

Sources and geographical origins of fine aerosols in Paris (France)

M. Bressi^{1,2}, J. Sciare¹, V. Ghersi³, N. Mihalopoulos⁴, J.-E. Petit^{1,5}, J. B. Nicolas^{1,2}, S. Moukhtar³, A. Rosso³, A. Féron¹, N. Bonnaire¹, E. Poulakis⁴ and C. Theodosi⁴.

¹Laboratoire des Sciences du Climat et de l'Environnement, LSCE, UMR8212, CNRS-CEA-UVSQ, Gif-sur-Yvette, 91191, France.

²French Environment and Energy Management Agency, ADEME, 20 avenue du Grésillé, BP90406 49004, Angers Cedex 01, France.

³AIRPARIF, Surveillance de la Qualité de l'Air en Ile-de-France, Paris, 75004, France.

⁴Environmental Chemical Processes Laboratory, ECPL, Heraklion, Voutes, Greece.

⁵INERIS, DRC/CARA/CIME, Parc Technologique Alata, BP2, Verneuil-en-Halatte, 60550, France.

Correspondence to: michael.bressi@ensiacet.fr

Supplementary Material

S1. Complement to Sect. 1.2.2 "Data preparation"

The construction of the concentration dataset (X matrix) demands a precise and critical analysis of the chemical dataset. We initially used the whole chemical database described in Bressi et al. (2013) and Poulakis et al. (2012), in addition with the species mentioned in the Sect.1.1.2. This comprises PM_{2.5} mass and 29 chemical components: PM_{grav}, EC, OM, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺, Al, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Pb, levoglucosan, mannosan, arabitol and mannitol. Unfortunately, for specific elements, atmospheric concentrations were mostly below the Method Quantification Limit (MQL) and therefore difficult to interpret. After testing different threshold values, we decided to exclude every compound exhibiting less than 40% of their values above the MQL, which concerns Al, Cr, As, arabitol and mannitol. In addition, to avoid redundant species, Ca was excluded from the dataset but Ca²⁺ was taken into account, and levoglucosan and mannosan concentrations were subtracted from organic matter levels.

We also had to deal with missing species measurements due to analytical problems for specific samples. PMF3.0 does not allow missing data to be implemented in the X matrix. These data are generally replaced by "virtual" values having larger uncertainties in order to lower their influences in the PMF modelling (Huang et al., 1999; Polissar et al., 1998; Reff et al., 2007). Every

1 missing data was here replaced by the median of the corresponding species, and associated an
2 uncertainty of four times the species-specific median as suggested in Norris et al. (2008). The
3 detailed list of every processed missing data can be found in Table S2.

4

5 The second dataset, dealing with uncertainties associated with every species' measurement,
6 was constructed following the procedure described by Norris et al. (2008) and adapted from Polissar
7 et al. (1998). This is an equation-based method that requires each species Method Detection Limit
8 (MDL) -in the same unit as the species concentration (here in $\mu\text{g}/\text{m}^3$)- and its analytical uncertainty
9 (u) -in percentage-. The PMF uncertainty (σ) is calculated as:

$$10 \text{ if } x_{ij} \leq MDL_i, \forall j, \text{ then } \sigma_{ij} = \frac{5}{6} * MDL_i \quad (S1a)$$

$$11 \text{ if } x_{ij} > MDL_i, \forall j, \text{ then } \sigma_{ij} = \sqrt{(u_i * x_{ij})^2 + (MDL_i)^2} \quad (S1b)$$

12 MDLs and analytical uncertainties of our dataset were initially taken from Bressi et al. (2013),
13 Poulakis et al. (2012), and Gherzi et al. (2010), but lead to meaningless and non-robust results. An
14 adjustment was empirically made for every chemical species to have i) normally distributed scaled
15 residuals, ii) relevant MDL and u regarding the analytical method used and iii) mathematically
16 satisfactory and robust PMF results (see Sect. 2). Several empirical tests showed that when the
17 theoretical MDL of a given compound is not appropriate, taking its 5th percentile as its MDL often
18 leads to suitable results. In fact, this method lowers the statistical weight of the smallest
19 concentrations of a given chemical species. MDLs and analytical uncertainties chosen are reported in
20 Table S3.

