

Fig.S1 Collection characteristics of RDI #1 using QFF or aluminium as substrate (mean \pm sd)

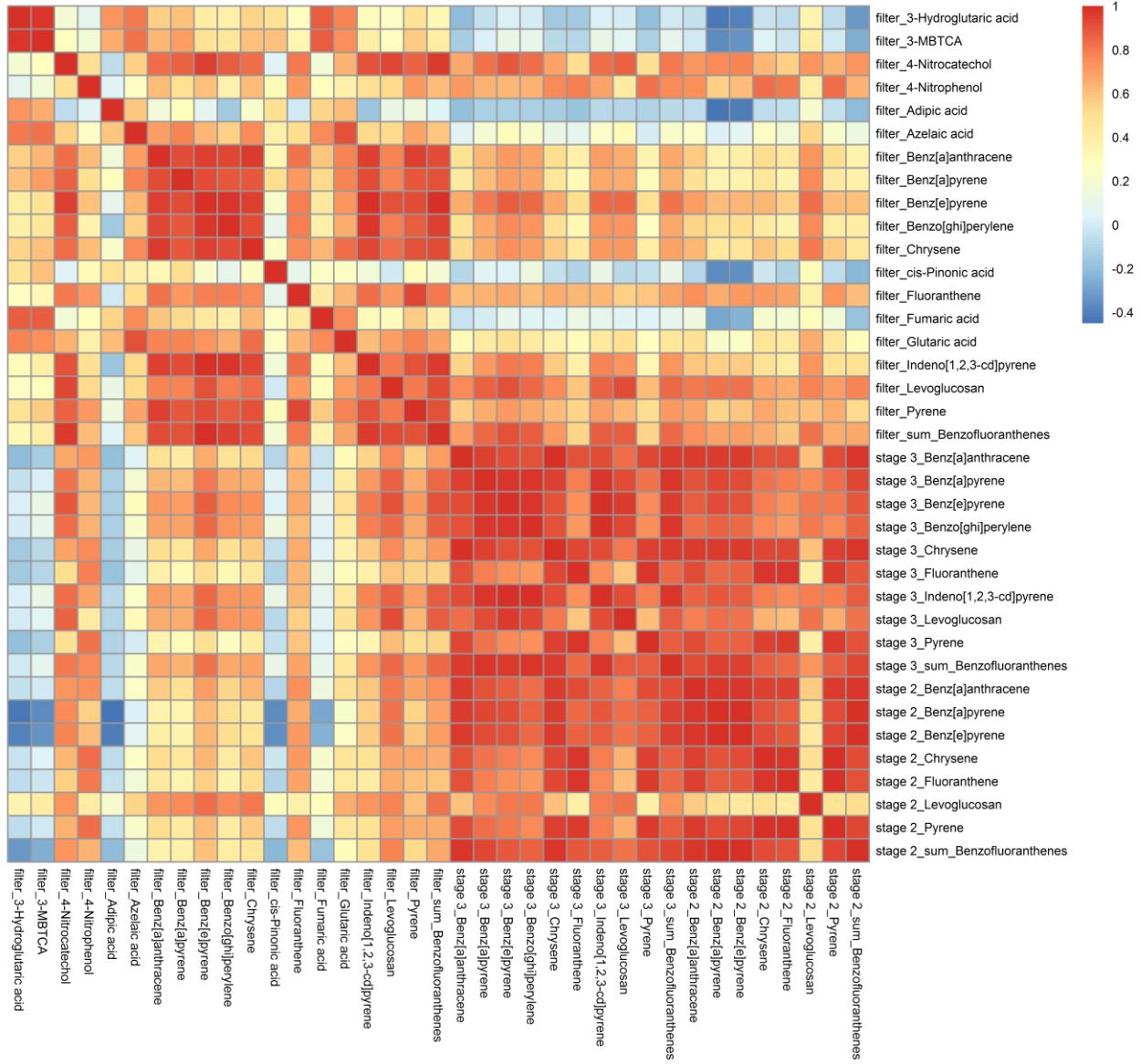


Fig. S2 Correlation matrix of measured compounds from different PM size fractions (stage 2, 1 - 2.4 μm ; stage 3, 0.36 - 1 μm ; filter, < 0.36 μm)

