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Supplement of

Mass spectral characterization of secondary organic aerosol from urban cooking and vehicular sources

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Fig. S1. Schematic depiction of the simulation and measurement system for the cooking and vehicle experiments.

**Source emission**
- deep-frying chicken;
- shallow-frying tofu;
- stir-frying cabbage;
- kung pao chicken

**Vehicle Engine:**
- GDI China phase V gasoline
  - Engine speed: 1500rpm; 1750rpm; 2000rpm
  - Engine torque: 16Nm: 32Nm: 40Nm

**Oxidation**

**SMPS**

**Go: PAM**

**GAS (SO₂, CO₂)**

**HR-ToF-AMS**
### Table S1. Details of cooking and sampling procedures.

| Cooking Dish        | Cooking Material                              | Oil Temperature | Cooking Time | Numbers for Each Dish | Sampling Time | Fuel                       | Sampling Temperate |
|---------------------|-----------------------------------------------|-----------------|--------------|------------------------|---------------|----------------------------|--------------------|
| Deep-fried chicken  | 170g chicken, 500ml corn oil                 | 145–155°C       | 66 min       | 8                      | 90 min        |                            |                    |
| Shallow-frying tofu | 500g tofu, 200ml corn oil                     | 100–110°C       | 64 min       | 8                      | 60 min        | Liquefied petroleum gas    | 20–25°C            |
| Stir-frying cabbage | 300g cabbage, 40ml corn oil                  | 95–105°C        | 47 min       | 8                      | 58 min        | Iron work                  |                    |
| Kung Pao chicken    | 150g chicken, 50g peanut, 50g cucumber, 40ml corn oil | 90–105°C        | 40 min       | 8                      | 60 min        |                            |                    |

### Table S2. Details of vehicle and sampling procedures.

| Running Condition | Sampling Time | Parallels | Fuel               | Sampling Temperate |
|-------------------|---------------|-----------|--------------------|--------------------|
| Rotating speed    | Torque        |           |                    |                    |
| 1500 rpm          | 16 Nm         | 60 min    | 5                  | Commercial         | 20–25°C       |
| 1750 rpm          | 16 Nm         | 60 min    | 5                  |                    |               |
| 2000 rpm          | 16 Nm         | 60 min    | 5                  | Commercial         | 20–25°C       |
| 2000 rpm          | 32 Nm         | 60 min    | 5                  | China V gasoline   |               |
| 2000 rpm          | 40 Nm         | 60 min    | 5                  |                    |               |
Table S3. The OH exposure and photochemical age for all conditions in cooking and vehicle experiments

| O₃ concentration (ppbv) | RH (%) & Temperature (°C) | Description of Go: PAM | OH exposure (molecules cm³ s⁻¹) | Photochemical Age (day) | O₃ concentration (ppbv) | RH (%) & Temperature (°C) | Description of Go: PAM | OH exposure (molecules cm³ s⁻¹) | Photochemical Age (day) |
|------------------------|---------------------------|------------------------|-------------------------------|------------------------|------------------------|---------------------------|------------------------|-------------------------------|------------------------|
| 0                      |                           | Sample flow (7 L/min)   | 4.3E+10                       | 0.3                    | 0                      |                           | Sample flow (4 L/min)   | 7.8E+10                       | 0.6                    |
| 310                    | 18–23%                    | and oxidant flow (3 L/min); Residence time: 55 s | 9.6E+10                       | 0.7                    | 2367                   | 44–49%                    | oxidant flow (1 L/min); Residence time: 110 s | 2.1E+11                       | 1.7                    |
| 1183                   | &16–19°C                  |                         | 1.4E+11                       | 1.1                    | 4433                   | &19–22°C                  |                         | 3.7E+11                       | 2.9                    |
| 2217                   |                           |                         | 2.7E+11                       | 2.1                    | 6533                   |                           |                         | 5.4E+11                       | 4.2                    |
| 4025                   |                           |                         |                               |                        |                        |                           |                         |                               |                        |
Table S4. The mass concentrations of primary organic aerosol (POA) for all conditions in vehicle experiments

