



# Supplement of

# Size-segregated particle number and mass concentrations from different emission sources in urban Beijing

Jing Cai et al.

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#### 1. RIEs and CO2<sup>+</sup>/ NO3 artifact correction of ACSM

The relative ionization efficiencies (RIEs) for sulfate, nitrate, ammonium, chloride and organics were 0.86, 1.05, 4.0, 1.5 and 1.4, respectively. RIEs of ammonium, sulfate and chloride were obtained by pure standard of  $NH_4NO_3$  (99%, Tianjin-Fuchen Corp. China), ( $NH_4$ )<sub>2</sub>SO<sub>4</sub> (99 %, Sigma-Aldrich Corp. U.S.) and  $NH_4Cl$  (99.5%, Sigma-Aldrich Corp. U.S.). Ionization efficiency (IE) in this study was obtained from ammonium nitrate calibration, which was 167.6 ions/pg under air beam of  $3.1 \times 10^5$  ions/s.

Recently, it was discovered that NO<sub>3</sub> induces a positive bias on organic CO<sub>2</sub><sup>+</sup> concentrations in the AMS/ACSM systems by Pieber et al. (2016), which can be described as a function of ambient NO<sub>3</sub> ( $\mu$ g/m<sup>3</sup>) in combination with the CO<sub>2</sub><sup>+</sup>/NO<sub>3</sub> ratio from pure NH<sub>4</sub>NO<sub>3</sub> measurements (CO<sub>2</sub><sup>+</sup>/NO<sub>3</sub>)<sub>AN</sub>:

For pure  $NH_4NO_3$  aerosol from calibrations, we determined the magnitude of the  $CO_2^+/NO_3$  artifact(Pieber et al., 2016) and parametrized it as a function of the fragmentation pattern of  $NO_3 (NO^+/NO_2^+)$  to account for changes in the vaporizer in the ACSM:

$$(CO_2^+/NO_3)_{NH4NO3} = 0.025 \pm 0.002 \times (NO^+/NO_2^+)_{NH4NO3}$$

Then we determined the  $CO_2$  concentration from OA using a two week moving average (NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup>) from ambient observations:

$$(CO_2^+)_{OA,meas} = (CO_2^+)_{meas} - (CO_2^+/NO_3)_{NH4NO3} \times (NO_3)_{meas}$$

Further, we propagated the uncertainty of the subtraction when computing the PMF input matrices.

$$\sigma_{(CO2+)OA,meas} = \sqrt{\sigma_{((CO2+)OA,meas)counting}^2 + \sigma_{((CO2+NO3)AN,meas\times(NO3)meas)}^2}$$



Fig. S1 Comparison of calculated PM<sub>SMPS</sub> with measured NR-PM<sub>2.5</sub> plus BC in all sampling days



**Fig. S2** Average diurnal evolution of number fractions of (a) *N*<sub>20-100</sub> (20–100 nm) and *N*<sub>20-100</sub> (100–680 nm), (b) number concentrations in *N*<sub>20-40</sub> (20–40 nm), *N*<sub>40-100</sub> (40–100 nm) and *N*<sub>100-680</sub> (100–680 nm) during non-NPF days



**Fig. S3** Average diurnal evolution of particle number size distribution during all days (NPF days are also concluded). (a) Particle size distribution, (b) number concentrations in  $N_{20.40}$  (20–40 nm),  $N_{40-100}$  (40–100 nm) and  $N_{100-680}$  (100–680 nm), (c) number fractions, (d) the comparison between SMPS and NR-PM<sub>2.5</sub>+BC, (e) different component concentrations (f) and diurnal variations of the mass fractions of different components

And to compare our sampling period with long-term measurement, we also present the NR-PM<sub>2.5</sub> and BC measured from Feb 2018 to Jun 2019, which is shown in Figure S4.



