Supporting Information

Polyurethane Foam (PUF) Disk Samplers for Measuring Trace Metals in Ambient Air

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Text S1. Analysis of the samples and QA/QC

All measurements were performed using an Agilent Technologies 7900 ICP-MS system (Agilent Technologies, Wilmington, DE, USA), equipped with an octopole collision/reaction system (ORS), a MicroMist nebulizer, a Peltier cooled (2 ◦C) quartz Scott-type double pass spray chamber and an Agilent ASX-520 autosampler. The ICP-MS MassHunter software (Agilent Technologies, Inc., 2015) was used to control all instrument operations including tuning, data acquisition, and data analysis. The instrument was tuned in both modes (normal and reaction cell) using a tuning solution containing 1 μg/ L of Li, Y, Tl, Ce and Co in 2% (v/v) HNO_3. The doubly charged ratio ($^{70}Ce^{++}/^{140}Ce^{+}$) was kept at 0.50 ± 0.02 % and the oxide ratio ($^{156}CeO/^{140}Ce$) was 1.5 ± 0.2 %. Quantification was performed using multi-element matrix-matched calibration standard solutions. Spectral interferences were minimized or eliminated either by choosing a non-interfered isotope (^{27}Al, ^{47}Ti, ^{51}V, ^{55}Mn, ^{59}Co, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{88}Sr, ^{95}Mo, ^{107}Ag, ^{111}Cd, ^{118}Sn, ^{121}Sb, ^{133}Cs, ^{137}Ba, ^{139}La, ^{140}Ce, ^{205}Tl, ^{208}Pb and ^{238}U) or by using the octopole reaction system (ORS) with H_2 as the reaction gas (^{52}Cr, ^{78}Se, and ^{56}Fe). Internal standardization with 0.5 mg/L solution of ^{45}Sc, ^{89}Y, ^{115}In, and ^{165}Ho was used to correct for the instrumental drifts and non-spectral interferences. The internal standard was added on line and was automatically mixed with the sample solution before being introduced into the nebulizer.

The precision and accuracy of the analysis method were checked by analyzing certified reference materials, spikes and duplicates prepared in the same matrix as the digested samples. The 95% expanded relative analytical uncertainties above limits of quantitation (3 times limits of detection) ranged from 10 to 25%. The limits of detection (LOD) were calculated as 3 times standard deviation from replicate analysis of reagent blanks (N=25) treated the same way as samples. The LODs were in the range of 2 to 3000 ng/g (Table SI.3). Urban dust SRM 1648a (National Institute for Science and Technology, Gaithersburg, MD, USA) was also analyzed regularly to check the accuracy of the method and satisfactory recoveries were obtained

Text S2. Crustal Enrichment Factors

Trace metals measured at the 6 sampling sites were further evaluated by calculating Crustal

Enrichment Factors (EFc), which is used to attribute trace metals present in air to crustal sources e.g., soil dust. The Crustal Enrichment Factor is calculated using a double-normalization technique as follows:

$$\text{EFc} = \frac{(Cx/CAl)air}{(Cx/CAl)crust}$$

where Al is used as a reference crust element. The choice of reference element is optional but Fe, Si Ti and Al are typically used as reference elements which are the most abundant elements in earth soil, which have limited sources other than soil.[1] Mason's global compilation of soil was used as reference soil in the calculations.[2] In the formula, $({Cx}/{Al})_{\text{air}}$ is the concentration ratio of an element X to that of Al in the air and $\left({Cx}/{Al}\right)crust$ is the corresponding ratio in earth crust[1] .