21

22 With the PMF3.0 software, additional uncertainties can be optionally attributed to single
23 species and/or to the overall dataset. Single species additional uncertainties were suggested by
24 Paatero and Hopke (2003) with a calculation based on the signal-to-noise ratio (S/N), which is
25 defined in PMF3.0 as:

$$26 \left(\frac{S}{N}\right)_j = \sqrt{\frac{\sum_{i=1}^n (x_{ij} - \sigma_{ij})^2}{\sum_{i=1}^n \sigma_{ij}^2}} \quad (S2)$$

27 According to S/N values, species are categorised as "bad" ($S/N < 0.2$), "weak" ($0.2 \leq S/N \leq 2$) or "strong"
28 ($S/N > 2$), although the authors mention the relatively arbitrary nature of these figures. The bad
29 categorisation excludes the species from the dataset, the weak triples the PMF uncertainty whereas
30 the strong does not add supplemental uncertainties. According to these criteria, Cd, Zn and Cu were
31 defined weak whereas all other species were categorised strong. PM mass was defined as a "Total
32 Variable" i.e. was regarded as being the sum of all the PM chemical components, and was

1 automatically categorised weak in order to lower its influence in the final PMF results. An optional
2 "Extra Modeling Uncertainty" can be added in the PMF3.0 version but was not applied here.

3

4 **S2. Complement to Sect. 1.2.3 "Robustness of PMF results"**

5

6 Bootstrap is a statistical inference method introduced by Efron (1979) to "estimate the sampling
7 distribution of some prespecified random variable on the basis of the observed dataset". It gives
8 information on the accuracy of an estimate or a statistic. The key idea is to "resample from the
9 original data to create replicate datasets from which the variability of the quantities of interest will
10 be assessed" (Davison and Hinkley, 1997). Resampling can either be performed from an empirical
11 data distribution (nonparametric bootstrap) or via a fitted model describing a specific distribution
12 (parametric bootstrap). This led to different bootstrap versions reported in Wehrens et al. (2000).
13 One major assumption required for having reliable bootstrap results, is that data are independent
14 and identically distributed (Singh, 1981), which is questionable for real atmospheric samples. A way
15 to solve this dependence issue is to use the so-called "blocked bootstrap" method, which is randomly
16 selecting blocks of successive data, instead of selecting individual elements (see Lahiri, 2003 for more
17 details). The relevant block length to be chosen is discussed in Politis and White (2004). More
18 information concerning the bootstrap theory can be found in Efron and Tibshirani (1993).

19 We used the bootstrap method suggested in the PMF3.0 software which is a nonparametric
20 "blocked bootstrap" version. Non-overlapping blocks of consecutive samples are randomly selected
21 with replacement to create a new "bootstrap matrix" that has the same dimensions as the original X
22 matrix (bootstrap matrices will be noted with an "*" later on). PMF is then run on the X* matrix and
23 bootstrap factors are assigned to base run factors by comparing their contribution to PM mass (i.e.
24 by comparing G* and G columns, respectively). Bootstrap factors are "mapped" with base run factors
25 if their G* versus G column correlation is higher than a user defined threshold (see Sect. 1.2.4), and
26 "unmapped" otherwise. Further details concerning the bootstrap method used here can be found in
27 Norris et al. (2008).

1 Table S1. Days discarded from the PMF chemical dataset.

2

09/09	15, 16
10/09	7
11/09	7
12/09	16, 17, 18
01/10	7, 11, 12
02/10	19, 25, 28
03/10	1, 27, 28, 29
04/10	3, 4, 5, 6, 11, 12
05/10	-
06/10	-
07/10	16, 17, 18, 19
08/10	7
09/10	-
Total (days)	28

3

1 Table S2. List of the missing data replaced by the median of the corresponding species' concentrations.

2

	09/09	10/09	11/09	12/09	01/10	02/10	03/10	04/10	05/10	06/10	07/10	08/10	09/10	Total
PM	15	-	-	17	28	-	28	4, 5	-	9	16-19	7	-	12
OM	15, 16	7, 13, 14	7, 15	16-18	7, 11, 12	19, 25, 28	13, 27, 28	3-5, 11	17, 19, 20	9, 17, 18	16-19	7	-	34
EC	-	7	-	16-18	7	19, 25	14, 28	4, 5, 29	-	5, 9	17, 18	-	-	16
NO3	28	28	-	-	-	-	2, 28	4, 5	-	-	17, 18	-	-	8
SO4	17, 18	28	-	-	-	-	2, 28	4, 5	-	-	17, 18	-	-	9
NH4	28	-	-	-	-	-	28	4, 5	-	-	17, 18	-	-	6
Na	-	-	-	-	-	-	28	4, 5	-	-	17, 18	-	-	5
Cl	-	28	-	12	13	-	2, 28	4, 5	-	-	17, 18	-	-	9
Mg	-	-	11	-	1	-	28	4, 5	-	-	17, 18	-	-	7
K	-	-	8-10	-	1	-	28	4, 5	-	-	17, 18	-	-	9
Lev	-	13, 14	15	17, 18	-	-	13, 27, 28	4, 5	17	-	17, 18	-	-	13
Man	-	13, 14	15	17, 18	-	-	13, 27, 28	4, 5	17	-	17, 18	-	-	13
V	-	19, 20	-	20, 30	16	-	9, 27, 28	4, 5, 19	21	15	17, 18, 21	-	-	16
Ni	16, 25	3, 20	-	30	16	16	9, 27, 28	4, 5, 18, 28-30	1-20, 22, 27-31	1, 3, 4, 8-13, 15-16	2, 17, 18, 21, 25	23	-	59
Fe	-	-	20	30	-	4, 14	9, 27, 28	4, 5	-	15	17, 18, 21	-	-	13
Mn	-	-	-	27, 30	-	-	9, 28-31	4, 5	-	14, 15	14, 17, 18, 21, 24	17	-	17
Cu	-	-	20	17, 25, 30	1	14	9, 27, 28	4, 5, 27	-	15	17, 18, 21	-	-	16
Cd	23, 29	29	25	17, 30	25-27	-	9, 28-31	4, 5	13	15, 23-30	1-5, 17, 18, 21	-	-	34
Pb	-	-	-	17, 25, 30	26	-	9, 28-31	4, 5, 15	-	14, 15	17, 18, 21	-	-	17