| Experiment          | POA Mass concentration (μg/m³) |
|---------------------|--------------------------------|
|                     | Average | Standard Deviation |
| 1500rpm_16Nm        | 1.20    | 0.30               |
| 1750rpm_16Nm        | 1.26    | 0.61               |
| 2000rpm_16Nm        | 1.14    | 0.30               |
| 2000rpm_32Nm        | 1.29    | 0.62               |
| 2000rpm_40Nm        | 1.23    | 0.31               |
Fig. S2. The mass spectra of aged HOA emission from different vehicle running conditions under different EPA.
Table S5. The $\theta$ angles among the mass spectra of aged HOA under EPA 1.7 days, 2.9 days, and 4.1 days.

|          | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------|--------------|--------------|--------------|--------------|--------------|
| 1500rpm_16Nm | 0            | 8            | 8            | 16           | 18           |
| 1750 rpm_16Nm | 0            | 1            | 9            | 11           |
| 2000 rpm_16Nm | 0            | 9            | 11           |
| 2000 rpm_32Nm | 0            | 4            |
| 2000 rpm_40Nm | 0            |

|          | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------|--------------|--------------|--------------|--------------|--------------|
| 1500rpm_16Nm | 0            | 14           | 14           | 29           | 19           |
| 1750 rpm_16Nm | 0            | 2            | 15           | 6            |
| 2000 rpm_16Nm | 0            | 14           | 5            |
| 2000 rpm_32Nm | 0            | 9            |
| 2000 rpm_40Nm | 0            |

|          | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------|--------------|--------------|--------------|--------------|--------------|
| 1500rpm_16Nm | 0            | 8            | 8            | 3            | 29           |
| 1750 rpm_16Nm | 0            | 1            | 7            | 21           |
| 2000 rpm_16Nm | 0            | 7            | 21           |
| 2000 rpm_32Nm | 0            | 26           |
| 2000 rpm_40Nm | 0            |
Fig. S3. The mass spectra of aged COA emission from different Chinese dishes under different EPA.
Table S6. The θ angles among the mass spectra of POA and aged COA emission from different Chinese dishes under EPA 0.3 day, 1.1 days, and 2.1 days.

|                | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|------------------|
| **POA θ angles** |                     |                     |                     |                  |
| deep-frying chicken | 0                   | 31                  | 29                  | 24               |
| stir-frying cabbage  | 0                   | 12                  | 11                  |                  |
| shallow-frying tofu  | 0                   | 10                  | 10                  |                  |
| Kung Pao chicken     | 0                   |                     |                     |                  |

|                | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|------------------|
| **EPA 0.3 day θ angles** |                     |                     |                     |                  |
| deep-frying chicken | 0                   | 23                  | 22                  | 17               |
| stir-frying cabbage  | 0                   | 10                  | 10                  |                  |
| shallow-frying tofu  | 0                   | 10                  | 10                  |                  |
| Kung Pao chicken     | 0                   |                     |                     |                  |

|                | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|------------------|
| **EPA 1.1 days θ angles** |                     |                     |                     |                  |
| deep-frying chicken | 0                   | 20                  | 17                  | 15               |
| stir-frying cabbage  | 0                   | 10                  | 10                  | 14               |
| shallow-frying tofu  | 0                   | 10                  | 10                  | 16               |
| Kung Pao chicken     | 0                   |                     |                     |                  |

|                | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|------------------|
| **EPA 2.1 days θ angles** |                     |                     |                     |                  |
| deep-frying chicken | 0                   | 22                  | 18                  | 17               |
| stir-frying cabbage  | 0                   | 10                  | 10                  | 13               |
| shallow-frying tofu  | 0                   | 10                  | 10                  | 12               |
| Kung Pao chicken     | 0                   |                     |                     |                  |
Fig. S4. The changes in mass spectra of aged HOA emissions from different conditions.
Table S7. The θ angles among the mass spectra under different EPA at one vehicle condition (1500rpm_16Nm, 1750rpm_16Nm, 2000rpm_16Nm, 2000rpm_32Nm, and 2000rpm_40Nm, respectively).