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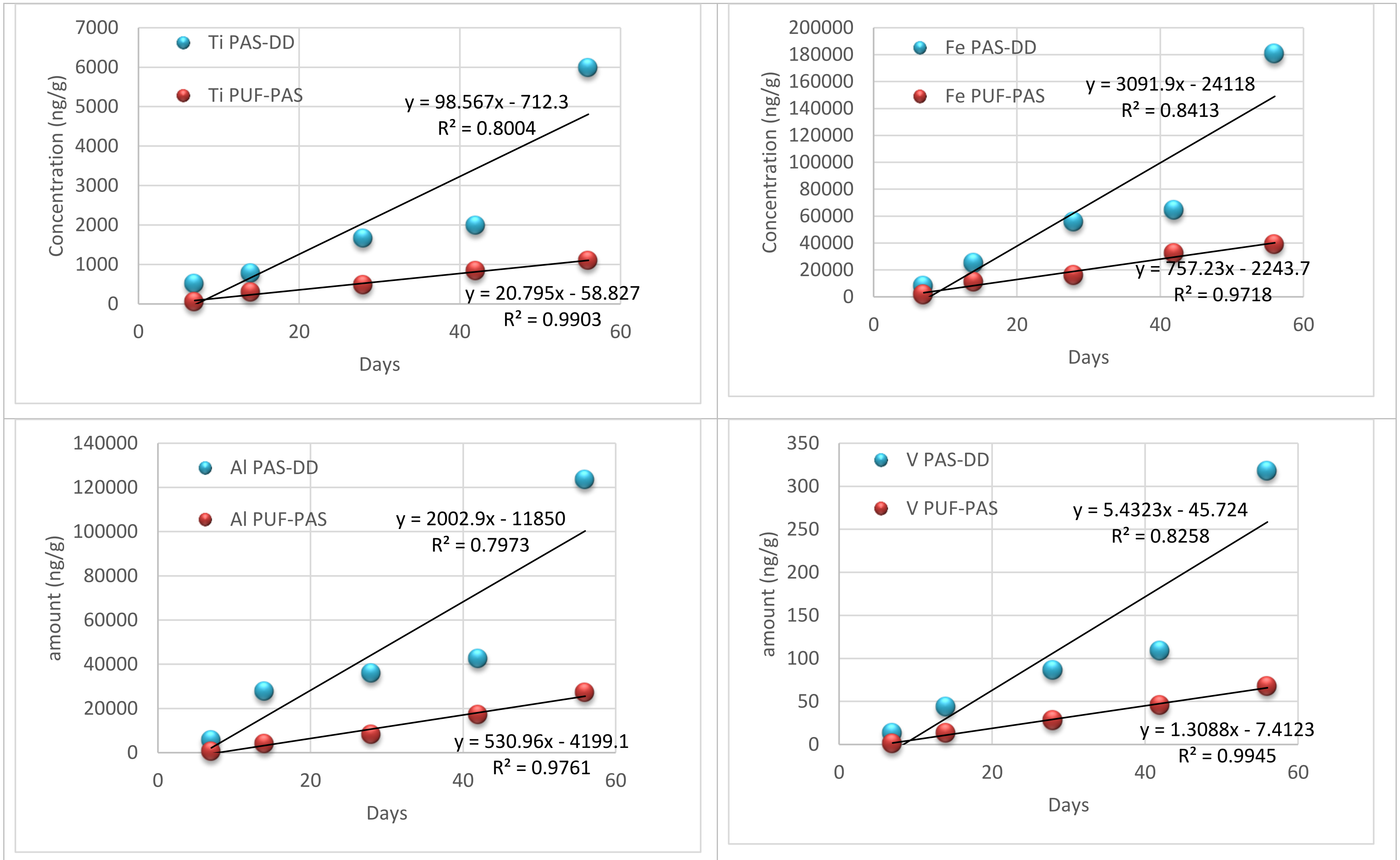
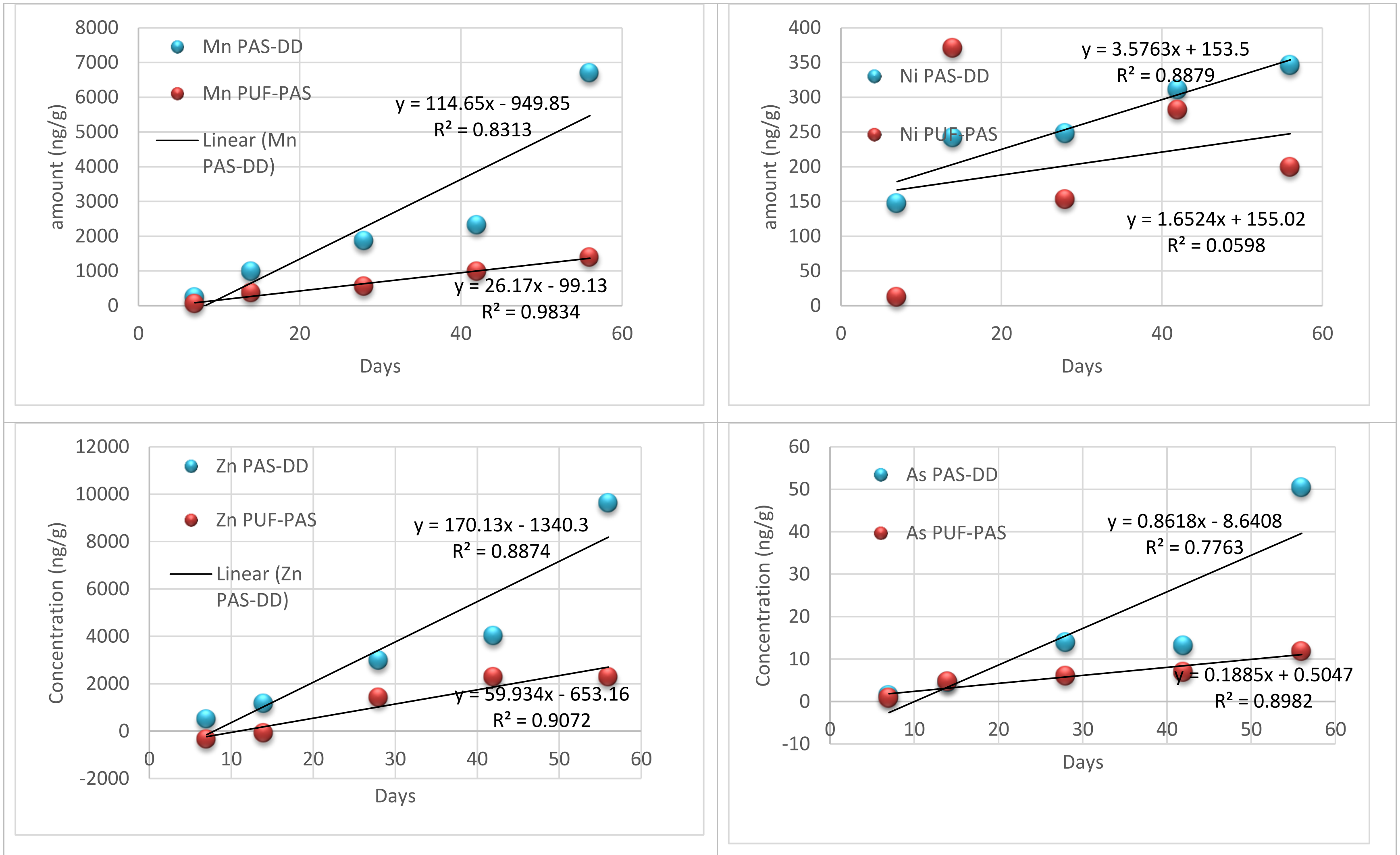