Fig. S4 Long-term (Feb 2018 to Jun 2019) measurement of NR-PM2.5 from ACSM and BC from aethalometer



**Figure S5.** The ratio of m/z 115 to total OA in our sampling period (April 6 to July 2, 2018), winter period (Dec 2018 to Feb 2019) and the whole year measurement (Feb 2018 to Jun 2019).

### 2. PMF analysis and validations

#### 2.1 Size-PMF validations

Month	Day of the Month		
April, 2018	10, 11, 12, 15, 16, 17, 24, 25, 27, 30		
May, 2018	1, 2, 3, 5, 6, 7, 8, 9, 18, 22, 23, 24, 27, 28, 29, 30		
June, 2018	1, 2, 4, 13, 17, 19, 20, 21, 23, 24, 25, 27, 28		

Table S1. NPF days during the sampling period (April 6, 2018 to July 2, 2018)

PMF (2, ver. 4.2 (Ulbrich et al., 2009)) is applied to the size-resolved particle number concentrations.  $Q/Q_{exp}$  decreases when the number of factors increases (Fig. S6). Five factors can already explain the whole dataset well. After adding more factors, large size components are separated to several small sub-groups, which cannot explained by environmental process. Therefore, we employ five factors in this analysis.



Fig. S6  $Q/Q_{exp}$  as a function of the number of factors in Size-PMF analysis

Fpeak tests are conducted with the range of -1 to 1 (increments 0.2). Compared to base case runs, fpeak rotations only change the separation and profiles of the factors explaining particles with large sizes and hypothetical regional origin (Factor 4 and 5, Fig S7) with negative fpeak values. The most significant variations occur in Factors 5 with negative fpeak values, which has very low concentrations (Fig. S7 (f)). Positive fpeak values can help to pull the profile of large size components into several modes, which is more similar to the particles coming from regional sources. In addition, according to the residuals, the explanations of large particles are also better with positive Fpeak runs. Thus, we take the fpeak of 1.0 as the results of Size-PMF.

Both profiles and contributions from primary sources (Factor 1, 2 and 3) are stable among all the fpeak runs (Fig. S7 and S6), which is also validated by bootstrap analysis (Fig. S8). Those analyses show the stability of the Size-PMF results.



**Fig. S7** Profiles of different factors with different Fpeak values resolved in size-PMF (sub-fig (a) to (e)) and time series of different factors with different Fpeak values resolved in size-PMF (sub-fig (f) to (j))

Further, we assess the stability of the Size-PMF results through bootstrapping the PMF input (200 iterations). Primary sources (Factor 1, 2 and 3) are stable both in terms of their factor profiles as well as their contributions, while regional sources show larger variations in the large size range, supporting the state that the primary sources are well resolved. The residual to uncertainty ratio (scaled residual) also suggests larger uncertainties in explaining the variability of the largest particles. The time series of scale residual exhibits no clear trend (Fig. S9 (b)).



**Fig. S8** Number profile and volume profiles of different factors in bootstrap runs (sub-fig (a) to (e)). Round dots and error bars represent the average value and one standard variation of all solutions, respectively. Average number concentration of different factors with one standard variation of all runs is displayed in sub-fig (f)



**Fig. S9** Scaled residual as a function of Dp (a) and date (b) in Size-PMF analysis, round dots represent mean Scaled residual and error bars represent the standard deviation. Rescale residual is defined as the ratio of residual to uncertainty

## 2.2 OA-PMF validations

Prior to OA-PMF analysis, we tested constraining cooking, gasoline and diesel profiles in order to test whether vehicular emissions can be separated according to the fuel type. We performed 300 PMF runs times using a random a-value in the range of 0 to 0.1 independently for HOA and COA (increment of 0.05). A relatively small a-value is applied due to the high similarity of the profiles from gasoline and diesel exhausts. The results show clear indications that gasoline and cooking emissions are mixed (similar diurnals, Fig. S10). Based on this test, it seems not possible to separate vehicular emissions in OA-PMF based on the fuel type used in our dataset.