3

1 Table S3. Method Detection Limits (MDL) and Analytical Uncertainties (u) chosen for PMF runs.

2

	MDL	u
	$\mu\text{g}\cdot\text{m}^{-3}$	%
PM	2.0E-01	5
OM	1.0E-01	15
EC	3.0E-01	15
NO3	1.3E-01	5
SO4	4.3E-01	5
NH4	3.0E-01	5
Na	2.6E-01	10
Cl	6.9E-02	10
Mg	5.5E-03	15
K	2.0E-02	10
Lev	5.2E-03	10
Man	5.1E-04	10
V	2.0E-04	10
Ni	2.9E-04	20
Fe	3.6E-02	20
Mn	5.6E-04	15
Cu	1.0E-03	20
Cd	2.1E-05	15
Pb	1.1E-03	15

3

1 Table S4. Using bootstrapping to determine the adequate number of factors in PMF.

2

3 *Legend: r-value: minimum coefficient of determination used to assign a boot factor to a base factor, rows: bootstrap factors, columns: base factors, Un:*
 4 *unmapped. The less robust bootstrap factor is boldfaced and coloured in red.*

5 Note: Bootstrapping was not performed on the same base run for the 0.6 and 0.7 r-value configurations. In fact, for each configuration, 20 base runs were
 6 conducted prior to bootstrapping and the base run showing the lowest Q-value was retained. Therefore, according to the r-value configuration, the different
 7 factor numbers will not correspond to the same physical source.
 8
 9

r-value=0.6

	1	2	3	4	5	6	Un
1	100	0	0	0	0	0	0
2	0	100	0	0	0	0	0
3	0	0	100	0	0	0	0
4	0	0	1	99	0	0	0
5	0	0	1	0	94	0	5
6	0	0	0	0	0	100	0

	1	2	3	4	5	6	7	Un
1	100	0	0	0	0	0	0	0
2	0	96	1	0	0	0	0	3
3	0	0	100	0	0	0	0	0
4	0	0	0	100	0	0	0	0
5	0	0	0	0	98	0	0	2
6	0	0	0	0	0	100	0	0
7	0	0	0	0	0	0	100	0

	1	2	3	4	5	6	7	8	Un
1	100	0	0	0	0	0	0	0	0
2	0	99	0	0	0	0	0	1	0
3	0	0	100	0	0	0	0	0	0
4	0	0	0	100	0	0	0	0	0
5	11	1	1	0	78	0	0	3	6
6	0	0	0	0	0	100	0	0	0
7	0	0	0	0	0	0	100	0	0
8	0	0	0	0	0	0	0	95	5

r-value=0.7

	1	2	3	4	5	6	Un
1	100	0	0	0	0	0	0
2	0	99	0	0	0	0	1
3	0	0	83	1	0	0	16
4	0	0	0	100	0	0	0
5	0	0	0	0	99	0	1
6	0	0	0	0	0	100	0

	1	2	3	4	5	6	7	Un
1	100	0	0	0	0	0	0	0
2	0	96	0	0	0	0	0	4
3	0	0	100	0	0	0	0	0
4	1	0	0	95	0	0	1	3
5	0	0	0	0	100	0	0	0
6	0	0	0	0	0	100	0	0
7	0	0	0	0	0	0	100	0

	1	2	3	4	5	6	7	8	Un
1	100	0	0	0	0	0	0	0	0
2	0	93	0	0	1	0	0	0	6
3	0	0	100	0	0	0	0	0	0
4	0	0	0	99	0	0	0	0	1
5	0	0	0	0	100	0	0	0	0
6	3	3	0	0	0	75	0	2	17
7	0	0	0	0	0	0	100	0	0
8	0	0	0	0	0	0	0	100	0

1 Table S5. Sum of the squared difference between the scaled residuals (d-values) calculated for each
 2 pair of base runs.