| EPA          | θ angles | HOA_ambient | 0.6 day | 1.7 days | 2.9 days | 4.1 days |
|--------------|----------|-------------|---------|----------|----------|----------|
| 1500rpm_16Nm |          | HOA_ambient | 0       | 27       | 45       | 63       |
|              |          | 0.6 day     | 0       | 24       | 46       | 46       |
|              |          | 1.7 days    | 0       | 22       | 1        | 1        |
|              |          | 2.9 days    | 0       | 0        | 0        | 0        |
|              |          | 4.1 days    |         |          |          |          |
| 1750rpm_16Nm |          | HOA_ambient | 0       | 29       | 40       | 51       |
|              |          | 0.6 day     | 0       | 14       | 29       | 35       |
|              |          | 1.7 days    | 0       | 15       | 7        | 7        |
|              |          | 2.9 days    | 0       | 0        | 0        | 0        |
|              |          | 4.1 days    |         |          |          |          |
| 2000rpm_16Nm |          | HOA_ambient | 0       | 30       | 35       | 41       |
|              |          | 0.6 day     | 0       | 7        | 13       | 38       |
|              |          | 1.7 days    | 0       | 10       | 37       | 37       |
|              |          | 2.9 days    | 0       | 0        | 28       | 28       |
|              |          | 4.1 days    |         |          |          |          |
| 2000rpm_32Nm |          | HOA_ambient | 0       | 29       | 36       | 48       |
|              |          | 0.6 day     | 0       | 10       | 24       | 21       |
|              |          | 1.7 days    | 0       | 19       | 13       | 13       |
|              |          | 2.9 days    | 0       | 0        | 12       | 12       |
|              |          | 4.1 days    |         |          |          |          |
| 2000rpm_40Nm |          | HOA_ambient | 0       | 29       | 36       | 48       |
|              |          | 0.6 day     | 0       | 10       | 24       | 21       |
|              |          | 1.7 days    | 0       | 19       | 13       | 13       |
|              |          | 2.9 days    | 0       | 0        | 12       | 12       |
|              |          | 4.1 days    |         |          |          |          |
Fig. S5. The mass spectra of aged COA oxidation under different OH exposure for different Chinese dishes.
Table S8. The θ angles among the mass spectra under different EPA for different Chinese dishes.

| Deep-frying chicken | POA | 0.3 day | 0.7 day | 1.1 days | 2.1 days |
|---------------------|-----|---------|---------|----------|---------|
| POA                 | 0   | 12      | 17      | 19       | 19      |
| 0.3 day             | 0   | 6       | 9       | 9        |         |
| 0.7 day             | 0   | 4       | 5       |          |         |
| 1.1 days            | 0   | 4       | 5       |          |         |
| 2.1 days            |     |         |         | 0        |         |

| Stir-frying cabbage | POA | 0.3 day | 0.7 day | 1.1 days | 2.1 days |
|---------------------|-----|---------|---------|----------|---------|
| POA                 | 0   | 5       | 10      | 15       | 18      |
| 0.3 day             | 0   | 6       | 10      | 14       |         |
| 0.7 day             | 0   | 6       | 9       | 9        |         |
| 1.1 days            | 0   | 5       |          | 5        |         |
| 2.1 days            |     |         |         | 0        |         |

| Shallow frying tofu | POA | 0.3 day | 0.7 day | 1.1 days | 2.1 days |
|---------------------|-----|---------|---------|----------|---------|
| POA                 | 0   | 7       | 12      | 15       | 21      |
| 0.3 day             | 0   | 6       | 9       | 14       |         |
| 0.7 day             | 0   | 3       | 9       |          |         |
| 1.1 days            | 0   | 6       |          | 6        |         |
| 2.1 days            |     |         |         | 0        |         |

