Supplement of Atmos. Chem. Phys. Discuss., 15, 14733–14781, 2015
http://www.atmos-chem-phys-discuss.net/15/14733/2015/
doi:10.5194/acpd-15-14733-2015-supplement

Atmospheric Chemistry and Physics
Discussions
Open Access

Supplement of

Advanced source apportionment of size-resolved trace elements at multiple sites in London during winter

S. Visser et al.

Correspondence to: J. G. Slowik (jay.slowik@psi.ch)

Supplement

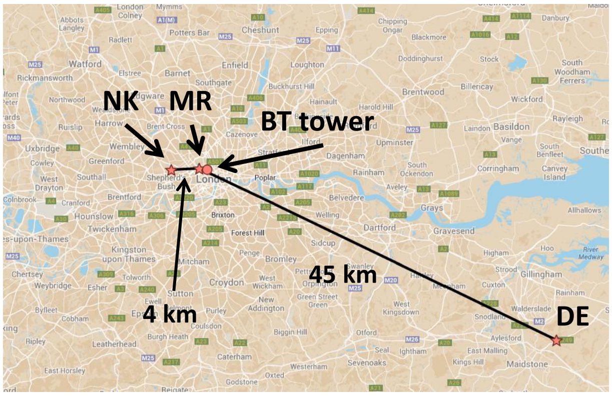

Figure S1. Map of southeastern UK. Indicated are the sampling sites Marylebone Road (MR, kerbside), North Kensington (NK, urban background), Detling (DE, rural), and the elevated BT Tower site for meteorological measurements (adapted from Google Maps).

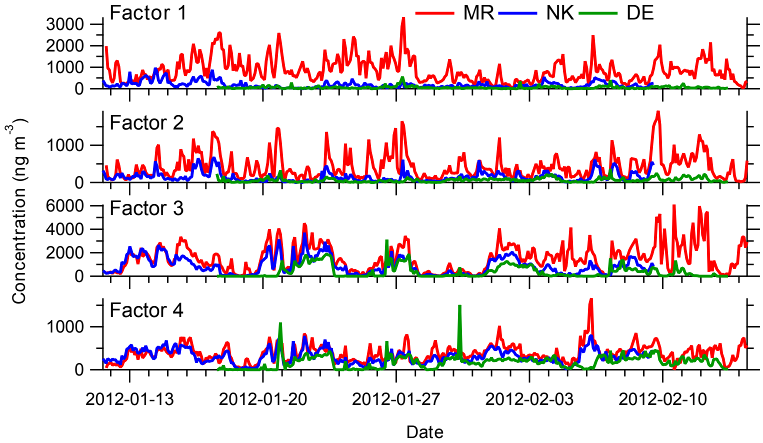

Figure S2. Non-optimal $PM_{10-2.5}$ source contributions (factor time series) with unconstrained ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Factor 1 indicates mixed traffic-related and brake wear; factor 2 resuspended dust; factor 3 sea / road salt; factor 4 aged sea salt. See Fig. S5 for accompanying source profiles.

The residuals of Ni, Cr and Mo remain large at DE. Unconstrained ME-2 on five or six factors leads to unstable results varying strongly with seed. The dust factor splits in factors rich in Al and Si, and in Ca, but without improving residuals. A brake wear factor or a factor with Ni, Cr and Mo does not appear with increasing number of factors.

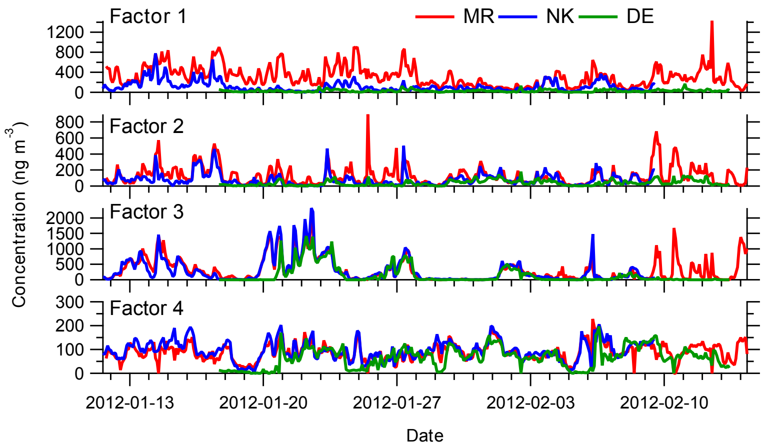

Figure S3. Non-optimal $PM_{2.5-1.0}$ source contributions (factor time series) with unconstrained ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Factor 1 indicates mixed traffic-related and brake wear; factor 2 resuspended dust; factor 3 sea / road salt; factor 4 mixed aged sea salt and regional transport. See Fig. S5 for accompanying source profiles.

Unconstrained ME-2 on five or six factors leads to unstable results varying strongly with seed. The dust factor splits in factors rich in Al, and in Si and Ca, but without improving residuals. A brake wear factor does not ap-pear with increasing number of factors. The factor containing mixed aged sea salt and regional transport cannot be unmixed in unconstrained ME-2.