Figure S2. Uptake profiles of trace elements in PUF-PAS and PAS-DD expressed as ng/g PUF, for samplers deployed March 29 – May 17, 2017 at the DV site. Concentrations are expressed as ng per g PUF where 1 g of PUF disk is equivalent to approximately 22.7% of the PUF disk mass or equivalently, 38 cm^2 of the exposed surface area for PUF-PAS and 32 cm^2 of the exposed surface area for the PAS-DD.



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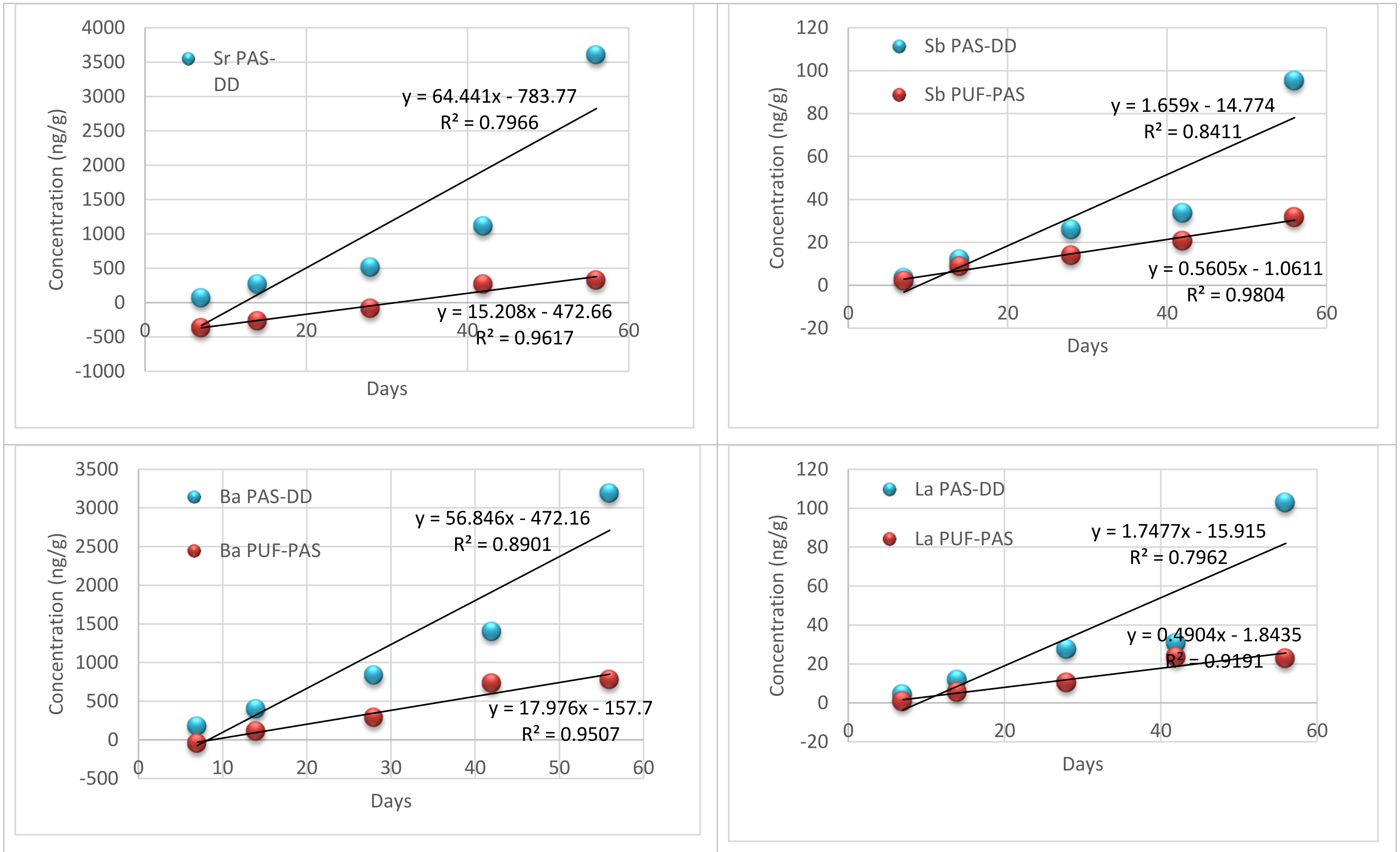

