



## Supplement of

# $\ensuremath{\text{PM}}_1$ composition and source apportionment at two sites in Delhi, India, across multiple seasons

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## 1 Supplement of: PM<sub>1</sub> composition and source apportionment at two sites in Delhi, India

## 2 across multiple seasons

**3** S1. Monitoring sites, meteorology and dates.



Figure S1 Location of the monitoring sites. Image taken from www.googlemaps.co.uk IGDTUW located at Old
Delhi Lat 28.588°, Lon 77.217° and IMD located at New Delhi Lat 28.664°, Lon 77.232°.



Figure S2 Maps showing the surroundings of ND (a) and OD (b). Red circle shows location of the monitoringsites.



- 35 Figure S3 Box plots with temperature (a), wind speed (b) and relative humidity (c) for the different seasons.
- 36 The marker represents the mean.

Instrument	Event	Aditional instrumentation
ACSM	Win_ND_A	Aethalometer AE33
HR-AMS_1	PreM_ND_H1	Aethalometer AE31
cToF-AMS	PreM_OD_C	Aethalometer AE31
HR-AMS_2	PreM_OD_H2	Aethalometer AE31
HR-AMS_2	Mon_OD_H2	Aethalometer AE31
HR-AMS_2	PostM_OD_H2	Aethalometer AE31, SP2, PTR-MS, Nox, CO
cToF-AMS	PostM_OD_C	Aethalometer AE31
HR-AMS_2	PostM_OD_T_H2	Aethalometer AE31
cToF-AMS	PostM_ND_T_C	МААР

37 Table S1 Collocated instruments with the mass spectrometers.

38

## 39 S2. AMS quality assurance analysis. NR-PM<sub>1</sub> concentrations and relative contribution

- 40 S2.1 Calibrations and collection efficiency estimation.
- 41 Table S2. Nitrate ion efficiency (IE) and relative IE (RIE) for NH4<sup>+</sup>, SO4<sup>2-</sup> and Cl<sup>-</sup> from calibrations performed on
- 42 the aerosol mass spectrometer instruments. <sup>a</sup>preflux period (11/10/18 03/11/18). <sup>b</sup>Diwali period (05/11/18 -
- 43 14/11/18). <sup>c</sup>post Diwali (14/11/18 23/11/18).

Instrument	Season	IE	$RIE_NH_4^+$	RIE_SO42-	RIE_CI <sup>-</sup>	CE
cToF-AMS	PreM	1.55E-07	4.01	1.17	1.5	0.5
cToF-AMS	PostM	2.40E-07	4.6	1.2	1.7	0.5
HR-AMS_1	PreM	3.25E-07	4	1.31	1.3	0.5
HR-AMS_2	PreM	2.92E-07	4	1.45	2.07	0.5
HR-AMS_2	Mon	2.92E-07	4	1.45	2.07	0.5
HR-AMS_2	<sup>a</sup> PostM	2.89E-07	4	1.45	2.07	0.5
HR-AMS_2	<sup>b</sup> PostM	3.14E-07	4	1.45	1.05	0.8
HR-AMS_2	<sup>c</sup> PostM	3.14E-07	4	1.45	1.05	0.5

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Collocated PM<sub>2.5</sub> measurements were performed in a Digitel sampler (DH-77 Digitel Enviro-sense) with 47 48 a flow rate of 500 L/min. collecting 12-hour samples in guartz fibre filters. The samples were analysed 49 with Ion Chromatography (IC) to measure anion and cation data, including blank subtraction. 50 Components analysed include phosphate, nitrate, bromide, sulphate, nitrite, chloride, fluoride, K<sup>+</sup>, 51 Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH4<sup>2-</sup> and Na<sup>+</sup>. Collected filters were kept frozen and transported to the University of 52 Birmingham for analysis. For the IC analysis, deionized water was used for blank determination and 53 subtraction. 10 mil of DI water were added to samples. Extraction tubes were sonicated for 1 hour 54 with bath temperature not exceeding 27 °C. Next day filter the extract solution for each sample tube 55 using a 10 ml plastic filter and 0.45 µm syringe filter (star labs) into a new labelled polypropylene tube, 56 finally, the sample is ready for IC analysis.