3 Note: Cf. Norris et al. (2008) and Sect. 2.1 for more information on this variable.

4

RUN#	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
2	0.028																		
3	0.019	0.003																	
4	0.020	0.005	0.005																
5	0.019	0.005	0.001	0.006															
6	0.007	0.020	0.011	0.012	0.010														
7	0.025	0.001	0.003	0.004	0.005	0.017													
8	0.013	0.019	0.012	0.010	0.009	0.003	0.018												
9	0.032	0.007	0.009	0.004	0.010	0.020	0.007	0.017											
10	0.025	0.012	0.010	0.007	0.008	0.011	0.012	0.006	0.009										
11	0.005	0.028	0.016	0.022	0.014	0.005	0.026	0.010	0.032	0.023									
12	0.019	0.008	0.005	0.004	0.004	0.008	0.007	0.005	0.008	0.002	0.017								
13	0.007	0.022	0.013	0.014	0.011	0.001	0.020	0.003	0.022	0.012	0.006	0.009							
14	0.041	0.088	0.071	0.087	0.069	0.063	0.085	0.075	0.106	0.099	0.037	0.084	0.062						
15	0.008	0.022	0.013	0.013	0.011	0.001	0.019	0.003	0.021	0.011	0.006	0.008	0.001	0.064					
16	0.017	0.005	0.002	0.004	0.001	0.007	0.005	0.007	0.009	0.006	0.013	0.002	0.009	0.073	0.008				
17	0.048	0.106	0.094	0.101	0.096	0.082	0.102	0.098	0.122	0.122	0.057	0.107	0.081	0.016	0.084	0.097			
18	0.018	0.016	0.011	0.009	0.009	0.006	0.015	0.003	0.012	0.002	0.016	0.003	0.007	0.087	0.006	0.006	0.109		
19	0.035	0.005	0.010	0.004	0.012	0.025	0.006	0.023	0.002	0.013	0.038	0.011	0.028	0.110	0.027	0.011	0.121	0.017	
20	0.031	0.006	0.009	0.003	0.010	0.019	0.006	0.015	0.003	0.006	0.033	0.006	0.021	0.108	0.020	0.008	0.122	0.010	0.003

5
6

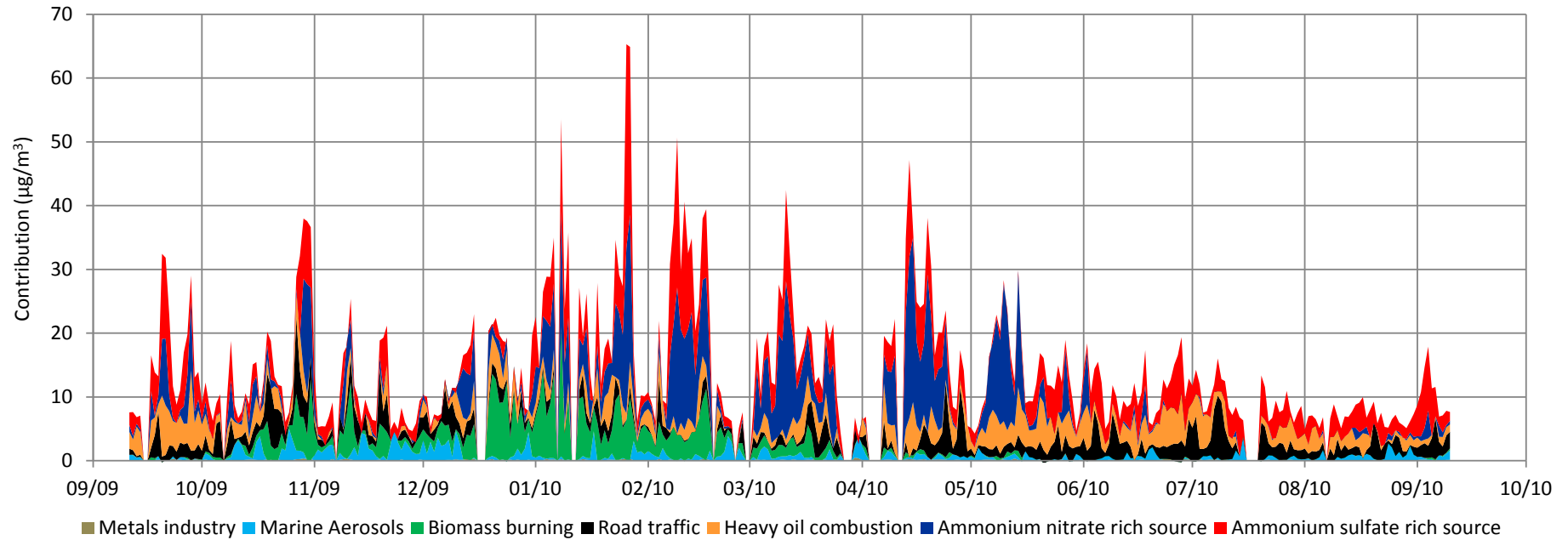
1 Table S6. Description of the different receptor model studies compared in Fig. 8 and discussed in
 2 Sect. 3.3.1.