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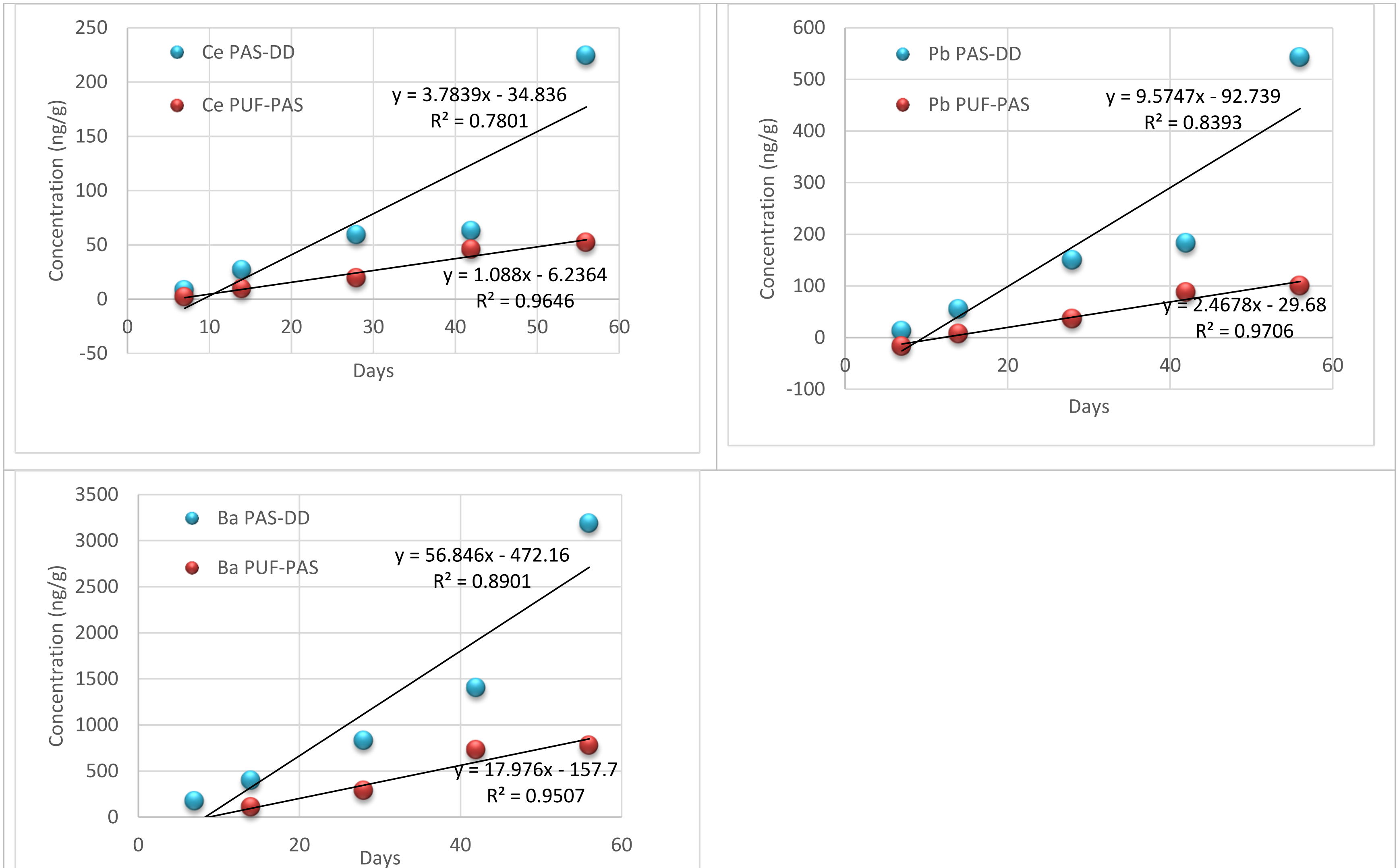