A Partisol (2025i, ThermoFisher Scientific) was deployed to perform 6-hour gravimetric PM<sub>2.5</sub> mass.
 These measurements are used in this manuscript to determine the collection efficiency of the HR AMS 2.

60 For HR-AMS\_2, a CE = 0.5 was used for preM and postM preflux tower periods, which was determined

by comparing AMS+BC with gravimetric  $PM_{2.5}$  (Fig. S4.b) and  $Cl^2$ ,  $NO_3^2$  and  $SO_4^{2^2}$  quantified by IC from

62 filter measurements (Fig. S6). In the PostM flux period, for the HR-AMS\_2, a CE = 1.0 was derived after

63 comparison with total  $PM_{2.5}$  (fig. S4.d). For the HR-AMS\_1 measurements, a CE = 0.5 after the

64 intercomparison with the HR-AMS\_2 (fig. S7). The ACSM manual recommends using a CE = 0.5.

Figure S5 presents the time series of PM<sub>1</sub> online measurements (HR-AMS\_2 + BC) and total gravimetric

66 PM2.5 concentrations. Using CE = 0.5 (Fig. 5.b) shows the best agreement between  $PM_1$  and  $PM_{2.5}$ ,

 $67 \qquad \text{with a } \mathsf{PM}_{2.5}:\mathsf{PM}_1 \text{ ratio going from } 0.8 \text{ to } 1.4.$ 



69



with HR-AMS\_2 and aethalometer (BC) measurements. All AMS+BC measurements are averaged according to filter
 sampling times.



76 Figure S5 Analysis of the HR-AMS\_2 and total gravimetric PM<sub>2.5</sub> for the PostMon preflux period. Time series of averaged PM<sub>1</sub> (AMS + BC) (black line) and gravimetric PM<sub>2.5</sub> (blue line) for CE =1 (a), CE = 0.5 77

78 (b) and CDCE (c). The PM<sub>2.5</sub>:PM<sub>1</sub> ratio is shown in red. All AMS+BC measurements are averaged

79 according to filter sampling times.





- 84 chromatography by the University of Birmingham for CE = 1, CE = 0.5 and composition dependant CE
- 85 (CDCE). All AMS+BC measurements are averaged according to filter sampling times.

86 S2.1 cToF-AMS and HR-AMS 2 intercomparison.

87 An intercomparison was performed between the cToF-AMS and the HR-ToF-AMS (fig. S7a and S7b), 88 deployed at OD over pre-monsoon in order to perform an intercomparison (28/May – 09/June), 89 obtaining average concentrations, in  $\mu$ gm<sup>-3</sup>, of 15.0 and 19.1 of Org, 1.7 and 1.6 of NO<sub>3</sub><sup>-</sup>, 6.8 and 8.3 of SO<sub>4</sub><sup>2-</sup>, 2.5 and 2.6 of NH<sub>4</sub><sup>+</sup>, 0.4 and 0.5 of Cl<sup>-</sup> for cToF-AMS and HR-ToF-AMS respectively. The total 90 91 aerosol concentration for cToF-AMS is 26.5  $\mu$ g.m<sup>-3</sup> and for HR-ToFAMS is 32.1  $\mu$ g.m<sup>-3</sup>, a difference of 92 21%. This is well within the range of previous AMS comparison studies. Crenn et al. (2015) estimated 93 an organic mass uncertainty of 19 %. Bahreini et al. (2009) estimated an overall uncertainty of 35 %, agreeing with other AMS studies (DeCarlo et al., 2008;Dunlea et al., 2009). Recently, a 50% uncertainty 94 95 has been reported by Shinozuka et al. (2020).

96 A PMF analysis was performed to the Org concentrations measured by the cToF-AMS and the HR-ToF-97 AMS. Figure 7c shows the mass spectra comparison of the factor profiles identified and figure 7.d shows the triangle plot, f44 – f43, to compare and describe OOA. The doted lines represent the space 98 99 proposed by Ng et al. (2011) to characterise OOA. The parameters f43 and f44 represent the ratio of 100 the integrated signal at m/z 43 and m/z 44 to the total signal in the organic component mass spectrum. 101 The same OA factors were identified in the two PMF analyses, HOA, MO-OOA, BBOA, COA and LO-

102 OOA. This analysis verifies the AMS intercomparison, with the same OA factors and similar ageing (f44f43).

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Figure S7. Average concentrations (S7.a) and relative contribution (S7.b) of Org, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and Cl<sup>-</sup> for 106 107 the cToF-AMS and the HR-ToF-AMS. Mass spectra comparison from PMF analysis (S7c) and f44-f43 triangle 108 plot (S7.d) to compare OA ageing according to Ng et al. (2011).