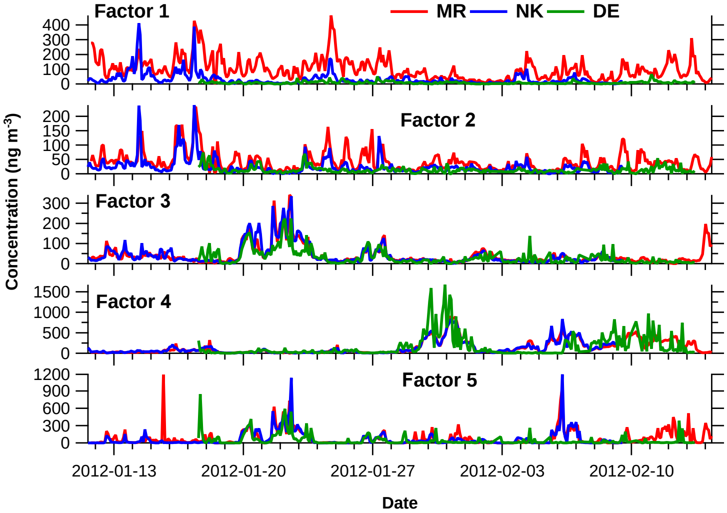

Figure S4. Non-optimal $PM_{1.0-0.3}$ source contributions (factor time series) with unconstrained ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Factor 1 indicates traffic-related; factor 2 resuspended dust; factor 3 aged sea salt; factor 4 mixed S-rich and solid fuel; factor 5 mixed sea / road salt and Cl-rich. See Fig. S5 for accompanying source profiles.

Unconstrained ME-2 on six or seven factors leads to unstable results varying strongly with seed. The S-rich and solid fuel factor splits in a factor with only S as indicative for S-rich, but the second factor contains K without S. In a solid fuel source S can be expected. The mixed sea / road salt and Cl rich source (factor 5) is visible from the time series from roughly 20-24 January. This episode correlates strongly with factor 3 and with western wind, indicative of sea salt. Contrary, the episode from 5-7 February is absent in factor 3 and at the rural site, indicative of a source with fine Cl.

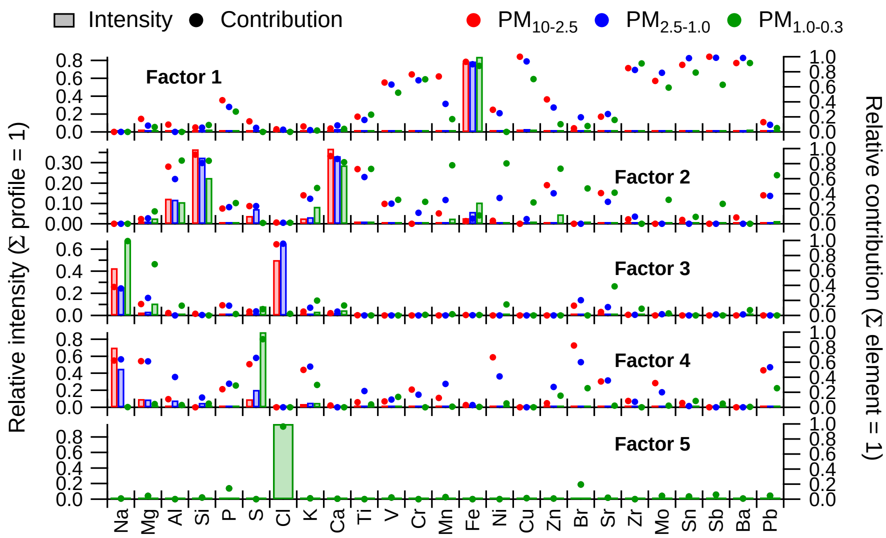

Figure S5. Non-optimal source profiles of unconstrained ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). The bars (left y-axis) represent the average element intensity to each factor in ng ng^{-1}, the circles (right y-axis) the fraction of the total predicted concentration for a given element. See Figs. S2-4 for an indication of the sources and why these profiles are considered non-optimal.

Table S1. Source profiles of ME-2 results on combined data of the MR-NK-DE sites for $\mathrm{PM}_{10-2.5}$ with mean $\pm$ 1 standard deviation (std) from the anchor sensitivity analysis. Relative intensity in ng ng^{-1} represents the average element contribution to the factor ($\sum$ profile = 1). Relative contribution denotes the fraction of the total predicted concentration for a given element ($\sum$ contribution = 1). See also Fig. 2.