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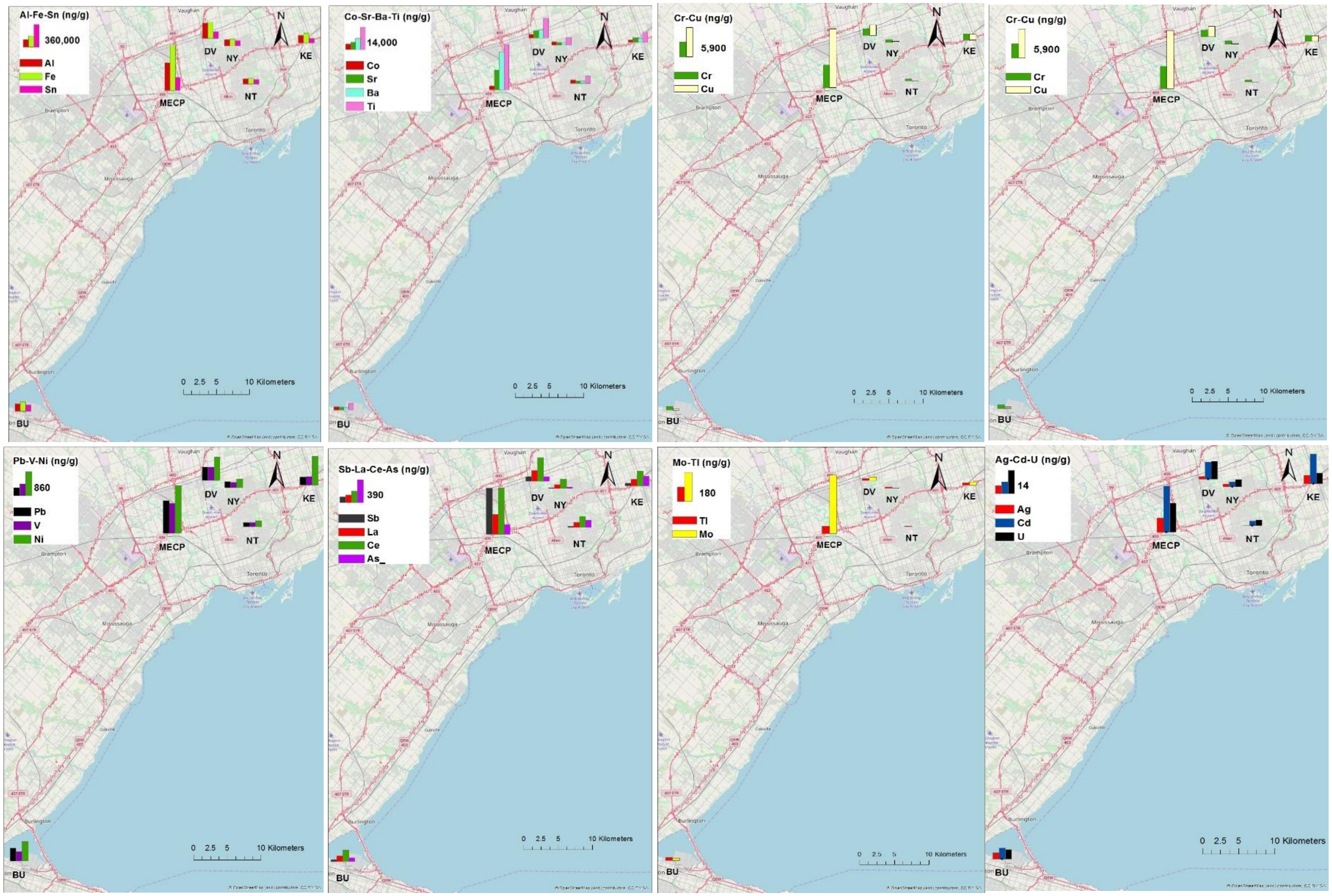