| Kung Pao chicken    | POA | 0.3 day | 0.7 day | 1.1 days | 2.1 days |
|---------------------|-----|---------|---------|----------|---------|
| POA                 | 0   | 7       | 13      | 19       | 23      |
| 0.3 day             | 0   | 8       | 13      | 17       |         |
| 0.7 day             | 0   | 7       | 10      |          |         |
| 1.1 days            | 0   | 7       |          | 7        |         |
| 2.1 days            |     |         |         | 0        |         |
Fig. S6. Van Krevelen diagram of POA, aged COA and aged HOA from vehicle and cooking.
Fig. S7. Diagnostic plots of the PMF analysis on OA mass spectral matrix for stir-frying cabbage. (a) $Q/Q_{\text{exp}}$ as a function of number of factors (P) selected for PMF modeling. For the four-factor solution (i.e., the best P), (b) $Q/Q_{\text{exp}}$ as a function of fPeak, (c) The fractions of OA factors vs. fPeak, (d) The $Q/Q_{\text{exp}}$ values for each m/z
Fig. S8. Mass spectra of the (a) 2-factor, and (b) 3-factor solution using PMF method in stir-frying cabbage OA analysis.
Table S9. The optimum choices for PMF factors in stir-frying cabbage OA analysis.

| Factor number | Fpeak | Seed | Q/Q_{exp} | Solution Description |
|---------------|-------|------|-----------|---------------------|
| 1             | 0     | 0    | 1.62      | Too few factors, large residuals at time series and key m/z |
| 2             | 0     | 0    | 0.85      | **Optimum choices for PMF factors (POA and SOA).** Time series, mass spectra and diurnal variations of PMF factors are reasonable. |
| 3-5           | 0     | 0    | 0.77-0.82 | Factor split. Take 3 factor number solution as an example, POA was split into two factors with similar spectra. |
Fig. S9. Diagnostic plots of the PMF analysis on aged HOA mass spectral matrix for 2000rpm_32Nm. (a) $Q/Q_{\text{exp}}$ as a function of number of factors ($P$) selected for PMF modeling. For the four-factor solution (i.e., the best $P$), (b) $Q/Q_{\text{exp}}$ as a function of $f_{\text{Peak}}$, (c) The fractions of OA factors vs. $f_{\text{Peak}}$, (d) The $Q/Q_{\text{exp}}$ values for each m/z.
Fig. S10. Mass spectra of the (a) 2-factor, and (b) 3-factor solution using PMF method in 2000rpm_32Nm aged HOA analysis.
Table S10. The optimum choices for PMF factors in 2000rpm_32Nm aged HOA analysis.

| Factor number | Fpeak | Seed | Q/Q_{exp} | Solution Description |
|---------------|-------|------|-----------|----------------------|
| 1             | 0     | 0    | 15.44     | Too few factors, large residuals at time series and key m/z |
|               |       |      |           | **Optimum choices for PMF factors (LO-SOA and MO-SOA). Time** |
|               |       |      |           | **series, mass spectra and diurnal variations of PMF factors are** |
|               |       |      |           | **reasonable.** |
| 2             | 0     | 0    | 2.87      | Factor split. Take 3 factor number solution as an example, LO-SOA |
|               |       |      |           | was split into two factors with similar spectra. |
Table S11. A summary of dominant peaks among cooking PMF POA.

|                | Deep-frying chicken | Stir-frying cabbage | Shallow frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|-----------------|
| $f_{28}$       | 0.0508              | 0.0560              | 0.0682              | 0.0685          |
| $f_{43}$       | 0.0802              | 0.0365              | 0.0489              | 0.0597          |
| $f_{55}$       | 0.0641              | 0.0664              | 0.0842              | 0.0757          |
| $f_{57}$       | 0.0966              | 0.0411              | 0.0473              | 0.0612          |
| $f_{67}$       | 0.0211              | 0.0382              | 0.0404              | 0.0333          |
| $f_{69}$       | 0.0486              | 0.0343              | 0.0383              | 0.0376          |

Table S12. A summary of dominant peaks among cooking PMF SOA.