109 Table S3. Statistics of NR-PM1 and BC measurements for the various seasons. Minimum, maximum, average, 110 standard deviation, median and number of points.

	Org						no3							so4					
	min	max	avg	SD	median	npnts	min	max	avg	SD	median	npnts	min	max	avg	SD	median	npnts	
Win_ND_A	5.27	197.60	39.88	32.22	29.98	2169	0.18	21.05	3.69	2.96	2.82	2169	0.84	9.77	3.90	1.82	3.66	2169	
PreM_ND_H1	-0.67	788.67	23.59	21.76	19.58	40569	-0.05	16.44	2.33	2.21	1.55	40569	-0.11	21.57	5.03	3.24	4.10	40569	
PreM_OD_C	3.01	283.67	30.20	26.21	24.10	3196	0.42	15.27	3.41	2.59	2.48	3196	0.24	30.12	13.54	5.11	12.39	3196	
PreM_OD_H2	-0.05	291.76	29.16	22.96	26.12	11808	0.02	16.28	2.31	2.39	1.50	11808	0.00	43.17	13.05	6.39	12.71	11808	
Mon_OD_H2	-0.11	195.86	24.98	16.63	21.43	4384	0.02	22.88	3.14	2.66	2.41	4384	-0.01	37.77	11.29	5.56	9.90	4384	
PostM_OD_H2	9.02	458.35	95.35	68.11	75.06	6798	1.65	58.66	11.45	8.15	9.25	6798	2.14	30.09	11.01	5.33	10.24	6798	
PostM_OD_C	13.19	206.78	62.97	39.39	55.12	1044	0.72	35.73	7.69	6.80	5.24	1044	1.71	25.19	8.82	3.54	8.77	1044	
PostM_OD_T_H2	10.37	1211.58	87.85	77.59	75.98	1785	1.79	28.09	12.02	5.19	12.20	1785	0.55	52.66	6.77	4.61	6.22	1785	
PostM_ND_T_C	13.93	331.73	73.23	42.31	62.77	1506	1.56	42.72	14.44	8.23	13.51	1506	-1.76	35.49	6.58	3.13	6.55	1506	
	nh4						chl						BC						
	min	max	avg	SD	median	npnts	min	max	avg	SD	median	npnts	min	max	avg	SD	median	npnts	
Win_ND_A	-2.40	28.74	4.54	4.11	3.07	2169	-0.10	67.59	5.96	9.14	2.35	2169	1.064	77.02	15.91	13.62	10.69	2144	
PreM_ND_H1	-1.50	16.58	2.63	1.78	2.18	40568	-0.30	26.57	1.15	1.75	0.54	40569	0.354	8.929	2.529	1.46	2.324	529	
PreM_OD_C	-6.19	13.66	5.02	1.90	4.63	3196	0.03	22.73	0.80	1.78	0.26	3196	0.027	51.53	5.918	4.731	4.646	2684	
PreM_OD_H2	-0.01	20.79	4.16	2.35	3.90	11808	0.00	33.75	1.29	2.57	0.44	11808	0.027	51.53	5.895	5.367	4.435	6688	
Mon_OD_H2	0.00	18.23	3.75	2.04	3.39	4384	0.00	34.30	0.99	1.66	0.37	4384	0.076	20.02	3.936	2.642	3.208	4314	
PostM_OD_H2	0.66	37.60	7.47	5.31	6.06	6798	0.07	59.96	6.05	7.48	2.91	6798	0.608	53.76	11.59	10.01	8.08	5499	
PostM_OD_C	-8.22	15.18	5.02	3.19	4.22	1044	0.07	28.69	3.91	5.00	2.08	1044	1.114	31.76	7.783	5.563	6.93	914	
PostM_OD_T_H2	0.55	44.50	8.75	5.16	8.69	1785	0.30	73.80	9.67	10.91	5.43	1785	1.585	59.52	17.27	13.56	12.24	1497	
PostM_ND_T_C	-5.54	28.43	7.41	4.04	7.09	1506	0.15	56.52	5.03	5.66	3.07	1506	3.316	38.17	16.52	7.708	16.03	1103	



Figure S8. Aerosol time series of the various measurements. All concentrations are in μg.m<sup>-3</sup>. OA concentrations are plotted on the right axis and the rest of the compounds are plotted on the left axis.