Fig. S10 (a) comparison of source profiles for vehicular exhaust tests, data source: Diesel exhaust 1 (Mohr et al., 2009), Diesel 2 (Sage et al., 2008), Diesel bus exhaust (Canagaratna et al., 2004), gasoline exhaust (Mohr et al., 2009), (b) correlations of source profiles of vehicular exhausts. PMF results (constrained gasoline, diesel, and cooking reference spectra from Mohr et al. (2009), (Canagaratna et al., 2004) and Crippa et al. (2013b), respectively with a-value between 0 and 0.1), c) PMF factor profiles, (d) diurnal variations of PMF factor time series, (e) resolved COA concentrations as a function of gasoline factor concentrations

In OA-PMF analysis, organic mass spectra are imported and analyzed by the multi-linear engine (ME-2) algorithm (ME-2) implemented in the toolkit SoFi, Source Finder. Unconstrained runs exhibit that a mixed POA (primary OA) component and a SOA (secondary OA) component. According to numerous previous researches in Beijing, during our sampling period, BBOA (biomass burning OA) and CCOA (coal combustion OA) are usually the lowest throughout the year(Sun et al.,

2018;Hu et al., 2017). To separate the primary sources, the mass spectral fingerprint of HOA and COA from Crippa (Crippa et al., 2013a;Crippa et al., 2013b) are applied to constrain the PMF runs. Since adding factors cannot further decrease  $Q/Q_{exp}$  ratio, four-factor result is chosen (Fig. S11). In the next step, we perform sensitivity tests by performing 2500 PMF runs times using a random a-value in the range of 0 to 1 independently for HOA and COA(increment of 0.1).



Fig. S11  $Q/Q_{exp}$  as a function of the number of factors in OA-PMF analysis

We evaluate the environment interpretability of all the PMF runs by the following criteria. Only PMF runs that meet all the criteria below are accepted.

- 1. HOA should correlate with BC and NO<sub>x</sub> (significance level of 0.05)
- 2. HOA should correlate better with BC and NOx than COA (significance level of 0.05)
- 3. Lunch peak of COA (12 p.m.) should be higher than the concentration in the morning (9 a.m.)
- 4. Mass spectra of COA is more similar with the reference COA than HOA (assessed by Kendall-Tau)
- 5. Mass spectra of HOA is more similar with the reference HOA than COA (assessed by Kendall-Tau)

Thus, we accept 451 runs and take the average profiles and time series as the final OA-PMF solution. The averaged residuals and scaled residuals of the profiles are displayed in Fig. S12 (a) and (b), respectively. The daily residual of m/z 60 and m/z 115 are displayed in Fig. S12 (c) and (d). The residual profiles exhibit no sign of significant unexplained primary fragments, suggesting the primary sources are well resolved. No significant trend of m/z 115 is observed. The daily average m/z 60 shows a slightly higher level for the first period with very large variations. This may due to the uncertainties in PMF analysis or a bit more biomass burning activities, which is similar with the trend of large particles in the Size-PMF (Regional-related 1 factor). However, considering the low m/z 60 concentrations and residuals as well as the large size of biomass burning particles, it will not significantly affect the primary source estimations in this study.



**Fig. S12** Residual (a) and scaled residual (b, defined as residual to uncertainty) of OA-PMF. Daily average scaled residual of fragments of m/z 60 and 115 are displayed in (c) and (d). The round dots represent the daily average scaled residual and the error bars represent one standard variation



# 2.3 Other supporting information

**Fig. S13** Diurnal patterns of (a) mixing layer height (hourly height within 25<sup>th</sup> and 75<sup>th</sup> percentiles), (b) light-duty vehicle flow, (c) truck flow, and (d) bus flow during sampling period



**Fig. S14** Diurnal patterns of NOx (a) in winter time (Dec  $1^{\text{st}}$ , 2018 to Feb 28<sup>th</sup>, 2019). The range represented hourly concentrations within 25<sup>th</sup> and 75<sup>th</sup> percentiles.