|                | Deep-frying chicken | Stir-frying cabbage | Shallow frying tofu | Kung Pao chicken |
|----------------|---------------------|---------------------|---------------------|-----------------|
| $f_{28}$       | 0.0504              | 0.0451              | 0.0463              | 0.0682          |
| $f_{29}$       | 0.0481              | 0.0796              | 0.0675              | 0.0644          |
| $f_{41}$       | 0.0501              | 0.0590              | 0.0679              | 0.0547          |
| $f_{43}$       | 0.1032              | 0.0865              | 0.0944              | 0.1023          |
| $f_{44}$       | 0.0609              | 0.0596              | 0.0584              | 0.0800          |
| $f_{55}$       | 0.0534              | 0.0586              | 0.0636              | 0.0495          |
| $f_{57}$       | 0.0665              | 0.0376              | 0.0421              | 0.0364          |

Table S13. A summary of dominant peaks among vehicle PMF LO-SOA.

|                | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------------|--------------|--------------|--------------|--------------|--------------|
| $f_{28}$       | 0.0579       | 0.0551       | 0.0527       | 0.0493       | 0.0081       |
| $f_{41}$       | 0.0417       | 0.0493       | 0.0443       | 0.0386       | 0.0574       |
| $f_{43}$       | 0.1571       | 0.1495       | 0.1523       | 0.1670       | 0.1632       |
| $f_{44}$       | 0.0663       | 0.0653       | 0.0623       | 0.0597       | 0.0183       |
| $f_{55}$       | 0.0384       | 0.0393       | 0.0386       | 0.0339       | 0.0447       |
| $f_{57}$       | 0.0246       | 0.0270       | 0.0253       | 0.0226       | 0.0329       |

Table S14. A summary of dominant peaks among vehicle PMF MO-SOA.

|                | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------------|--------------|--------------|--------------|--------------|--------------|
| $f_{28}$       | 0.2077       | 0.1590       | 0.2141       | 0.2049       | 0.1099       |
| $f_{41}$       | 0.0139       | 0.0186       | 0.0124       | 0.0124       | 0.0242       |
| $f_{43}$       | 0.0722       | 0.1063       | 0.0777       | 0.0771       | 0.1431       |
| $f_{44}$       | 0.2190       | 0.1688       | 0.2239       | 0.2126       | 0.1208       |
| $f_{55}$       | 0.0127       | 0.0181       | 0.0120       | 0.0120       | 0.0238       |
| $f_{57}$       | 0.0042       | 0.0076       | 0.0026       | 0.0032       | 0.0127       |
Table S15. The θ angles among the mass spectra of cooking PMF SOA for different dishes.

| cooking SOA     | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|-----------------|---------------------|---------------------|---------------------|------------------|
| deep-frying chicken | 0                   | 21                  | 18                  | 19               |
| stir-frying cabbage | 0                   | 8                   | 13                  |                  |
| shallow-frying tofu | 0                   | 13                  |                     |                  |
| Kung Pao chicken  | 0                   |                     |                     |                  |

Table S16. The θ angles among the mass spectra of cooking PMF POA for different dishes.

| cooking POA     | deep-frying chicken | stir-frying cabbage | shallow-frying tofu | Kung Pao chicken |
|-----------------|---------------------|---------------------|---------------------|------------------|
| deep-frying chicken | 0                   | 31                  | 28                  | 20               |
| stir-frying cabbage | 0                   | 13                  | 17                  |                  |
| shallow-frying tofu | 0                   | 10                  |                     |                  |
| Kung Pao chicken  | 0                   |                     |                     |                  |

Table S17. The θ angles among the mass spectra of vehicle PMF LO-SOA at different conditions.

| Vehicle LO-SOA | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------------|--------------|--------------|--------------|--------------|--------------|
| 1500rpm_16Nm  | 0            | 3            | 3            | 6            | 19           |
| 1750rpm_16Nm  | 0            | 3            | 7            | 3            |              |
| 2000rpm_16Nm  | 0            | 6            | 3            |              |              |
| 2000rpm_32Nm  | 0            | 6            |              | 6            |              |
| 2000rpm_40Nm  |              |              | 0            |              |              |

Table S18. The θ angles among the mass spectra of vehicle PMF MO-SOA at different conditions.