### 124 S3. PMF analysis

- The selection of the optimal PMF solution was performed following recommendations in previous studies
  (Canonaco et al., 2013;Crippa et al., 2014;Reyes-Villegas et al., 2016) and exploring between 8-10 PMF solutions
  with 3, 4, 5 and 6 factors; looking at different seeds (a random starting point of the PMF solution), which resulted
  on analysing around 40 PMF solutions per season.
- 129 The following criteria was used to select the optimal PMF solution:
- 130 1. Residuals to be closest to zero.
- 131 2.  $Q/Q_{exp}$  value closest to one.
- **132** 3. High correlation between HOA and NOx.
- **133** The PMF analysis was performed using the Source Finder (SoFi 4.8) tool (Canonaco et al., 2013) In order to select
- the optimal PMF solution, we analysed the overall Q/Qexp (Figure S9), the row (time series) Q/Qexp (Figure
   S10) and the Squares of scaled residual (Figure S11). The overall Q/Qexp is calculated as the mean of the time
- 136 series Q/Qexp.
- 137 In all the PMF analyses, an improvement in the residuals and  $Q/Q_{exp}$  was observed when increasing the number
- 138 of factors (Fig. S9), with Q/Qexp values of around 5.52 for 4-factor solutions and values of around 5.2 for 5-factor
- solutions. However, the 6-factor solutions presented two factors with similar time series and mass spectra,characteristic of factor splitting. Hence, the 5-factor solution was chosen to be further analysed.
- 140 characteristic of factor splitting. Hence, the 5-factor solution was chosen to be further analysed.
- 141 The following figures show the PMF solution space to select the optimal PMF solution for the winter New Delhi
- 142 ACSM dataset, Win\_ND\_A. The same analysis was performed to the other datasets to determine the optimal PMF
- solution for each season for further analysis presented in the manuscript.
- 144 The solutions are labelled as follows: PMF\_4F\_S1 is the 4-factor solution (4F) seed number one (S1). The optimal
- solution of this season is PMF\_5F\_S2. This solution showed the lowest average residuals and Q/Qexp value
- 146 (Figure S9). In figure S12 the Pearson values from linear regressions between the PMF factors and NOx are
- displayed. NOx is a pollutant well-known to be related to traffic emissions, thus a high Pearson value is expected
- between HOA and NOx. High pearson values between 0.77 0.785 were observed with the highest Pearson value
- 149 to be found with the  $PMF_5F_S2$  solution (0.785).
- 150 The overall Q/Qexp = 5.2 is calculated as the mean of the time series Q/Qexp. The overall Q/Qexp can also be 151 estimated from the Squares of scaled residuals. We calculate the sum of the values (Squares of scaled residual)
- plotted in Figure S11 for the specific PMF run. For example, for the optimal solution (PMF\_5F\_S2) the sum =
  11269.56.
- 154 We calculate the Qexp = n \* m p\*(n + m) = 147127 for the 5-factor solution, where
- 155 n = num of samples = 2169, m = num of m/z = 73, p = num of factors = 5
- 156 We also divide the Qexp by m, which gives 2015.44.
- 157 Finally, Q/Qep = 11269.56/2015.44 = 5.59, which is close to the overall Q/Qexp = 5.2.
- Figures S10 and S11 are used to identify particularly high Q/Qexp values for m/z or episodes with high Q/Qexp on time series that might require further analysis. For example, we can see an episode with high Q/Qexp values on 20/20/2018 (Figure S10), which is related to an episode with high Org concentrations. Also, we can see an improvement on both time series (Figure S10) and m/z (Figure S11) plots when going from 4-factor to 5-factor solutions. However, no remarkable improvement
- between 5-factor solution was observed. Hence, we can use the overall Q/Qexp to select the optimalsolution.
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- 166



Figure S9. Summary plots of residuals and Q/Qexp values for 4 and 5 factor solutions (a) and a close up to the 5-factor solutions (b)



## 170

## 171 Figure S10. Time series of residuals and Q/Qexp values.



173 Figure S11. Residuals and Squares of scaled residual for m/z.





176 Figure S12. Pearson values of the linear regressions between the PMF factors and NOx (a) and babs\_950t (b).

Table S4. Statistical parameters [ $\mu$ g.m<sup>-3</sup>] of the PMF factors obtained from OA, measured with the HR- AMS\_2,

178 for all the various seasons. This data is analysed on detail by Cash et al. (2020), we present this data here to

179 compare with the other PMF-AMS datasets.