Relative intensity												
Element	Brake wear		Traffic		Dust		Sea / road salt		Aged sea salt		Industrial	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.000	0.000	0.434	0.013	0.668	0.009	0.000	0.000
Mg	0.030	0.003	0.020	0.003	0.018	0.002	0.024	0.002	0.089	0.004	0.000	0.000
Al	0.034	0.004	0.011	0.002	0.095	0.007	0.004	0.000	0.025	0.002	0.079	0.008
Si	0.000	0.000	0.014	0.002	0.292	0.015	0.002	0.000	0.028	0.002	0.069	0.007
P	0.016	0.002	0.006	0.001	0.004	0.000	0.002	0.000	0.003	0.000	0.007	0.001
S	0.000	0.000	0.019	0.003	0.039	0.004	0.008	0.001	0.088	0.006	0.003	0.000
Cl	0.000	0.000	0.009	0.001	0.068	0.006	0.512	0.020	0.000	0.000	0.000	0.000
K	0.000	0.000	0.003	0.000	0.023	0.002	0.003	0.000	0.035	0.002	0.007	0.001
Ca	0.000	0.000	0.005	0.001	0.290	0.020	0.002	0.000	0.052	0.004	0.000	0.000
Ti	0.011	0.001	0.002	0.000	0.008	0.001	0.000	0.000	0.001	0.000	0.000	0.000
V	0.014	0.002	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Cr	0.019	0.002	0.004	0.001	0.001	0.000	0.000	0.000	0.000	0.000	0.381	0.029
Mn	0.000	0.000	0.012	0.002	0.003	0.000	0.000	0.000	0.001	0.000	0.030	0.004
Fe	0.000	0.000	0.890	0.016	0.143	0.012	0.008	0.001	0.000	0.000	0.000	0.000
Ni	0.007	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.318	0.027
Cu	0.360	0.028	0.002	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.003	0.000
Zn	0.090	0.010	0.000	0.000	0.007	0.001	0.000	0.000	0.001	0.000	0.027	0.003
Br	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.003	0.000	0.007	0.001
Sr	0.007	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.002	0.000
Zr	0.017	0.002	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mo	0.033	0.004	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.053	0.006
Sn	0.048	0.006	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.007	0.001
Sb	0.051	0.006	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.004	0.000
Ba	0.264	0.024	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Pb	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.002	0.000	0.001	0.000

Relative contribution												
Element	Brake wear		Traffic		Dust		Sea / road salt		Aged sea salt		Industrial	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.000	0.000	0.394	0.007	0.606	0.007	0.000	0.000
Mg	0.164	0.013	0.112	0.011	0.101	0.007	0.130	0.009	0.493	0.014	0.000	0.000
Al	0.137	0.011	0.043	0.005	0.383	0.017	0.015	0.001	0.101	0.006	0.321	0.019
Si	0.000	0.000	0.035	0.004	0.721	0.015	0.004	0.000	0.070	0.005	0.170	0.014
P	0.431	0.025	0.153	0.017	0.097	0.008	0.058	0.005	0.083	0.005	0.179	0.016
S	0.000	0.000	0.117	0.017	0.250	0.022	0.052	0.006	0.560	0.026	0.021	0.003
Cl	0.000	0.000	0.015	0.003	0.116	0.012	0.869	0.013	0.000	0.000	0.000	0.000
K	0.000	0.000	0.042	0.008	0.320	0.030	0.046	0.006	0.495	0.032	0.097	0.014
Ca	0.000	0.000	0.014	0.003	0.831	0.015	0.005	0.001	0.149	0.014	0.000	0.000
Ti	0.515	0.039	0.080	0.014	0.344	0.034	0.000	0.000	0.062	0.006	0.000	0.000
V	0.818	0.021	0.067	0.012	0.056	0.007	0.000	0.000	0.017	0.002	0.042	0.007
Cr	0.046	0.007	0.010	0.002	0.002	0.000	0.000	0.000	0.001	0.000	0.941	0.008
Mn	0.002	0.000	0.255	0.038	0.070	0.009	0.000	0.000	0.015	0.002	0.658	0.040
Fe	0.000	0.000	0.855	0.012	0.137	0.012	0.007	0.001	0.000	0.000	0.000	0.000
Ni	0.022	0.004	0.001	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.975	0.004
Cu	0.975	0.002	0.006	0.001	0.009	0.001	0.001	0.000	0.000	0.000	0.009	0.001
Zn	0.717	0.030	0.003	0.000	0.058	0.007	0.000	0.000	0.008	0.001	0.214	0.028
Br	0.000	0.000	0.017	0.004	0.000	0.000	0.053	0.007	0.285	0.029	0.645	0.033
Sr	0.664	0.033	0.000	0.000	0.053	0.007	0.011	0.001	0.063	0.006	0.210	0.028
Zr	0.902	0.012	0.047	0.009	0.029	0.004	0.001	0.000	0.021	0.002	0.000	0.000
Mo	0.377	0.041	0.004	0.001	0.006	0.001	0.000	0.000	0.008	0.001	0.605	0.042
Sn	0.838	0.022	0.009	0.002	0.013	0.002	0.000	0.000	0.008	0.001	0.131	0.020
Sb	0.926	0.012	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.071	0.012
Ba	0.991	0.001	0.000	0.000	0.007	0.001	0.001	0.000	0.000	0.000	0.000	0.000
Pb	0.000	0.000	0.106	0.020	0.238	0.027	0.000	0.000	0.356	0.030	0.300	0.036

Table S2. Source profiles of ME-2 results on combined data of the MR-NK-DE sites for $\mathrm{PM}_{2.5-1.0}$ with mean $\pm$ 1 standard deviation (std) from the anchor sensitivity analysis. Relative intensity in ng ng^{-1} represents the average element contribution to the factor ($\sum$ profile = 1). Relative contribution denotes the fraction of the total predicted concentration for a given element ($\sum$ contribution = 1). See also Fig. 2.