3 Note: Sites are indicated as: "City (Country)-Type of sites". Urb: urban, Rur: rural, Kerb: kerbside.
 4

	Authors	Site	Receptor model	Year	PM fraction	PM mass ($\mu\text{g}/\text{m}^3$)	Source contribution ($\mu\text{g}/\text{m}^3$)	Source contribution (%)
Ammonium nitrate + ammonium sulfate rich sources	Vallius et al., 2005	Amsterdam (Neth) - Urb	PCA	1998-1999	PM _{2.5}	20.0	6.8	34
	Andersen et al., 2007	Copenhagen (Den) - Urb	COPREM	1999-2004	PM ₁₀	23.3	6.9	29
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	7.4	51
	Mooibroek et al., 2011	Schiedam (Neth) - Urb	EPA PMF3.0	2007-2008	PM _{2.5}	13	8.6	66
	Mooibroek et al., 2011	Hellendoorn (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	12.5	9.1	73
	Mooibroek et al., 2011	Rotterdam (Neth) - Kerb	EPA PMF3.0	2007-2008	PM _{2.5}	16.4	10.0	61
	Mooibroek et al., 2011	Vredepeel (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	14.5	10.7	74
	Mooibroek et al., 2011	Cabauw (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	17.5	12.6	72
Quass et al., 2004	Duisburg (Ger) - Urb	PMF	2003-2004	PM _{2.5}	22.8	13	57	
Ammonium nitrate rich source	Andersen et al., 2007	Copenhagen (Den) - Urb	COPREM	1999-2004	PM ₁₀	23.3	3.3	14
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	3.5	24
	Mooibroek et al., 2011	Schiedam (Neth) - Urb	EPA PMF3.0	2007-2008	PM _{2.5}	13.0	5.6	43
	Mooibroek et al., 2011	Hellendoorn (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	12.5	6.0	48
	Mooibroek et al., 2011	Vredepeel (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	14.5	6.4	44
	Mooibroek et al., 2011	Rotterdam (Neth) - Kerb	EPA PMF3.0	2007-2008	PM _{2.5}	16.4	6.7	41
	Mooibroek et al., 2011	Cabauw (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	17.5	7.7	44
Ammonium sulfate rich source	Mooibroek et al., 2011	Schiedam (Neth) - Urb	EPA PMF3.0	2007-2008	PM _{2.5}	13.0	3.0	23
	Mooibroek et al., 2011	Hellendoorn (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	12.5	3.1	25
	Mooibroek et al., 2011	Rotterdam (Neth) - Kerb	EPA PMF3.0	2007-2008	PM _{2.5}	16.4	3.3	20
	Andersen et al., 2007	Copenhagen (Den) - Urb	COPREM	1999-2004	PM ₁₀	23.3	3.5	15
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	3.9	27
	Mooibroek et al., 2011	Vredepeel (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	14.5	4.4	30
	Mooibroek et al., 2011	Cabauw (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	17.5	4.9	28
Road traffic	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	2.1	14
	Lee et al., 2003	Toronto (Ca) - Urb	PMF	2000-2001	PM _{2.5}	12.8	2.3	18
	Maykut et al., 2003	Seattle (USA) - Urb	PMF and UNMIX	1996-1999	PM _{2.5}	9.2	2.3	25
	Minguillón et al., 2012	Zurich (Swi) - Urb	EPA PMF3.0	Summer and Winter 2009	PM _{1.0}	10.2	3.8	37
	Perronne et al., 2012	Milan (It) - Urb	CMB	2006-2009	PM _{2.5}	35.5	7.8	22
Biomass combustion	Karanasiou et al., 2009	Athens (Gr) - Urb	PMF3	2002	PM _{2.0}	5.3	0.8	15
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	1.8	12
	Perronne et al., 2012	Milan (It) - Urb	CMB	2006-2009	PM _{2.5}	35.5	7.1	16
Andersen et al., 2007	Copenhagen (Den) - Urb	COPREM	1999-2004	PM ₁₀	23.3	7.3	15	
Heavy oil combustion	Andersen et al., 2007	Copenhagen (Den) - Urb	COPREM	1999-2004	PM ₁₀	23.3	2.2	9
	Alleman et al., 2010	Dunkirk (Fr) - Urb	PMF2	2003-2005	PM ₁₀	25.0	2.3	9
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	2.4	16
Marine aerosols	Vallius et al., 2005	Amsterdam (Neth) - Urb	PCA	1998-1999	PM _{2.5}	20.0	3.5	9
	Mooibroek et al., 2011	Hellendoorn (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	12.5	0.8	6
	This study	Paris (Fr) - Urb	EPA PMF3.0	2009-2010	PM _{2.5}	14.7	0.8	6
	Mooibroek et al., 2011	Rotterdam (Neth) - Kerb	EPA PMF3.0	2007-2008	PM _{2.5}	16.4	0.8	5
	Mooibroek et al., 2011	Vredepeel (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	14.5	0.9	6
	Vallius et al., 2005	Helsinki (Fin) - Urb	PCA	1998-1999	PM _{2.5}	12.2	0.9	7
	Karanasiou et al., 2009	Athens (Gr) - Urb	PMF3	2002	PM _{2.0}	5.3	1.1	21
	Mooibroek et al., 2011	Schiedam (Neth) - Urb	EPA PMF3.0	2007-2008	PM _{2.5}	13.0	1.2	9
	Mooibroek et al., 2011	Cabauw (Neth) - Rur	EPA PMF3.0	2007-2008	PM _{2.5}	17.5	1.6	9