		All_OD_H	2 (includes PreM, Mon and PostM)												
	COA	HOA_	LVOOA	nHOA	SFOA	<b>SVBBOA</b>	SVOOA	HOA	BBOA	POA	SOA	TOA	POA ratio		
mean	7.44	9.35	13.72	4.87	6.64	7.03	5.36	14.21	13.67	35.32	19.08	54.40	0.55		
median	5.26	4.31	12.14	0.30	0.74	0.04	4.00	4.88	1.80	14.74	17.81	32.83	0.55		
sd	9.55	14.30	10.89	12.65	16.05	12.81	5.85	25.71	26.45	51.35	13.21	58.69	0.21		
min	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
max	202.87	189.56	90.90	337.90	665.03	91.39	66.84	337.90	709.60	1250.37	103.93	1250.37	1.00		
		PreM_OD	_H2												
	COA	HOA_	LVOOA	nHOA	SFOA	<b>SVBBOA</b>	SVOOA	HOA	BBOA	POA	SOA	TOA	POA ratio		
mean	7.84	5.13	10.59	0.83	1.16	0.53	3.26	5.96	1.68	15.48	13.85	29.33	0.52		
median	5.61	3.07	10.68	0.00	0.07	0.00	1.57	3.25	0.43	10.15	14.18	26.44	0.50		
sd	8.73	6.90	7.36	2.46	3.51	1.18	4.76	8.80	3.91	17.95	9.70	23.54	0.22		
min	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00		
max	131.02	118.38	53.01	30.06	75.91	12.97	56.72	136.58	84.70	233.64	61.50	290.36	1.00		
		Mon OD H2		Mon_OD_H2											
	COA	HOA_	LVOOA	nHOA	SFOA	<b>SVBBOA</b>	SVOOA	HOA	BBOA	POA	SOA	TOA	POA ratio		
mean	2.83	6.55	6.71	0.76	1.36	0.01	6.81	7.31	1.37	11.51	13.52	25.03	0.45		
median	1.79	4.84	6.32	0.19	0.50	0.00	5.41	5.18	0.51	8.58	12.90	21.35	0.43		
sd	3.57	5.62	5.11	1.33	3.33	0.11	5.80	6.68	3.34	11.09	8.37	16.77	0.19		
min	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01		
max	41.95	59.34	35.68	14.61	60.78	3.07	66.84	67.70	60.78	130.85	66.84	197.70	1.00		
		PostM O	stM OD H2												
	COA	HOA	LVOOA	nHOA	SFOA	<b>SVBBOA</b>	SVOOA	НОА	BBOA	POA	SOA	TOA	POA ratio		
mean	8.54	17.78	20.56	12.95	14.90	16.85	6.97	30.73	31.75	71.02	27.53	98.55	0.63		
median	6.70	8.67	19.34	4.47	7.98	11.98	5.77	13.20	21.52	48.61	25.28	77.06	0.64		
sd	7.72	21.96	11.51	17.97	18.49	15.35	5.83	39.27	30.56	64.55	13.07	69.33	0.17		
min	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	3.25	0.00	9.05	0.24		
max	101.75	189.56	90.90	119.01	186.96	91.39	44.53	281.55	247.80	456.41	103.93	460.48	1.00		
		PostM_OD_T_H2													
	COA	HOA	LVOOA	nHOA	SFOA	<b>SVBBOA</b>	SVOOA	НОА	BBOA	POA	SOA	TOA	POA ratio		
mean	9.56	11.21	24.39	9.26	21.35	26.87	8.65	20.46	48.22	78.25	33.04	111.29	0.64		
median	7.22	6.51	21.51	2.93	11.05	25.44	7.89	10.19	37.41	61.87	30.82	96.55	0.63		
sd	15.55	13.09	14.51	21.27	36.38	13.00	5.53	28.83	44.79	80.12	14.63	81.24	0.14		
min	0.00	0.00	0.00	0.00	0.00	1.76	0.00	0.00	2.72	5.57	0.00	14.65	0.38		
max	202.87	86.99	63.69	337.90	665.03	76.43	29.20	337.90	709.60	1250.37	70.89	1250.37	1.00		