Relative intensity												
Element	Brake wear		Traffic		Dust		Sea / road salt		Aged sea salt		S-rich	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.000	0.000	0.225	0.010	0.640	0.005	0.000	0.000
Mg	0.021	0.002	0.012	0.002	0.007	0.001	0.033	0.002	0.100	0.003	0.050	0.004
Al	0.034	0.004	0.011	0.001	0.089	0.006	0.000	0.000	0.067	0.003	0.005	0.000
Si	0.000	0.000	0.038	0.005	0.284	0.013	0.005	0.000	0.028	0.001	0.038	0.003
P	0.017	0.002	0.006	0.001	0.005	0.000	0.003	0.000	0.005	0.000	0.009	0.001
S	0.000	0.000	0.001	0.000	0.005	0.000	0.027	0.002	0.085	0.004	0.576	0.020
Cl	0.000	0.000	0.020	0.003	0.000	0.000	0.672	0.025	0.000	0.000	0.036	0.003
K	0.000	0.000	0.003	0.000	0.020	0.002	0.010	0.001	0.046	0.002	0.058	0.005
Ca	0.000	0.000	0.040	0.005	0.300	0.020	0.020	0.001	0.000	0.000	0.014	0.001
Ti	0.012	0.001	0.003	0.000	0.009	0.001	0.000	0.000	0.003	0.000	0.004	0.000
V	0.014	0.002	0.002	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Cr	0.019	0.002	0.004	0.001	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mn	0.000	0.000	0.010	0.001	0.007	0.001	0.000	0.000	0.005	0.000	0.000	0.000
Fe	0.000	0.000	0.820	0.023	0.252	0.020	0.003	0.000	0.014	0.001	0.170	0.015
Ni	0.007	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Cu	0.332	0.027	0.011	0.002	0.005	0.000	0.000	0.000	0.000	0.000	0.007	0.001
Zn	0.092	0.010	0.003	0.000	0.007	0.001	0.000	0.000	0.002	0.000	0.014	0.001
Br	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.001	0.000	0.005	0.001
Sr	0.009	0.001	0.000	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.001	0.000
Zr	0.018	0.002	0.002	0.000	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000
Mo	0.034	0.004	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Sn	0.049	0.005	0.003	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Sb	0.052	0.006	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ba	0.290	0.025	0.008	0.001	0.003	0.000	0.001	0.000	0.000	0.000	0.003	0.000
Pb	0.000	0.000	0.001	0.000	0.002	0.000	0.000	0.000	0.001	0.000	0.005	0.000
Relative contribution												
Element	Brake wear		Traffic		Dust		Sea / road salt		Aged sea salt		S-rich	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.000	0.000	0.260	0.006	0.740	0.006	0.000	0.000
Mg	0.096	0.007	0.053	0.005	0.032	0.002	0.148	0.007	0.447	0.009	0.225	0.011
Al	0.164	0.012	0.052	0.005	0.435	0.014	0.000	0.000	0.326	0.011	0.022	0.002
Si	0.000	0.000	0.097	0.009	0.722	0.011	0.013	0.001	0.072	0.003	0.096	0.007
P	0.384	0.022	0.140	0.014	0.102	0.008	0.063	0.004	0.108	0.005	0.203	0.015
S	0.000	0.000	0.002	0.000	0.007	0.001	0.039	0.003	0.123	0.006	0.829	0.007
Cl	0.000	0.000	0.027	0.005	0.000	0.000	0.923	0.008	0.000	0.000	0.050	0.006
K	0.000	0.000	0.020	0.004	0.147	0.019	0.075	0.009	0.332	0.022	0.426	0.031
Ca	0.000	0.000	0.106	0.016	0.803	0.019	0.053	0.005	0.000	0.000	0.037	0.005
Ti	0.385	0.037	0.087	0.016	0.308	0.031	0.000	0.000	0.086	0.006	0.133	0.017
V	0.769	0.025	0.086	0.015	0.061	0.008	0.001	0.000	0.004	0.000	0.079	0.010
Cr	0.748	0.030	0.159	0.026	0.061	0.008	0.000	0.000	0.019	0.001	0.014	0.002
Mn	0.004	0.001	0.450	0.043	0.306	0.038	0.000	0.000	0.239	0.019	0.000	0.000
Fe	0.000	0.000	0.651	0.017	0.200	0.016	0.002	0.000	0.011	0.001	0.135	0.013
Ni	0.808	0.021	0.041	0.008	0.042	0.005	0.000	0.000	0.000	0.000	0.109	0.014
Cu	0.933	0.007	0.032	0.005	0.015	0.002	0.001	0.000	0.000	0.000	0.020	0.002
Zn	0.781	0.021	0.022	0.004	0.058	0.008	0.000	0.000	0.017	0.001	0.122	0.015
Br	0.000	0.000	0.084	0.020	0.000	0.000	0.122	0.016	0.155	0.014	0.638	0.034
Sr	0.767	0.020	0.012	0.002	0.051	0.007	0.026	0.003	0.064	0.005	0.080	0.011
Zr	0.825	0.020	0.098	0.017	0.040	0.005	0.000	0.000	0.038	0.003	0.000	0.000
Mo	0.895	0.013	0.053	0.010	0.011	0.002	0.004	0.000	0.011	0.001	0.026	0.004
Sn	0.922	0.011	0.052	0.010	0.010	0.001	0.002	0.000	0.000	0.000	0.014	0.002
Sb	0.968	0.005	0.024	0.005	0.005	0.001	0.003	0.000	0.000	0.000	0.000	0.000
Ba	0.955	0.005	0.025	0.004	0.008	0.001	0.003	0.000	0.000	0.000	0.009	0.001
Pb	0.000	0.000	0.083	0.018	0.191	0.027	0.000	0.000	0.169	0.015	0.557	0.039

Table S3. Source profiles of ME-2 results on combined data of the MR-NK-DE sites for $\mathrm{PM}_{1.0-0.3}$ with mean $\pm$ 1 standard deviation (std) from the anchor sensitivity analysis. Relative intensity in ng ng^{-1} represents the average element contribution to the factor ($\sum$ profile = 1). Relative contribution denotes the fraction of the total predicted concentration for a given element ($\sum$ contribution = 1). See also Fig. 2.