**Fig. S15** (a) Sampling site and surrounding environment (modified from Kokkonen et al. (2019) with authors'permission) and, (b) wind frequencies and wind speed for different time of the day in the sampling period. The area of dots are proportional to the wind frequencies. N freq. = North ( $337.5^{\circ}$  to  $22.5^{\circ}$ ) wind frequencies, NE freq. = North East ( $22.5^{\circ}$  to  $67.5^{\circ}$ ) wind frequencies, E freq. = East ( $67.5^{\circ}$  to  $112.5^{\circ}$ ) wind frequencies, SE freq. = South East ( $112.5^{\circ}$  to  $157.5^{\circ}$ ) wind frequencies, S freq. = South ( $157.5^{\circ}$  to  $202.5^{\circ}$ ) wind frequencies, SW freq. = South West ( $202.5^{\circ}$  to  $247.5^{\circ}$ ) wind frequencies, W freq. = West ( $247.5^{\circ}$  to  $292.5^{\circ}$ ) wind frequencies, NW freq. = North West ( $292.5^{\circ}$  to  $337.5^{\circ}$ ) wind frequencies, and WS = wind speed



**Fig. S16** The comparison of relative ionization efficiency of COA from this study and from cooking emission source tests. Square dots represented the median ACSM COA and SMPS cooking concentrations of the time of the day and coloured with different cooking hours. Error bars represent  $25^{\text{th}}$  and  $75^{\text{th}}$  percentiles. The diamonds represented the AMS and SMPS concentrations from cooking emission source tests (shown in black) and the error bars means the standard deviations of different experiments (Reyes-Villegas et al., 2018). In this plot, the concentrations and error bars from cooking source tests are multiplied with a factor of 0.5. The green and orange regions showed the RIEs for OA typically applied (Xu et al., 2018) and cooking source test (Reyes-Villegas et al., 2018). The yellow and green dash line represent the average RIE<sub>COA</sub> of 2.29 from cooking sources tests and typical default RIE<sub>OA</sub> of 1.4.

#### 3. Gas phase molecular marker



Fig. S17 Peak fitting of (a) linoleic acid (C<sub>18</sub>H<sub>32</sub>O<sub>2</sub>) and (b) pyrogluamic acid (C<sub>5</sub>H<sub>7</sub>NO<sub>3</sub>)



**Fig. S18** Daily comparison between COA mass concentration and normalized signal intensity of (a) linoleic acid ( $C_{18}H_{32}O_2$ ) and (b) pyrogluamic acid ( $C_5H_7NO_3$ ). The normalized signals intensities are defined as the ratio of the raw signal intensities of those compounds to the total reagent ions (sum of  $NO_3^-$ ,  $HNO_3 \cdot NO_3^-$  and  $(HNO_3)_2 \cdot NO_3^-$ ). Daily averaged concentrations are applied due to it is less affected by diurnal variations of temperature and photochemistry. To compare with gas phase procurers in a long period, here COA concentration was extracted from all sampling days, which has a good correlation with the COA extracted from non-NPF days in this study (Slope=0.99, r=0.89).



Fig. S19 Average particle size distribution of NPF days, Haze days, no Haze nor NPF event days and days used in PMF.



**Fig. S20** Normalized median diurnal variations (normalized by their highest median hourly value) of (a) HOA plus BC (black dash line) and Traffic-fine resolved from Size-PMF (black line); (b) Traffic-ultrafine from Size-PMF; (c) COA from ACSM (yellow dash line) and Cooking-related particles resolved from SMPS (yellow line); (d) Secondary aerosols (SIA+SOA) from ACSM (green dash line) and Regional-related from Size-PMF(green line)