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POA = primary OA, SOA = secondary OA, TOA = SOA +POA. This analysis identified 7 PMF factors, we are adding
 HOA = HOA\_ + nHOA and BBOA = SFOA + SVBBOA to compare with our 5-factor solutions in the main manuscript.

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185 Figure S13 show the triangle plot to analyse f44 – f43 compare and describe OOA. The doted lines represent the 186 space proposed by Ng et al. (2011) to characterise OOA. The parameters f43 and f44 represent the ratio of the 187 integrated signal at m/z 43 and m/z 44 to the total signal in the organic component mass spectrum. We can see 188 the typical behaviour of MOOOA with high f44 values compared to LOOOA, characteristic of a more aged, 189 oxygenated OA. MOOOA, in black, while having different values, is found in a distinct area in the plot with f44 190 between 0.18 -0.26, while LOOA with low f44, 0.10 and lower, and high f43, characteristic of fresher OOA when 191 compared with MOOOA. HOA, in brown, has a low f44, close to zero, and distinct f43 values of 0.08 - 0.13. oPOA, 192 in purple, has slightly high values of both f44 and f43, agreeing with the identification as to be oxygenated 193 primary organic aerosol. This analysis suggests a good separation on the oxygenated species between factor 194 profiles and shows an f43 cluster of HOA.

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Figure S13. f44 vs f43 for all the periods and all the factor profiles and identified with PMF analysis. The symbols representthe sampling periods/sites and the colours define the PMF factor profiles.

#### 199 S4. Aethalometer analysis

### 200 S4.1 Aethalometer AE-31 correction and model OD

The data collected with the aethalometer model AE-31 needs to be corrected from loading and scattering effects. The Weingarten model (Weingartner et al., 2003) has been applied using a filter loading factor f=1.30 and a multiple scattering constant C=2.8, which was calculated as the slope between BC from SP2 measurements and BC from Aethalometer after filter loading corrections. Figure S14 shows the corrected BC concentrations

from the Aethalometer (red) and the BC concentrations of the SP2.





207 Figure S14. Intercomparison of BC measurements between aethalometer AE-31 and SP2

208 The aethalometer model was applied following the Sandradewi approach (Sandradewi et al., 2008) using 209 absorption angstrom exponent traffic  $\alpha_{\rm ff} = 0.8$  and wood burning  $\alpha_{\rm bb} = 2.0$  (Fig. S15).



Figure S15. Aethalometer model absorption coefficients for fossil fuels (babs\_950ff) and biomass burning (babs\_470bb).

A sensitivity test was performed to determine  $\alpha_{ff} = 0.8$ . and  $\alpha_{bb} = 2.0$ , varying  $\alpha_{ff}$  from 0.4 to 2 and  $\alpha_{bb}$  from 1.4 to 2.6 and increments of 0.1. Figure S16 shows example plots of the performed analysis. An improvement was observed when using  $\alpha_{ff}$  of 0.8 compared to 1.0, thus a value of 0.8 was derived (figures S16.a and S16.b). No significant changes were observed when testing different  $\alpha_{bb}$  values (figures S16.c and S16.d), thus the default value of 2.0 was used (Fig. S16). A similar analysis was performed to select values for the subsequent Aethalometer model analyses.



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Figure S16. Sensitivity test to select  $\alpha_{\rm ff} = 0.8$ . The peak marked in panel (a) relates to the Diwali celebrations.

## 222 S4.2 Aethalometer model outputs AE-31 PreM\_ND.

223 The aethalometer model was applied following the Sandradewi approach (Sandradewi et al., 2008) using an 224 absorption angstrom exponent for fossil fuel of  $\alpha_{ff} = 0.8$  and for biomass burning of  $\alpha_{bb} = 2.0$  (Fig. S17), after 225 doing a sensitivity test.