| Vehicle MO-SOA | 1500rpm_16Nm | 1750rpm_16Nm | 2000rpm_16Nm | 2000rpm_32Nm | 2000rpm_40Nm |
|----------------|--------------|--------------|--------------|--------------|--------------|
| 1500rpm_16Nm  | 0            | 12           | 2            | 2            | 29           |
| 1750rpm_16Nm  | 0            | 12           | 11           | 17           |              |
| 2000rpm_16Nm  | 0            | 3            |              | 28           |              |
| 2000rpm_32Nm  |              | 0            | 27           |              |              |
| 2000rpm_40Nm  |              |              |              |              | 0            |
Fig. S11. The $\theta$ angles between vehicle PMF LO-SOA and PMF MO-SOA under five running conditions.
Fig. S12. Mass spectral profiles of cooking POA, cooking SOA, vehicle LO-SOA, and vehicle MO-SOA as the primary and secondary spectrum constraints in ME-2 model.
Table S19. The $\theta$ angles between ambient COA, HOA, LO-OOA and MO-OOA factors and the cooking PMF POA, SOA, and the vehicle PMF LO-SOA, MO-SOA.

| $\theta$ angles       | HOA_ambient | COA_ambient | LO-OOA_ambient | MO-OOA_ambient | Cooking_POA | Cooking_SOA | Vehicle_LO-SOA | Vehicle_MO-SOA |
|-----------------------|-------------|-------------|----------------|----------------|--------------|-------------|----------------|----------------|
| HOA_ambient           | 0           | 21          | 36             | 56             | 21           | 27          | 30             | 61             |
| COA_ambient           | 21          | 0           | 31             | 49             | 18           | 22          | 34             | 55             |
| LO-OOA_ambient        | 36          | 31          | 0              | 37             | 18           | 28          | 32             | 52             |
| MO-OOA_ambient        | 56          | 49          | 37             | 0              | 18           | 28          | 33             | 18             |
| Cooking_POA           | 21          | 18          | 18             | 18             | 0            | 31          | 39             | 64             |
| Cooking_SOA           | 27          | 22          | 28             | 28             | 31           | 0           | 19             | 46             |
| Vehicle_LO-SOA        | 30          | 34          | 32             | 33             | 39           | 19          | 0              | 46             |
| Vehicle_MO-SOA        | 61          | 55          | 52             | 18             | 64           | 46          | 46             | 0              |
Fig.S13. (a) 5-factor solution performed by ME-2 on organic mass spectra; (b) 7-factor solution performed by ME-2 on organic mass spectra during the wintertime in Shanghai.
Fig. S14. (a) 2-factor solution performed by PMF on organic mass spectra during the wintertime in Shanghai; (b) 4-factor solution performed by PMF on organic mass spectra during the wintertime in Shanghai.
Table S20. Pearson r between the factors identified by using PMF model (4-factor solution), and the external tracers during the wintertime observations in Shanghai.

| Pearson r | Sulfate | CO$_2^+$ | C$_2$H$_4$O$_2^+$ | C$_{10}$H$_8^+$ |
|-----------|---------|----------|-------------------|-----------------|
| MO-OOA_PMF | 0.89    | 0.96     | 0.67              | 0.61            |
| LO-OOA_PMF | 0.04    | 0.31     | 0.44              | 0.51            | 0.59            |

| Pearson r | COA_PMF |
|-----------|---------|
| C$_6$H$_{10}$O$_+$ | 0.81    |

| Pearson r | HOA_PMF |
|-----------|---------|
| NO$_x$    | 0.73    |
Fig.S15. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the winter observation. (a) Q/Qexp as a function of number of factors (P) selected for PMF modeling. For the four-factor solution (i.e., the best P), (b) Q/Qexp as a function of fPeak, (c) The fractions of OA factors vs. fPeak, (d) The correlations among PMF factors.
Fig. S16. Diagnostic plots of the PMF analysis on OA mass spectral matrix for the winter observation. (a) Time series of the measured organic mass and the reconstructed organic mass, (b) Variations of the residual (= measured − reconstructed) of the fit, and the Q/Q_{exp} for each point in time, and (c) The Q/Q_{exp} values for each m/z.
### Table S21. Descriptions of PMF solutions for organic aerosol in the winter study of Shanghai