Relative intensity														
Element	Traffic		Dust		Sea / road salt		Aged sea salt		S-rich		Solid fuel		Reacted Cl	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.223	0.005	0.705	0.004	0.000	0.000	0.000	0.000	0.000	0.000
Mg	0.011	0.002	0.013	0.001	0.033	0.002	0.147	0.005	0.004	0.000	0.013	0.001	0.010	0.001
Al	0.004	0.001	0.084	0.007	0.001	0.000	0.055	0.002	0.005	0.000	0.000	0.000	0.002	0.000
Si	0.020	0.003	0.240	0.014	0.003	0.000	0.005	0.000	0.013	0.001	0.035	0.003	0.004	0.000
P	0.008	0.001	0.006	0.000	0.003	0.000	0.000	0.000	0.008	0.001	0.013	0.001	0.004	0.000
S	0.000	0.000	0.001	0.000	0.043	0.003	0.000	0.000	0.949	0.004	0.463	0.022	0.000	0.000
Cl	0.000	0.000	0.000	0.000	0.659	0.017	0.000	0.000	0.000	0.000	0.000	0.000	0.964	0.004
K	0.000	0.000	0.000	0.000	0.012	0.001	0.047	0.002	0.000	0.000	0.312	0.022	0.008	0.001
Ca	0.011	0.002	0.346	0.022	0.019	0.001	0.029	0.001	0.001	0.000	0.000	0.000	0.000	0.000
Ti	0.003	0.000	0.010	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000
V	0.003	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000
Cr	0.005	0.001	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mn	0.011	0.002	0.012	0.001	0.001	0.000	0.001	0.000	0.000	0.000	0.005	0.000	0.000	0.000
Fe	0.836	0.020	0.246	0.019	0.000	0.000	0.000	0.000	0.005	0.001	0.048	0.004	0.001	0.000
Ni	0.001	0.000	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Cu	0.029	0.004	0.010	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.002	0.000	0.000	0.000
Zn	0.009	0.001	0.020	0.002	0.000	0.000	0.000	0.000	0.001	0.000	0.064	0.006	0.000	0.000
Br	0.003	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.007	0.001	0.011	0.001	0.004	0.000
Sr	0.001	0.000	0.001	0.000	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Zr	0.004	0.001	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Mo	0.004	0.001	0.001	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Sn	0.006	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000
Sb	0.005	0.001	0.000	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ba	0.025	0.003	0.002	0.000	0.000	0.000	0.006	0.000	0.001	0.000	0.000	0.000	0.000	0.000
Pb	0.001	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.003	0.000	0.026	0.002	0.001	0.000
Relative contribution														
Element	Traffic		Dust		Sea / road salt		Aged sea salt		S-rich		Solid fuel		Reacted Cl	
	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std	mean	std
Na	0.000	0.000	0.000	0.000	0.240	0.003	0.760	0.003	0.000	0.000	0.000	0.000	0.000	0.000
Mg	0.048	0.005	0.058	0.004	0.143	0.007	0.635	0.011	0.019	0.002	0.054	0.004	0.044	0.004
Al	0.030	0.004	0.558	0.013	0.008	0.000	0.363	0.011	0.030	0.003	0.000	0.000	0.012	0.001
Si	0.062	0.006	0.752	0.011	0.008	0.000	0.016	0.000	0.042	0.003	0.109	0.008	0.011	0.001
P	0.193	0.018	0.130	0.010	0.074	0.004	0.005	0.000	0.192	0.015	0.306	0.019	0.100	0.009
S	0.000	0.000	0.001	0.000	0.029	0.002	0.000	0.000	0.652	0.009	0.318	0.010	0.000	0.000
Cl	0.000	0.000	0.000	0.000	0.406	0.009	0.000	0.000	0.000	0.000	0.000	0.000	0.594	0.009
K	0.000	0.000	0.000	0.000	0.032	0.003	0.125	0.007	0.001	0.000	0.820	0.010	0.022	0.003
Ca	0.027	0.005	0.852	0.010	0.046	0.004	0.072	0.004	0.003	0.000	0.000	0.000	0.000	0.000
Ti	0.188	0.032	0.603	0.040	0.000	0.000	0.000	0.000	0.007	0.001	0.202	0.026	0.000	0.000
V	0.432	0.041	0.216	0.024	0.000	0.000	0.000	0.000	0.071	0.010	0.239	0.028	0.042	0.006
Cr	0.673	0.037	0.318	0.037	0.000	0.000	0.009	0.001	0.000	0.000	0.000	0.000	0.000	0.000
Mn	0.366	0.043	0.399	0.039	0.037	0.004	0.028	0.002	0.000	0.000	0.170	0.022	0.000	0.000
Fe	0.736	0.016	0.216	0.016	0.000	0.000	0.000	0.000	0.004	0.001	0.042	0.004	0.001	0.000
Ni	0.256	0.037	0.197	0.025	0.331	0.028	0.000	0.000	0.056	0.009	0.160	0.022	0.000	0.000
Cu	0.690	0.032	0.230	0.028	0.007	0.001	0.011	0.001	0.000	0.000	0.057	0.008	0.005	0.001
Zn	0.095	0.019	0.209	0.027	0.000	0.000	0.000	0.000	0.012	0.002	0.684	0.035	0.000	0.000
Br	0.124	0.023	0.000	0.000	0.055	0.006	0.000	0.000	0.260	0.033	0.412	0.039	0.149	0.023
Sr	0.148	0.032	0.195	0.028	0.084	0.011	0.438	0.028	0.000	0.000	0.118	0.021	0.015	0.003
Zr	0.839	0.010	0.000	0.000	0.000	0.000	0.152	0.010	0.009	0.002	0.000	0.000	0.000	0.000
Mo	0.620	0.033	0.179	0.024	0.000	0.000	0.136	0.008	0.048	0.008	0.000	0.000	0.018	0.003
Sn	0.605	0.039	0.035	0.005	0.000	0.000	0.000	0.000	0.038	0.006	0.287	0.034	0.035	0.005
Sb	0.702	0.027	0.056	0.008	0.000	0.000	0.077	0.005	0.070	0.010	0.071	0.011	0.024	0.004
Ba	0.728	0.020	0.067	0.010	0.000	0.000	0.183	0.011	0.021	0.004	0.000	0.000	0.000	0.000
Pb	0.026	0.006	0.064	0.010	0.000	0.000	0.000	0.000	0.096	0.016	0.781	0.027	0.034	0.006