226





Figure S17. Aethalometer model absorption coefficients for traffic (b<sub>abs\_950 ff</sub>) and wood burning (b<sub>abs\_470 bb</sub>).

229

#### 231 S4.3 Aethalometer model outputs AE-33 ND-Winter.

The aethalometer model was applied following the Sandradewi approach (Sandradewi et al., 2008) using absorption angstrom exponent fossil fuel  $\alpha_{\rm ff} = 1.0$  and biomass burning  $\alpha_{\rm bb} = 2.0$  (Fig. S18), after doing a sensitivity test.



237 Figure S18. Aethalometer model applied to AE-33 model in Winter.





Figure S19. Polar plots of OA factors median concentrations [μg.m<sup>-3</sup>]. Due to the low number of data points for
 OD\_PreM\_cToF\_AMS to plot polar plots, pollution roses are presented.



243 Figure S20. Polar plots of various aerosols. Median concentrations [μg.m<sup>-3</sup>].

## 245 **References**

- Bahreini, R., Ervens, B., Middlebrook, A. M., Warneke, C., de Gouw, J. A., DeCarlo, P. F., Jimenez, J. L.,
  Brock, C. A., Neuman, J. A., Ryerson, T. B., Stark, H., Atlas, E., Brioude, J., Fried, A., Holloway, J. S.,
  Peischl, J., Richter, D., Walega, J., Weibring, P., Wollny, A. G., and Fehsenfeld, F. C.: Organic aerosol
  formation in urban and industrial plumes near Houston and Dallas, Texas, Journal of Geophysical
  Research: Atmospheres, 114, <u>https://doi.org/10.1029/2008JD011493</u>, 2009.
- 252 Canonaco, F., Crippa, M., Slowik, J. G., Baltensperger, U., and Prevot, A. S. H.: SoFi, an IGOR-based 253 interface for the efficient use of the generalized multilinear engine (ME-2) for the source 254 apportionment: ME-2 application to aerosol mass spectrometer data, Atmos Meas Tech, 6, 3649-255 3661, DOI 10.5194/amt-6-3649-2013, 2013.
- Cash, M. J., Langford, B., Di Marco, C., Mullinger, N., Allan, J., Reyes-Villegas, E., Ruthambara, J., Heal,
   M., Acton, W., F. J., Drysdale, W., Tuhin, M., Shivani, Ranu, G., and Nemitz, E.: Seasonal analysis of
   submicron aerosol in Old Delhi using high resolution aerosol mass spectrometry: Chemical
   characterisation, source apportionment and new marker identification, In-preparation, 2020.
- 260 Crenn, V., Sciare, J., Croteau, P. L., Verlhac, S., Fröhlich, R., Belis, C. A., Aas, W., Äijälä, M., Alastuey, A.,
- Artiñano, B., Baisnée, D., Bonnaire, N., Bressi, M., Canagaratna, M., Canonaco, F., Carbone, C., Cavalli,
   F., Coz, E., Cubison, M. J., Esser-Gietl, J. K., Green, D. C., Gros, V., Heikkinen, L., Herrmann, H., Lunder,
- 263 C., Minguillón, M. C., Močnik, G., O'Dowd, C. D., Ovadnevaite, J., Petit, J. E., Petralia, E., Poulain, L.,
- 264 Priestman, M., Riffault, V., Ripoll, A., Sarda-Estève, R., Slowik, J. G., Setyan, A., Wiedensohler, A.,
- 265 Baltensperger, U., Prévôt, A. S. H., Jayne, J. T., and Favez, O.: ACTRIS ACSM intercomparison Part 1:
- Reproducibility of concentration and fragment results from 13 individual Quadrupole Aerosol
   Chemical Speciation Monitors (Q-ACSM) and consistency with co-located instruments, Atmos. Meas.
- 268 Tech., 8, 5063-5087, 10.5194/amt-8-5063-2015, 2015.
- 269 Crippa, M., Canonaco, F., Lanz, V. A., Aijala, M., Allan, J. D., Carbone, S., Capes, G., Ceburnis, D.,
- 270 Dall'Osto, M., Day, D. A., DeCarlo, P. F., Ehn, M., Eriksson, A., Freney, E., Ruiz, L. H., Hillamo, R., Jimenez,
- J. L., Junninen, H., Kiendler-Scharr, A., Kortelainen, A. M., Kulmala, M., Laaksonen, A., Mensah, A.,
- 272 Mohr, C., Nemitz, E., O'Dowd, C., Ovadnevaite, J., Pandis, S. N., Petaja, T., Poulain, L., Saarikoski, S.,
- 273 Sellegri, K., Swietlicki, E., Tiitta, P., Worsnop, D. R., Baltensperger, U., and Prevot, A. S. H.: Organic
- aerosol components derived from 25 AMS data sets across Europe using a consistent ME-2 based
  source apportionment approach, Atmos Chem Phys, 14, 6159-6176, DOI 10.5194/acp-14-6159-2014,
  2014.
- 277 DeCarlo, P. F., Dunlea, E. J., Kimmel, J. R., Aiken, A. C., Sueper, D., Crounse, J., Wennberg, P. O.,
- 278 Emmons, L., Shinozuka, Y., Clarke, A., Zhou, J., Tomlinson, J., Collins, D. R., Knapp, D., Weinheimer, A.