1

2

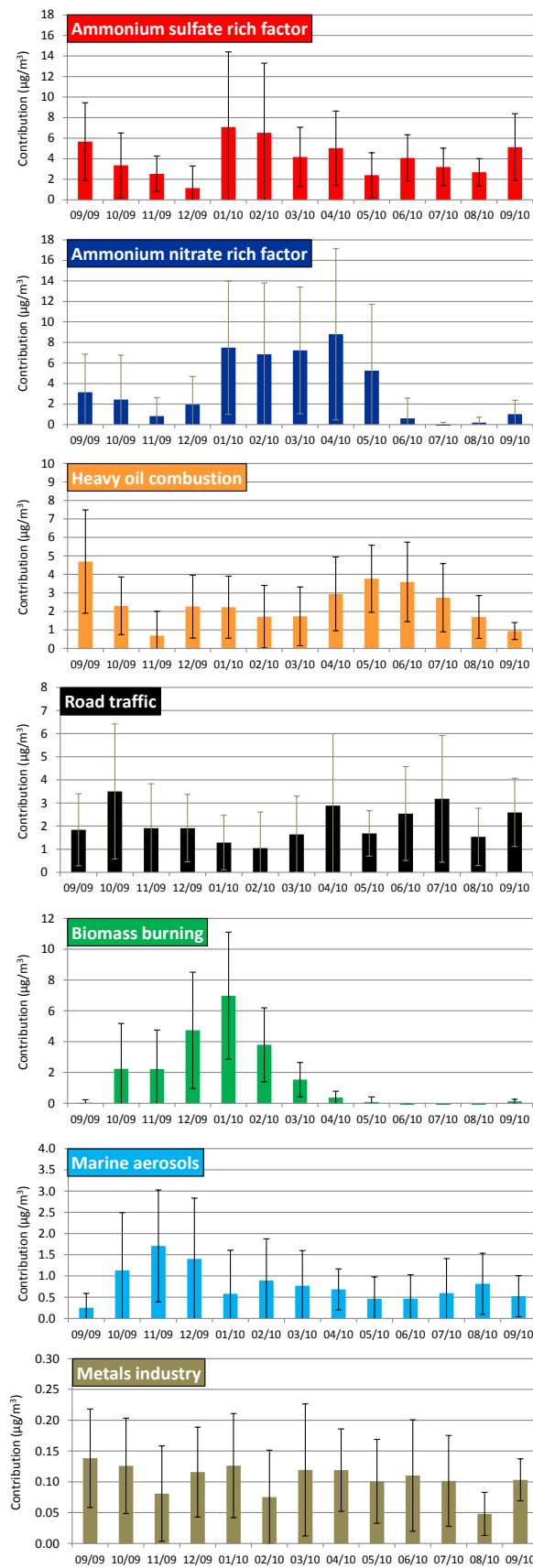


3

4

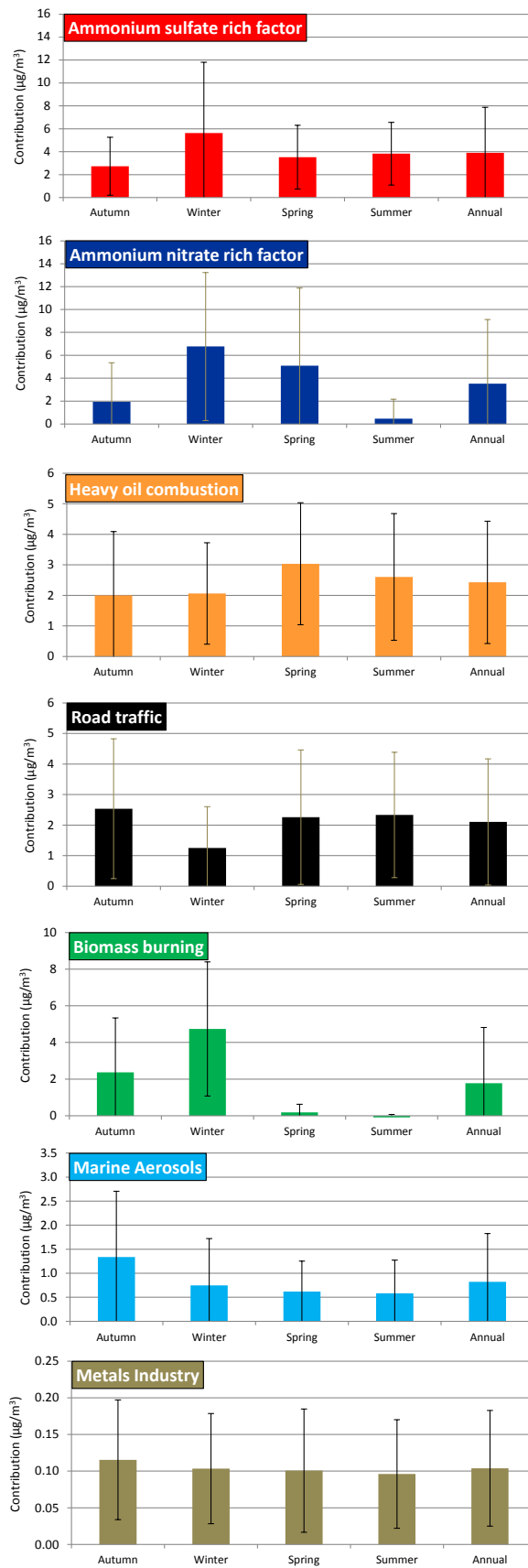
5 Fig. S1. Daily contribution ($\mu\text{g}/\text{m}^3$) of the seven sources to $\text{PM}_{2.5}$ mass from 11 September 2009 to 10 September 2010.

6 Note: results were taken from the base run exhibiting the lowest Q_{robust} .



1
2
3
4
5

Fig. S2. Monthly mean contribution ($\mu\text{g}/\text{m}^3$) of $\text{PM}_{2.5}$ sources from 11 September 2009 to 10 September 2010. Constructed from the base run exhibiting the lowest Q_{robust} . Error bars represent $\pm 1\sigma$.



1
2
3
4
5

Fig. S3. Seasonal mean contribution ($\mu\text{g}/\text{m}^3$) of PM_{2.5} sources from 11 September 2009 to 10 September 2010. Constructed from the base run exhibiting the lowest Q_{robust} . Error bars represent $\pm 1\sigma$.

1 **References**

2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31

Alleman, L. Y., Lamaison, L., Perdrix, E., Robache, A. and Galloo, J.-C.: PM10 metal concentrations and source identification using positive matrix factorization and wind sectoring in a French industrial zone, *Atmospheric Research*, 96(4), 612–625, doi:10.1016/j.atmosres.2010.02.008, 2010.