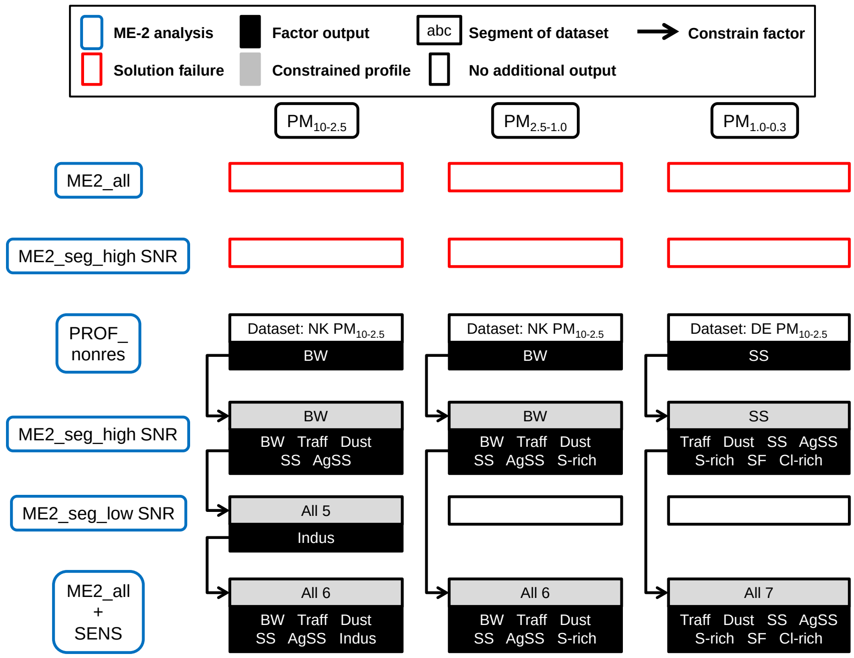

Figure S6. ME-2 analysis strategy for $PM_{10-2.5}$, $PM_{2.5-1.0}$ and $PM_{1.0-0.3}$ on MR-NK-DE sites combined (see Fig. 1 for explanation of blue boxes). MR: Marylebone Road, kerbside; NK: North Kensington, urban background; DE: Detling, rural site. Each step is followed by ME2_all, but always failed except in the last step. Input profiles are constrained with *a* value = 0.1. The model runs were performed on 3–10 factors and 10–20 seeds to explore local minima in the solution space to find those that are most meaningful. Sources: BW: brake wear; Traff: other traffic-related; Dust: resuspended dust; SS: sea / road salt; AgSS: aged sea salt; Indus: industrial; S-rich: S-rich; SF: solid fuel; Cl-rich: reacted Cl. See Sect. 2.3 for more details.