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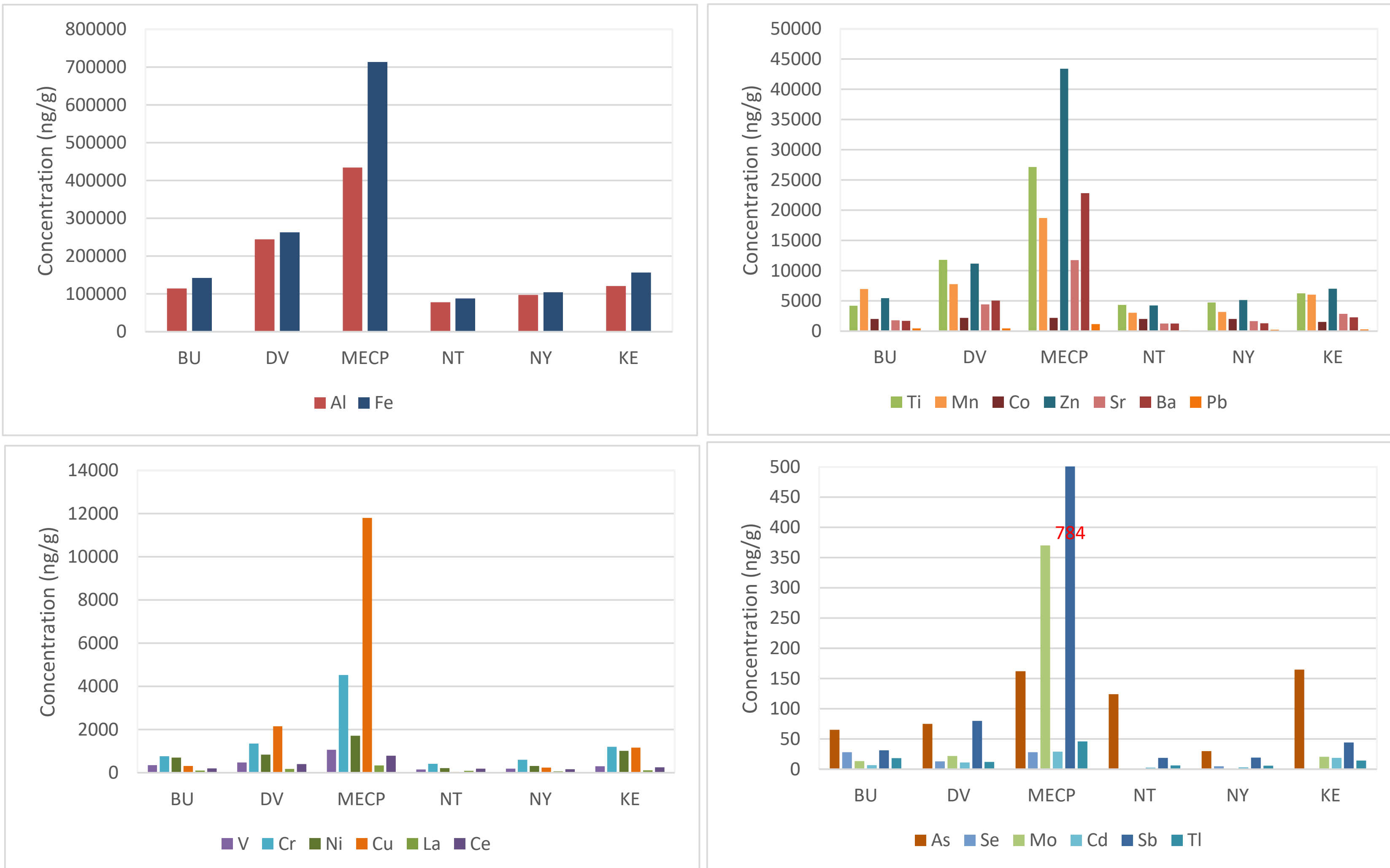