Sampling site	Sampling year	Source types	Sampling equipment	Reference
Augsburg, Germany	Winter 2007/07	Re-suspended dust, fresh/aged traffic, combustion, long-range transported dust, nucleation, secondary aerosols	UDMA- UCPC/DMA- CPC/APS	Gu et al. (2011)
Barcelona, Spain	Jan 2013–Dec 2016	Photonucleation, traffic nucleation, urban, secondary	SMPS	Rivas et al. (2020)

Table S1. Sources identified by Size-PMF method

	Nov 2003–Dec 2004	Road traffic, mineral dust, industry, sea spray, photonucleation, regional, combustion	DMPS	Pey et al. (2009)
Beijing, China	August 2008	Local/distant traffic, secondary nitrate, combustion	TDMPS	Wang et al. (2013)
	Aug–Sep, 2015	Nucleation, local primary emissions (e.g., cooking and traffic emissions), secondary	SMPS	Du et al. (2017)
Helsinki, Finland	Feb 2015–Aug 2017, Jan 2007–Dec 2016	Photonucleation, traffic nucleation, fresh traffic, urban, biogenic, secondary	DMPS	Rivas et al. (2020)
	Jan 2010–Dec 2016, Mar 2014–Dec 2016	Photonucleation, traffic nucleation, fresh traffic, urban, secondary	SMPS	Rivas et al. (2020)
London, UK	Oct–Nov, 2007	Road emissions (vehicle exhaust, brake dust, resuspension), Urban background (accumulation mode, suburban traffic, solid fuel/nitrate, regional, cooking, regional)	SMPS/APS	Harrison et al. (2011)
Pittsburgh, US	Jun–Aug, 2001	Local/distant traffic, secondary nitrate, regional transport, combustion	Nano- SMPS/SMPS/APS	Zhou et al. (2004)
Rochester, NY, US	Dec 2004–Nov 2005	Nucleation, traffic, industry, heating, secondary nitrate, secondary sulfate, regional transport	SMPS	Ogulei et al. (2007)
Zurich, Switzerland	Dec 2010–Oct 2014	Photonucleation, traffic nucleation, fresh traffic, urban, secondary	SMPS	Rivas et al. (2020)

Sampling time	Source	Prop. to OA	Size	Reference
Jul–Sep 2008	НОА	18.1%		
	COA	24.4%	$PM_1$	Huang et al. (2010)
	OOA1	33.7%		
	OOA2	34.7%		
	HOA	13%	PM <sub>1</sub>	Hu et al. (2016)
A C 2011	COA	21%		
Aug–Sep 2011	LO-OOA	28%		
	MO-OOA	37%		
	HOA	11.4%		
	COA	19.9%		Hu et al. (2017)
Jul–Aug 2012	LO-OOA	42.6%	$PM_1$	
	MO-OOA	26.1%		
	FFOA	9%		Sun et al. (2018)
	COA	13%	$PM_1$	
May 2013	BBOA	6%		
	LO-OOA	14%		
	MO-OOA	58%		
	FFOA	5.5%		
	COA	12.3%	$PM_1$	Zhou et al. (2018)
Oct 2014	BBOA	10.0%		
	LO-OOA	15.3%		
	OOA	53.5%		
	НОА	8%		
Jul–Aug 2015	COA	18%	$PM_1$	Duan et al. (2020)
	ISOOA (isoprene- oxidized OA)	5%	-	

Table S2. The mass fractions of resolved OA component in Beijing conducted in non-heating season period

	OOA	68%		
Jun 2017	HOA	13%		
	COA	15%	$PM_1$	$Y_{22} = (-1)(2010)$
	LO-OOA	45%		Xu et al. (2019)
	MO-OOA	27%		
	HOA	11%		
M 1 2019	COA	24%	DM	<b>X</b> (1(2010)
May–Jun 2018	LO-OOA	39%	$PM_1$	Xu et al. (2019)
	MO-OOA	26%		
Apr–July 2018	HOA	12%		
	COA	15%	PM <sub>2.5</sub> This st	
	LO-OOA	9%		This study
	MO-OOA	64%		

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