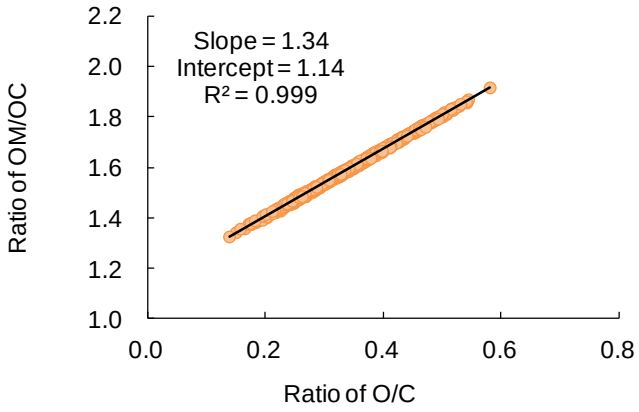


Fig. S-6 The scatter plot of ratio of O/C versus the ratio of OM/OC

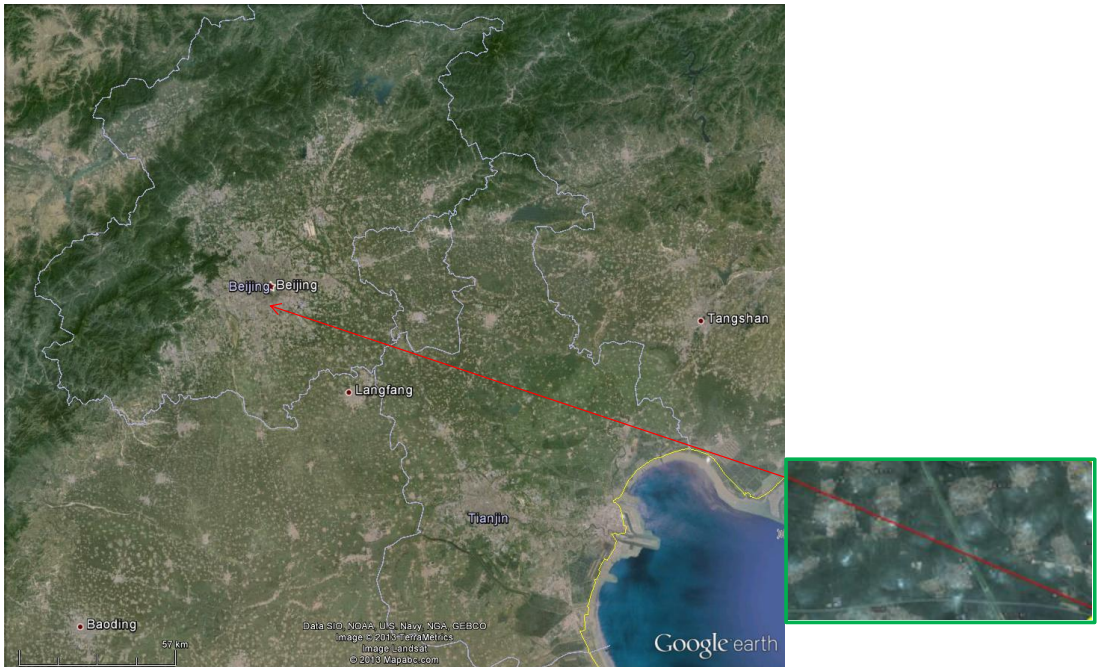


Fig. S-7 the satellite image of the sources around Beijing