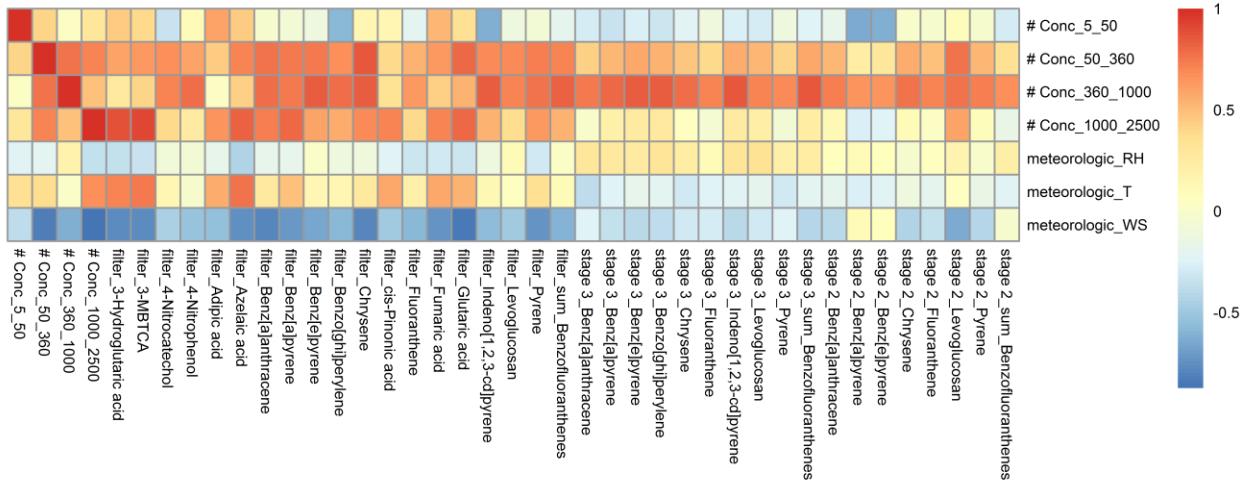


Fig. S3 Correlation matrix between different meteorological parameters, PNC and measured chemical concentrations from different PM size fractions (stage2, 1 - 2.4 μm ; stage 3; 0.36 - 1 μm ; filter, < 0.36 μm ; WS, wind speed; T, temperature; RH, relative humidity; # Conc_5_50, # Conc_50_360, # Conc_360_1000, and # Conc_1000_2500 corresponding to concentration of PM in size range 5 - 50 nm, 50 - 360 nm, 360 - 1000 nm and 1000 - 2500 nm, respectively

Table S1 Sample number and corresponding sampling start time

Sample #	Sampling start time	Weekdays
1	3/25/2014 17:00	Tuesday
2	3/26/2014 17:00	Wednesday
3	3/27/2014 17:00	Thursday
4	3/28/2014 17:00	Friday
5	3/29/2014 17:00	Saturday
8	4/1/2014 10:20	Tuesday
9	4/2/2014 10:20	Wednesday
10	4/3/2014 10:20	Thursday
11	4/4/2014 10:20	Friday
14	4/7/2014 12:10	Monday
15	4/8/2014 12:10	Tuesday
16	4/9/2014 12:10	Wednesday

Table S2 Limit of quantification (LOQ) of the quantified compounds

Quantified compounds	LOQ (ng)
Fluoranthene	0.007
Pyrene	0.006
Chrysene	0.010
Benz[a]anthracene	0.007
Benz[a]pyrene	0.013
Benz[e]pyrene	0.013
sum-Benzofluoranthenes	0.001
Indeno[1,2,3-cd]pyrene	0.008
Benzo[ghi]perylene	0.013
Levoglucosan	0.117
3-Hydroxyglutaric acid	0.052
4-Nitrocatechol	0.070
4- Nitrophenol	0.016
Adipic acid	0.200
Azelaic acid	0.187
cis-Pinonic acid	0.029
Fumaric acid	0.240
Glutaric acid	0.088

Table S3 *T* test summary of PAHs and levoglucosan, mean difference represents the mean of RDI #1 subtracted by mean of RDI #2. (* $P<0.05$)

	< 0.36 μm		0.36-1 μm		1-2.5 μm	
	Mean	P	Mean	P	Mean	P
	difference		difference		difference	
Fluoranthene	-0.015	0.008*	-0.001	0.673	0.002	0.222
Pyrene	-0.010	0.011*	-0.003	0.471	0.000	0.865
Chrysene	0.008	0.262	0.002	0.493	0.000	0.376
Benz[a]anthracene	-0.002	0.154	0.001	0.248	0.000	0.502
Benz[a]pyrene	0.000	0.846	0.000	0.920	0.000	0.173
Benz[e]pyrene	-0.005	0.139	0.001	0.774	0.000	0.482
sum-Benzofluoranthenes	-0.008	0.121	0.008	0.102	0.001	0.368
Indeno[1,2,3-cd]pyrene	-0.001	0.634	-0.002	0.672		
Benzo[ghi]perylene	-0.002	0.424	0.006	0.137		
Levoglucosan	-6.796	0.165	1.559	0.502	0.211	0.558

Table S4 *T* test result of polar compounds (* $P<0.05$)

	Mean	P
	difference	
3-Hydroxyglutaric acid	-0.073	0.535
3-MBTCA	-0.003	0.994
4-Nitrocatechol	-0.302	0.122
4-Nitrophenol	-0.035	0.172
Adipic acid	0.148	0.672
Azelaic acid	0.483	0.537
cis-Pinonic acid	0.161	0.679
Fumaric acid	-0.034	0.745
Glutaric acid	0.033	0.889