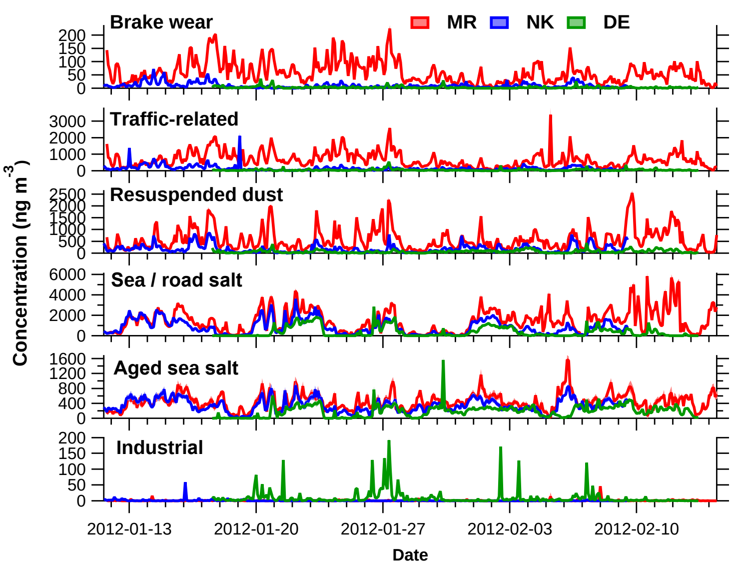

Figure S7. $PM_{10-2.5}$ source contributions (factor time series) according to the ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Data is given as mean ± 1 standard deviation (shaded area) from the anchor sensitivity analysis.

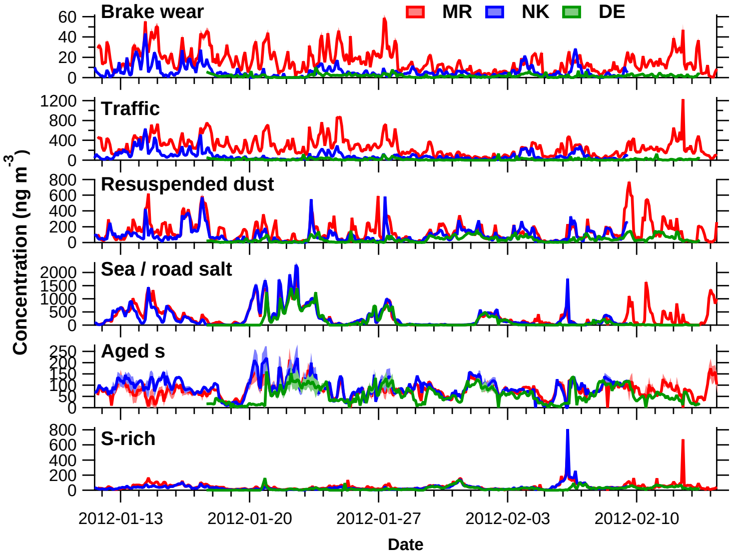

Figure S8. $PM_{2.5-1.0}$ source contributions (factor time series) according to the ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Data is given as mean ± 1 standard deviation (shaded area) from the anchor sensitivity analysis.

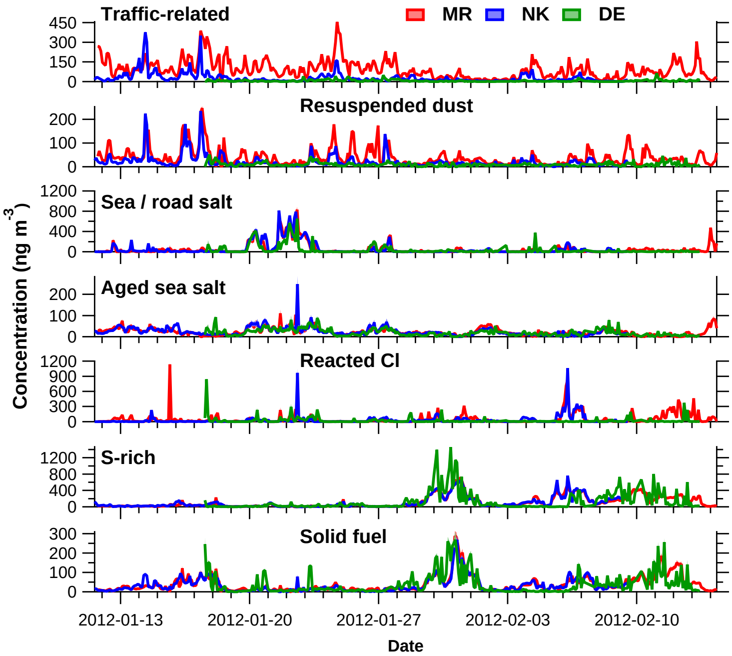

Figure S9. $PM_{1.0-0.3}$ source contributions (factor time series) according to the ME-2 analysis on combined data of the three sites (MR - Marylebone Road, kerbside; NK - North Kensington, urban background; DE - Detling, rural). Data is given as mean ± 1 standard deviation (shaded area) from the anchor sensitivity analysis.