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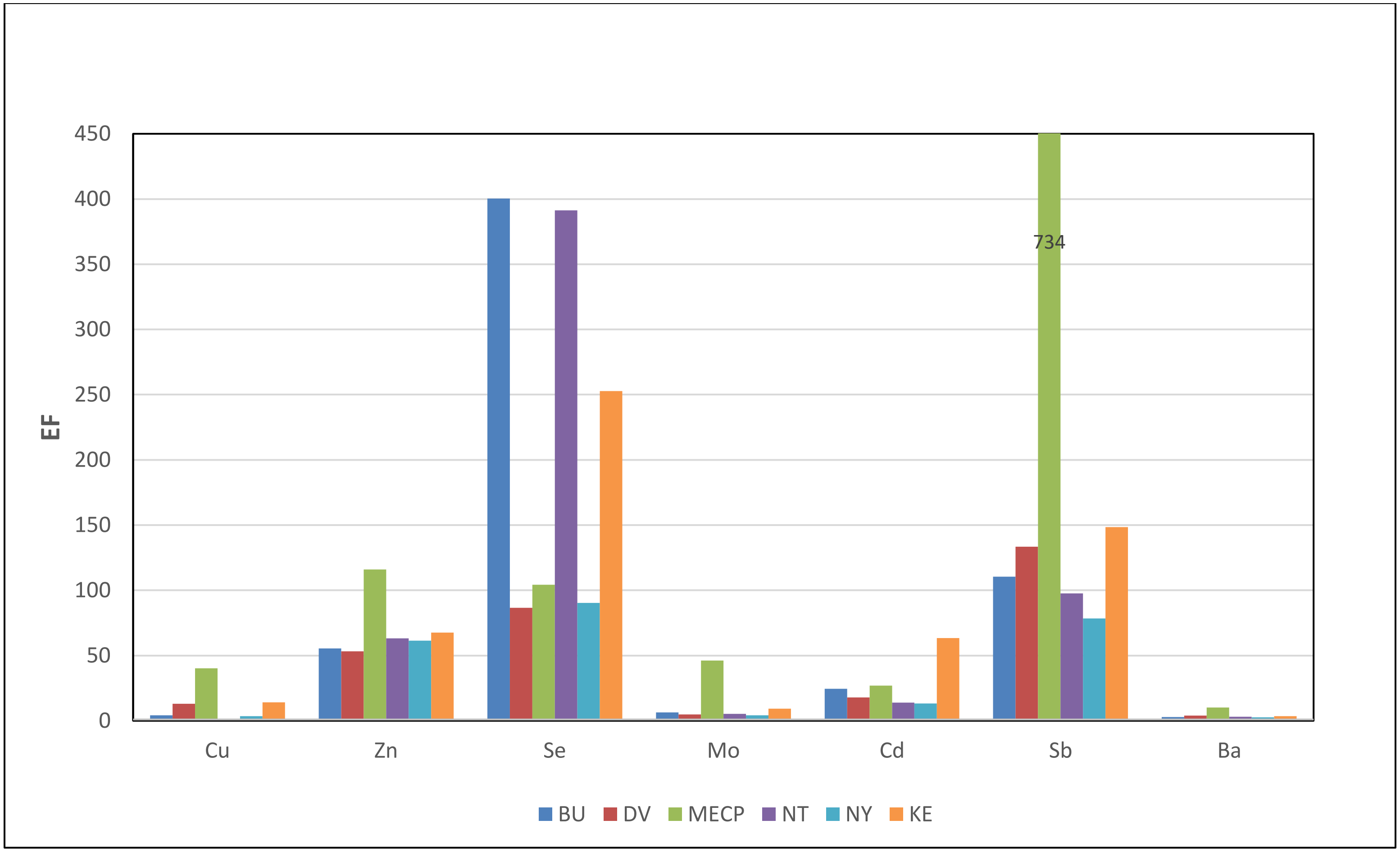

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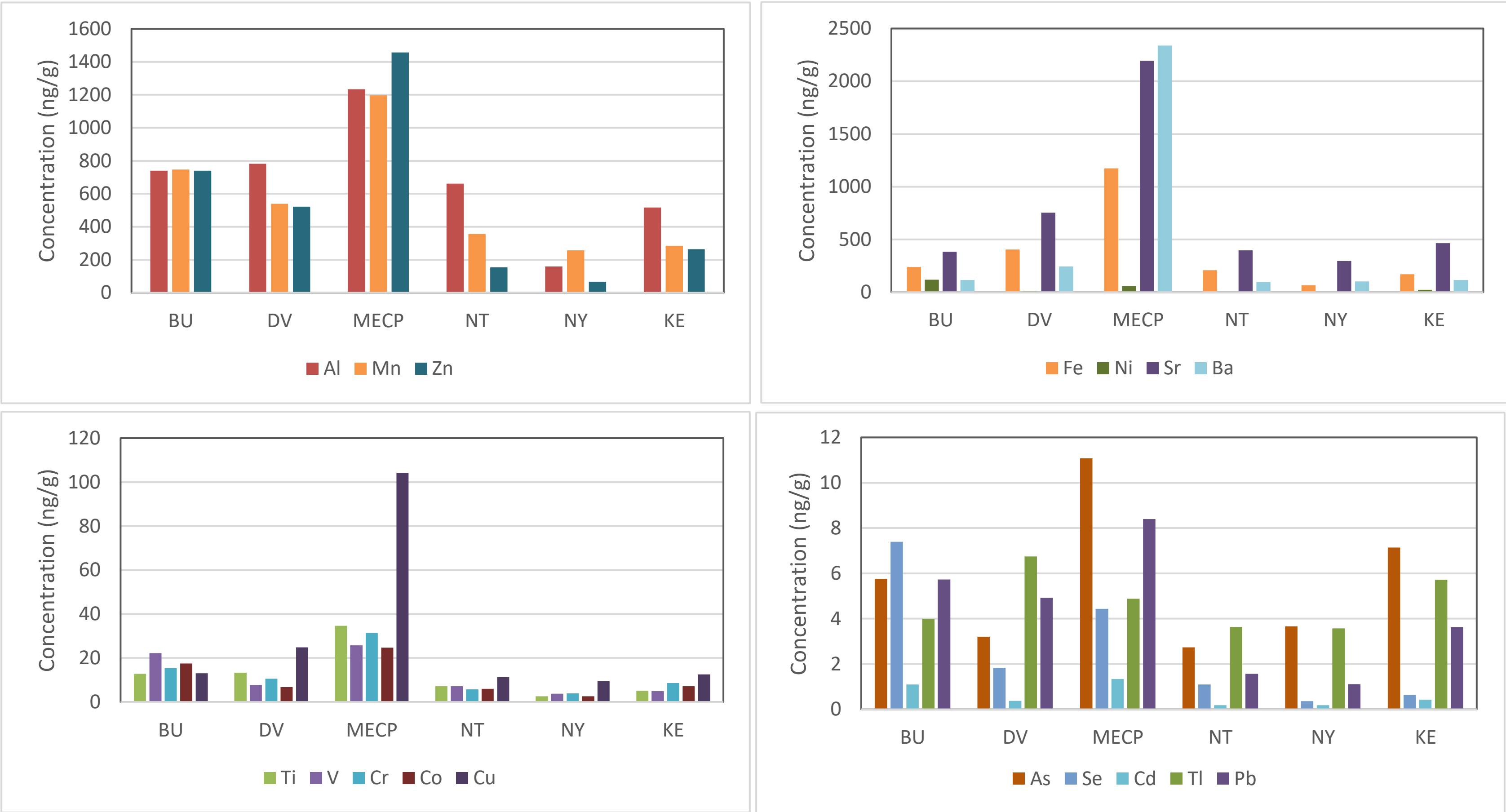