Table S5 Descriptive summary of the quantified compounds

Size ranges (μm)	Organic compounds	Mean	Median	SD	Min	Max
< 0.36	3-Hydroxyglutaric acid	1.015	0.812	0.903	0.09	2.943
< 0.36	3-MBTCA	2.846	2.5	2.403	0.256	6.651
< 0.36	4-Nitrocatechol	1.669	1.101	1.801	0.331	6.727
< 0.36	4-Nitrophenol	0.499	0.474	0.103	0.359	0.669
< 0.36	cis-Pinonic acid	10.046	7.683	10.202	1.614	36.537
< 0.36	Adipic acid	3.105	2.935	1.552	1.082	6.506
< 0.36	Azelaic acid	3.427	3.274	1.737	0.956	5.821
< 0.36	Benz[a]anthracene	0.033	0.03	0.018	0.012	0.07
< 0.36	Benz[a]pyrene	0.044	0.033	0.032	0.007	0.109
< 0.36	Benz[e]pyrene	0.07	0.059	0.052	0.009	0.195
< 0.36	Benzo[ghi]perylene	0.059	0.053	0.031	0.026	0.121
< 0.36	Chrysene	0.165	0.175	0.087	0.041	0.337
< 0.36	Fluoranthene	0.229	0.225	0.073	0.133	0.395
< 0.36	Fumaric acid	0.945	0.955	0.501	0.338	1.874
< 0.36	Glutaric acid	1.616	1.458	0.718	0.74	2.86
< 0.36	Indeno[1,2,3-cd]pyrene	0.027	0.023	0.014	0.011	0.056
< 0.36	Levoglucosan	72.0	70.4	38.3	22.6	177
< 0.36	Pyrene	0.168	0.164	0.065	0.094	0.314
< 0.36	sum-Benzofluoranthenes	0.178	0.154	0.117	0.048	0.477
0.36-1	Benz[a]anthracene	0.026	0.023	0.021	0.006	0.069
0.36-1	Benz[a]pyrene	0.058	0.049	0.045	0.008	0.168
0.36-1	Benz[e]pyrene	0.086	0.076	0.065	0.01	0.25
0.36-1	Benzo[ghi]perylene	0.064	0.055	0.049	0.008	0.176

0.36-1	Chrysene	0.086	0.075	0.067	0.017	0.224
0.36-1	Fluoranthene	0.102	0.091	0.074	0.03	0.263
0.36-1	Indeno[1,2,3-cd]pyrene	0.033	0.032	0.024	0.004	0.087
0.36-1	Levoglucosan	52.5	38.8	41.9	9.77	160
0.36-1	Pyrene	0.079	0.067	0.052	0.023	0.184
0.36-1	sum- Benzofluoranthenes	0.184	0.17	0.129	0.025	0.475
1-2.5	Benz[a]anthracene	0.005	0.003	0.004	0.001	0.013
1-2.5	Benz[a]pyrene	0.011	0.009	0.008	0.004	0.028
1-2.5	Benz[e]pyrene	0.016	0.011	0.01	0.006	0.039
1-2.5	Chrysene	0.018	0.015	0.012	0.004	0.042
1-2.5	Fluoranthene	0.041	0.032	0.029	0.012	0.099
1-2.5	Levoglucosan	12.9	13.5	4.81	5.19	22.1
1-2.5	Pyrene	0.023	0.019	0.016	0.007	0.054
1-2.5	Sum--Benzofluoranthenes	0.031	0.023	0.024	0.006	0.083

Method, brief description of IDTD-GC-TOF-MS method:

A 27 mm² size strip of filter samples or one fifth (2 mm) of sampled drum strips was placed in a glass-liner and spiked with isotope-labeled internal standard mixtures for quantification. An autosampler (Combi PAL, CTC Analytics AG, Zwingen, Switzerland) was programmed to moisten the filter with 10 µl silylation reagent N-methyl-N-trimethylsilyl-trifluoroacetamide (MSTFA) and insert the liner into the GC-injector (Optic 3, GL Sciences, Eindhoven, The Netherlands). Organic compounds were thermally extracted from the filter strip at 300 °C. During the desorption step the carrier gas was enriched with MSTFA to ensure the continuous derivatisation. The GC (Agilent 6890, Agilent, USA) was connected to a Time of Flight Mass Spectrometer (Pegasus III, LECO, USA). The capillary column for separation (BPX5, 25 m, 0.22 mm ID, 0.25 um film, SGE, Australia) was kept at 70 °C to focus the desorbed molecules during desorption time, heated up to 130 °C after the thermal desorption by a temperature ramp at 80 °C·min⁻¹ and then subjected to an 8 °C min⁻¹ ramp up to 330 °C followed by holding for 30 min. The column flow and split flow after desorption were set to 0.7 ml min⁻¹. The recorded mass range for TOFMS was 35 - 500 m/z with an acquisition frequency 25·s⁻¹.