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- J., Montzka, D. D., Campos, T., and Jimenez, J. L.: Fast airborne aerosol size and chemistry
   measurements above Mexico City and Central Mexico during the MILAGRO campaign, Atmos Chem
   Phys, 8, 4027-4048, 2008.
- 282 Dunlea, E. J., DeCarlo, P. F., Aiken, A. C., Kimmel, J. R., Peltier, R. E., Weber, R. J., Tomlinson, J., Collins,
- D. R., Shinozuka, Y., McNaughton, C. S., Howell, S. G., Clarke, A. D., Emmons, L. K., Apel, E. C., Pfister,
- G. G., van Donkelaar, A., Martin, R. V., Millet, D. B., Heald, C. L., and Jimenez, J. L.: Evolution of Asian
- aerosols during transpacific transport in INTEX-B, Atmos. Chem. Phys., 9, 7257-7287, 10.5194/acp-9-
- 286 7257-2009, 2009.
- Ng, N., Canagaratna, M., Jimenez, J., Chhabra, P., Seinfeld, J., and Worsnop, D.: Changes in organic
  aerosol composition with aging inferred from aerosol mass spectra, Atmos Chem Phys, 11, 6465-6474,
  2011.
- 290 Reyes-Villegas, E., Green, D. C., Priestman, M., Canonaco, F., Coe, H., Prévôt, A. S. H., and Allan, J. D.:
- 291 Organic aerosol source apportionment in London 2013 with ME-2: exploring the solution space with
- annual and seasonal analysis, Atmos. Chem. Phys., 16, 15545-15559, 10.5194/acp-16-15545-2016, 2016.
- 294 Sandradewi, J., Prévôt, A. S. H., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E., and
- 295 Baltensperger, U. R. S.: Using aerosol light abosrption measurements for the quantitative
- 296 determination of wood burning and traffic emission contribution to particulate matter, Environmental
- 297 Science and Technology, 42, 3316-3323, 10.1021/es702253m, 2008.
- 298 Shinozuka, Y., Saide, P. E., Ferrada, G. A., Burton, S. P., Ferrare, R., Doherty, S. J., Gordon, H., Longo,
- K., Mallet, M., Feng, Y., Wang, Q., Cheng, Y., Dobracki, A., Freitag, S., Howell, S. G., LeBlanc, S., Flynn,
  C., Segal-Rosenhaimer, M., Pistone, K., Podolske, J. R., Stith, E. J., Bennett, J. R., Carmichael, G. R., da
- 301 Silva, A., Govindaraju, R., Leung, R., Zhang, Y., Pfister, L., Ryoo, J. M., Redemann, J., Wood, R., and
- 302 Zuidema, P.: Modeling the smoky troposphere of the southeast Atlantic: a comparison to ORACLES
- airborne observations from September of 2016, Atmos. Chem. Phys., 20, 11491-11526, 10.5194/acp-
- 304 20-11491-2020, 2020.
- 305 Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B., and Baltensperger, U.: Absorption of
- 306 light by soot particles: determination of the absorption coefficient by means of aethalometers, J
- 307 Aerosol Sci, 34, 1445-1463, 10.1016/S0021-8502(03)00359-8, 2003.