Andersen, Z. J., Wahlin, P., Raaschou-Nielsen, O., Scheike, T. and Loft, S.: Ambient particle source apportionment and daily hospital admissions among children and elderly in Copenhagen, *Journal of Exposure Science and Environmental Epidemiology*, 17(7), 625–636, doi:10.1038/sj.jes.7500546, 2007.

Bressi, M., Sciare, J., Ghersi, V., Bonnaire, N., Nicolas, J. B., Petit, J.-E., Moukhtar, S., Rosso, A., Mihalopoulos, N. and Féron, A.: A one-year comprehensive chemical characterisation of fine aerosols (PM2.5) at urban, suburban and rural background sites in the region of Paris (France), *Atmos. Chem. Phys.*, 13, 7825–7844, doi:10.5194/acp-13-7825-2013, 2013.

Davison, A. C. and Hinkley, D. V.: *Bootstrap Methods and Their Application*, Cambridge University Press, 1997.

Efron, B.: 1977 Rietz Lecture - Bootstrap Methods - Another Look at the Jackknife, *Ann. Stat.*, 7(1), 1–26, doi:10.1214/aos/1176344552, 1979.

Efron, B. and Tibshirani, R.: *An Introduction to the Bootstrap*, Chapman & Hall, 1993.

Ghersi, V., Rosso, A., Moukhtar, S., Lameloise, P., Sciare, J., Bressi, M., Nicolas, J. B., Féron, A. and Bonnaire, N.: Etude de contribution des sources de particules fines (PM2,5) en Ile-de-France, *Pollution Atmosphérique*, 63–72, 2010.

Huang, S., Rahn, K. A. and Arimoto, R.: Testing and optimizing two factor-analysis techniques on aerosol at Narragansett, Rhode Island, *Atmospheric Environment*, 33(14), 2169–2185, doi:10.1016/S1352-2310(98)00324-0, 1999.