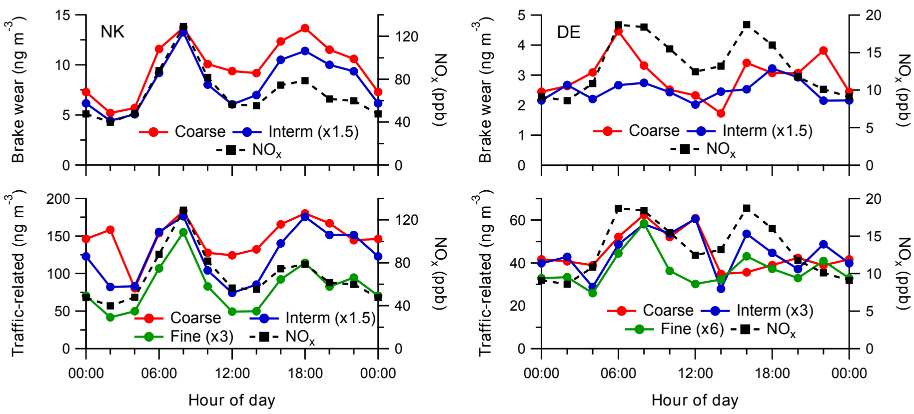

Figure S10. Diurnal variations of the brake wear ($PM_{10-2.5}$ - coarse, $PM_{2.5-1.0}$ - interm) and other traffic-related (coarse, interm, $PM_{1.0-0.3}$ - fine) factors at NK (left) and DE (right) compared to diurnal variations of NO_x. Hour of day is start of a 2 h sampling period (00:00 UTC means sampling from 00:00 to 02:00 UTC). Note the scaling applied to several tracers.

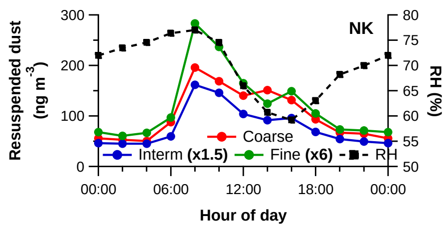

Figure S11. Diurnal variations of the resuspended dust ($PM_{10-2.5}$ - coarse, $PM_{2.5-1.0}$ - interm, $PM_{1.0-0.3}$ - fine) factors at NK compared to the diurnal variation of relative humidity (RH). Hour of day is start of a 2 h sampling period (00:00 UTC means sampling from 00:00 to 02:00 UTC). Note the scaling applied to several tracers.

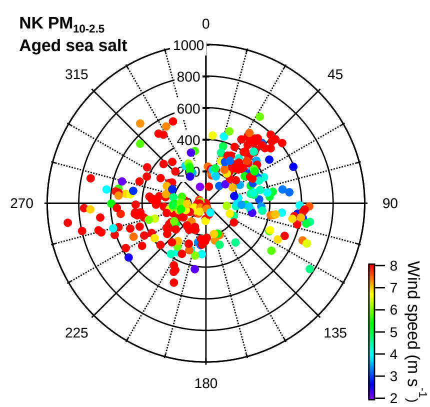

Figure S12. Wind rose of the aged sea salt factor at North Kensington for $PM_{10-2.5}$ (ng m^{-3}), color-coded by the wind speed. Data points with wind speed < 2 m s^{-1} are ignored. Wind roses are similar at Marylebone Road and Detling.

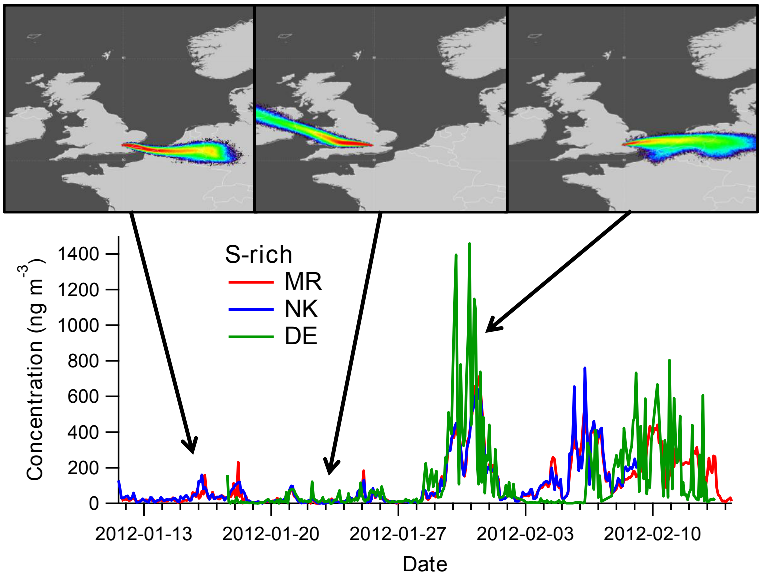

Figure S13. Time series of the S-rich factor in $PM_{1.0-0.3}$ at MR, NK and DE. The three footprints are simulated with the NAME model for particles released from the BT Tower and followed back at 0–100 m, a.g.l for the previous 24 h; particle concentrations increase from blue to red. Periods with high S-rich concentrations correspond to footprints from northern Europe (left and right), whereas low S-rich concentrations correspond to footprints from e.g. the west (centre).

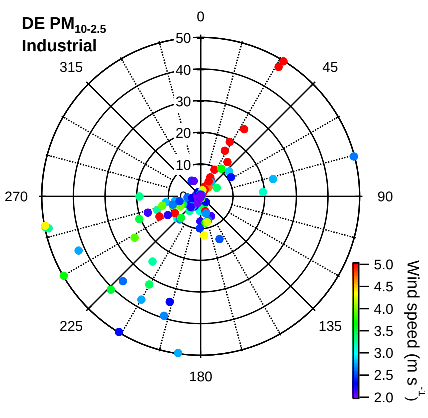

Figure S14. Wind rose of the industrial factor at DE for $PM_{10-2.5}$ (ng m^{-3}), color-coded by the wind speed. Data points with wind speed < 2 m s^{-1} are ignored. Note that data points ≥ 50 ng m^{-3} are set to 50 ng m^{-3} to improve visualisation.