| Factor number | Fpeak | Seed | Q/Q_{exp} | Solution Description |
|---------------|-------|------|-----------|----------------------|
| 1             | 0     | 0    | 3.97      | Too few factors, large residuals at time series and key m/z |
|               |       |      |           | Few factors (OOA- and HOA-like), large residuals at time series and key m/z. Factors are mixed to some extend based on the time series and spectra. |
| 2             | 0     | 0    | 2.26      | Optimum choices for PMF factors (OOA, HOA and COA). Time series and diurnal variations of PMF factors are consistent with the external tracers. The spectra of four factors are consistent with the source spectra in AMS spectra database. |
| 3             | 0     | 0    | 1.91      | Factor split. Take 4 factor number solution as an example, LO-OOA was split from other factors. |
| 4-6           | 0     | 0    | 1.63-1.73 |                       |
Fig.S17. The time-series correlations of all factors which resolved from PMF and ME-2 with external tracers during the wintertime observations in Shanghai.
Table S22. Pearson r between the factors identified by using PMF and ME-2 model, and the external tracers during the wintertime observations in Shanghai.

| Pearson r   | Sulfate | CO$_2^+$ | C$_2$H$_4$O$_2^+$ | C$_{10}$H$_8^+$ |
|-------------|---------|----------|-------------------|-----------------|
| OOA_PMF     | 0.90    | 0.96     | 0.65              | 0.96            |
| MO-OOA_ME-2 | 0.87    | 0.95     | 0.61              | 0.55            |

| Pearson r   | Nitrate | C$_2$H$_3$O$_2^+$ |
|-------------|---------|-------------------|
| OOA_PMF     | 0.94    | 0.90              |
| LO-OOA_ME-2 | 0.84    | 0.95              |

| Pearson r   | COA_PMF | COA_ME-2 |
|-------------|---------|----------|
| C$_6$H$_{10}$O$_2^+$ | 0.74    | 0.85     |

| Pearson r   | HOA_PMF | HOA_ME-2 |
|-------------|---------|----------|
| NO$_x$      | 0.70    | 0.64     |

| Pearson r   | C$_2$H$_4$O$_2^+$ | C$_{10}$H$_8^+$ |
|-------------|-------------------|-----------------|
| Other POA_ME-2 | 0.88              | 0.88            |
Fig. S18. The time-series correlations of all factors which resolved from ME-2 constraining two POA profiles and ME-2 constraining four factors spectral profiles with external tracers during the wintertime observations in Shanghai.
Fig. S19. The comparison of the mass spectra, the diurnal variation, and fraction between ME-2 constraining the spectral profiles of two primary factors (the cooking POA, ambient HOA) and ME-2 constraining four spectral profiles resolved factors during the wintertime in Shanghai. The black lines in the spectra and diurnal pattern are the result of ME-2 analysis by constraining two spectral profiles in the actual atmosphere in Shanghai winter. The four spectral profiles were two primary OA factors (the cooking POA, ambient HOA resolved in three cities) and two secondary OA factors (the cooking SOA, the vehicle LO-SOA).
The comparison of the mass spectra, the diurnal variation, and fraction between ME-2 and PMF resolved factors during the summertime in Shanghai.
Fig. S21. The time-series correlations of all factors which resolved from PMF and ME-2 with external tracers during the summertime observations in Shanghai.
Table S23. Pearson $r$ between the factors identified by using PMF and ME-2 model, and the external tracers during the summertime observations in Shanghai.

| Pearson r | Sulfate | $CO_2^+$ |
|-----------|---------|----------|
| MO-OOA_PMF | 0.94    | 0.79     |
| MO-OOA_ME-2 | 0.87    | 0.95     |

| Pearson r | Nitrate | $C_2H_3O^+$ |
|-----------|---------|-------------|
| LO-OOA_PMF | 0.53    | 0.94        |
| LO-OOA_ME-2 | 0.60    | 0.96        |

| Pearson r | COA_PMF | COA_ME-2 |
|-----------|---------|----------|
| $C_6H_{10}O^+$ | 0.23    | 0.36     |

| Pearson r | HOA_PMF | HOA_ME-2 |
|-----------|---------|----------|
| BC       | 0.52    | 0.55     |