Karanasiou, A. A., Siskos, P. A. and Eleftheriadis, K.: Assessment of source apportionment by Positive Matrix Factorization analysis on fine and coarse urban aerosol size fractions, *Atmospheric Environment*, 43(21), 3385–3395, doi:10.1016/j.atmosenv.2009.03.051, 2009.

Lahiri, S. N.: *Resampling Methods for Dependent Data*, Springer, 2003.

Lee, P. K. H., Brook, J. R., Dabek-Zlotorzynska, E. and Mabury, S. A.: Identification of the Major Sources Contributing to PM2.5 Observed in Toronto, *Environ. Sci. Technol.*, 37(21), 4831–4840, doi:10.1021/es026473i, 2003.

1 Maykut, N. N., Lewtas, J., Kim, E. and Larson, T. V.: Source Apportionment of PM_{2.5} at an Urban
2 IMPROVE Site in Seattle, Washington, *Environ. Sci. Technol.*, 37(22), 5135–5142,
3 doi:10.1021/es030370y, 2003.

4 Minguillón, M. C., Querol, X., Baltensperger, U. and Prévôt, A. S. H.: Fine and coarse PM composition and
5 sources in rural and urban sites in Switzerland: Local or regional pollution?, *Science of The Total
6 Environment*, 427–428(0), 191–202, doi:10.1016/j.scitotenv.2012.04.030, 2012.

7 Mooibroek, D., Schaap, M., Weijers, E. P. and Hoogerbrugge, R.: Source apportionment and spatial
8 variability of PM_{2.5} using measurements at five sites in the Netherlands, *Atmospheric
9 Environment*, 45(25), 4180–4191, doi:10.1016/j.atmosenv.2011.05.017, 2011.

10 Norris, G., Vedantham, R., Wade, K., Brown, S., Prouty, J. and Foley, C.: EPA Positive Matrix Factorization
11 (PMF) 3.0: fundamentals & user guide, U.S. Environmental Protection Agency, 2008.

12 Paatero, P. and Hopke, P. K.: Discarding or downweighting high-noise variables in factor analytic models,
13 *Analytica Chimica Acta*, 490(1-2), 277–289, 2003.

14 Perrone, M. G., Larsen, B. R., Ferrero, L., Sangiorgi, G., De Gennaro, G., Udisti, R., Zangrando, R.,
15 Gambaro, A. and Bolzacchini, E.: Sources of high PM_{2.5} concentrations in Milan, Northern Italy:
16 Molecular marker data and CMB modelling, *Science of The Total Environment*, 414(0), 343–355,
17 doi:10.1016/j.scitotenv.2011.11.026, 2012.

18 Polissar, A., Hopke, P. and Paatero, P.: Atmospheric aerosol over Alaska - 2. Elemental composition and
19 sources, *J. Geophys. Res.-Atmos.*, 103(D15), 19045–19057, doi:10.1029/98JD01212, 1998.

20 Politis, D. N. and White, H.: Automatic Block-Length Selection for the Dependent Bootstrap, *Econometric
21 Reviews*, 23(1), 53–70, doi:10.1081/ETC-120028836, 2004.

22 Poulakis, E., Theodosi, C., Sciare, J., Bressi, M., Ghersi, V. and Mihalopoulos, N.: Airborne mineral
23 components and trace metals in Paris region: Spatial and temporal variability, manuscript in
24 preparation, 2012.

25 Quass, U., Kuhlbusch, T. and Koch, M.: Identification of source groups of fine dust, Public report to the
26 Environment Ministry of North Rhine Westphalia, Germany. Available from:
27 http://www.lanuv.nrw.de/luft/berichte/FeinstaubNRW_2004_Summary.pdf (Accessed 24 July
28 2012), 2004.

29 Reff, A., Eberly, S. I. and Bhave, P. V.: Receptor modeling of ambient particulate matter data using
30 positive matrix factorization: review of existing methods, *Journal of the Air & Waste Management
31 Association*, 57(2), 146, 2007.

1 Singh, K.: On the Asymptotic Accuracy of Efron's Bootstrap, *Ann. Statist.*, 9(6), 1187–1195,
2 doi:10.1214/aos/1176345636, 1981.

3 Vallius, M., Janssen, N. A. H., Heinrich, J., Hoek, G., Ruuskanen, J., Cyrus, J., Van Grieken, R., de Hartog, J.
4 J., Kreyling, W. G. and Pekkanen, J.: Sources and elemental composition of ambient PM2.5 in
5 three European cities, *Science of The Total Environment*, 337(1–3), 147–162,
6 doi:10.1016/j.scitotenv.2004.06.018, 2005.

7 Wehrens, R., Putter, H. and Buydens, L.: The bootstrap: a tutorial, *Chemometrics and Intelligent*
8 *Laboratory Systems*, 54(1), 35–52, 2000.

9