## **SUPPLEMENTARY INFORMATION**

## Size-resolved characterization of organic aerosol in the North China

## Plain: new insights from high resolution spectral analysis

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Figure S1. Size distributions of  $Q/Q_{exp}$  in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.



Figure S2. Average OA composition resolved by size-resolved PMF (labeled as "unfix MS"), size-all PMF (labeled as "fix MS"), and bulk OA PMF (labeled as "Bulk").



Figure S3. Size distributions of differences in mass loadings identified by size-resolved PMF and

size-all PMF in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.



Figure S4. Size distributions of ratios of NO3 to SO4 in Beijing in winter.



Figure S5. Size distributions of the total OA species (OOA1, OOA2, SOA, OPOA, POA, FFOA, BBOA) compared to the size distributions of total OA in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.





Figure S6. High-resolution mass spectra of (a1-a12) POA, (b1-b12) OOA1 and (c6-c12) OOA2 in Beijing in summer. The number of captions represents the sequence of the bin. A summary of size ranges of 14 bins in the range of 100 - 1000 nm is shown in Table. S1.





Figure S7. High-resolution mass spectra of (a1-a14) POA, (b1-b14) SOA and (c1-c14) OPOA in Beijing in winter. The number of captions represents the sequence of the bin. A summary of size ranges of 14 bins in the range of 100 - 1000 nm is shown in Table. S1.





Figure S8. High-resolution mass spectra of (a1-a14) FFOA, (b1-b14) BBOA and (c1-c14) SOA in Gucheng in winter. The number of captions represents the sequence of the bin. A summary of size ranges of 14 bins in the range of 100 - 1000 nm is shown in Table. S1.



Figure S9. Diurnal evolution of the size distributions of OA in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.



Figure S10. Correlations of OA factors and tracers in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng in different size. POA (left), OOA1(middle) and OOA2(right) were identified in summer in Beijing. Comparatively, POA (left), OPOA (middle) and SOA (right) in Beijing and FFOA (left), BBOA (middle) and OOA(right) in Gucheng were identified during wintertime.



Figure S11. Size distributions of  $f_{44}/f_{43}$  (left),  $f_{55}/f_{57}$  (middle) and  $f_{44}/f_{60}$  (right) for OA factors in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.



Figure S12. Contribution of increase in the concentrations of OA species during moderately (left,  $35 \ \mu g \ m^{-3} < NR-PM_1 < 70 \ \mu g \ m^{-3}$ ) and seriously pollution (right, NR-PM<sub>1</sub> > 70 \ \mu g \ m^{-3}) in each size bin compared to clean days (NR-PM<sub>1</sub> < 35 \ \mu g \ m^{-3}) in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng. The differences in OA loading between moderately pollution and clean days (left), seriously pollution and clean days (right) are also shown.



Figure S13. The contribution for OA factors at NR-PM<sub>1</sub>< 35  $\mu$ g m<sup>-3</sup> (left), 35  $\mu$ g m<sup>-3</sup> < NR-PM<sub>1</sub> < 70  $\mu$ g m<sup>-3</sup> (middle) and NR-PM<sub>1</sub> > 70  $\mu$ g m<sup>-3</sup> (right) in (a) summer and (b) winter in Beijing, and (c) winter in Gucheng.

Bin number	PToF Size (nm)	
Bin#1	110.6-135.9	
Bin#2	135.9-164.8	
Bin#3	164.8-197.7	
Bin#4	197.7-235.1	
Bin#5	235.1-277.4	
Bin#6	277.4-325.1	
Bin#7	325.1-378.8	
Bin#8	378.8-438.9	
Bin#9	438.9-506.3	
Bin#10	506.3-581.7	
Bin#11	581.7-665.8	
Bin#12	665.8-759.7	
Bin#13	759.7-864.4	
Bin#14	864.4-981.0	

Table S1. A summary of size ranges of 14 bins in the range of 100 - 1000 nm

Table S2. A summary of frequencies of three different PM levels

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	$NR-PM_1 < 35 \ \mu g \ m^{-3}$	$35  \mu g  m^{-3} \! < \! NR \! - \! PM_1 \! < \! 70  \mu g  m^{-3}$	$NR-PM_1 > 70 \ \mu g \ m^{-3}$	
Summer <sub>Beijing</sub>	57.3%	30.9%	11.8%	
Winter <sub>Beijing</sub>	58.1%	27.6%	14.3%	
WinterGucheng	25.2%	37.2%	37.6%	