Figure S6. Concentrations of water-soluble trace metals in air (ng/g PUF), derived from PUF-PAS samplers deployed across the Greater Toronto Area during March 29 – May 17, 2017.

Water soluble concentrations of Al, Mn, Zn, Sr, Ba, Cu, As and Pb at the MECP site was almost 2 times higher than at other sampling sites. For the remaining trace metals the differences between sites was less pronounced . Following the MECP site, the DV site had the next highest levels of water soluble trace metals, suggesting an association with traffic emissions since both MECP and DV are traffic-impacted sites.

Table S1. Sampling site and sample collection information.

Sampling Site		Coordinate	Type	Main sources
Ontario Ministry of the Environment, Conservation and Parks	MECP	43°42'39.28"N 79°32'30.60"W	Traffic	High traffic volume and emissions associated with Highway 401.
Kennedy	KE	43°46'22.00"N 79°17'18.45"W	Urban/Traffic	Domestic heating emissions in the winter period, moderate traffic volume
Downsview	DV	43°46'53.41"N 79°28'6.78"W	Urban	Environment and Climate Change Canada (ECCC) field site. Moderate traffic volume and emissions from light industries in the area
Burlington	BU	43°17'55.45"N 79°48'4.68"W	Industrial	Major industrial area, including steel production and close to highway traffic on the Queen Elizabeth Way
North York	NY	43°46'15.21"N 79°25'38.61"W	Residential	Domestic heating emissions in the winter period, with relatively low vehicle traffic volume
North Toronto	NT	43°43'4.50"N 79°23'38.93"W	Residential	Domestic heating emissions in the winter period, with relatively low vehicle traffic volume

Table S2. Homogeneity test results of PUF samples

Sample ID	Mass g	Al ng/g	Ti ng/g	V ng/g	Mn ng/g	Fe ng/g	Co ng/g	Ni ng/g	Cu ng/g	Zn ng/g	As ng/g	Se ng/g	Sr ng/g	Mo ng/g	Cd ng/g	Sb ng/g	Ba ng/g	La ng/g	Ce ng/g	Tl ng/g	Pb ng/g
SD1-2	0.1099	34802	1394	92	1852	61407	1578	1243	1205	7671	19	45	626	65	10	54	2144	31	66	3	570
SD1-1	0.0920	22481	1225	67	1201	38340	1499	443	1006	6314	16	49	599	57	9	50	1236	41	76	3	593
SD1-3	0.1138	36057	1275	87	1627	51271	1539	1313	1106	6999	17	33	593	46	5	39	1306	28	58	3	599
Average		31113	1298	82	1560	50339	1539	1000	1106	6995	17	42	606	56	8	47	1562	33	67	3	588
Std deviation		7502	86	13	331	11562	39	483	100	678	2	8	18	9	2	8	505	7	9	0	15
% RSD (SD1)		24	7	16	21	23	3	48	9	10	9	19	3	17	30	16	32	22	14	5	3
SD2-1	0.0817	22567	1123	67	1215	38162	1515	351	1117	6178	16	62	538	41	8	47	1568	23	48	2	592
SD2-2	0.1133	27726	1320	95	1372	51426	1600	433	1246	7527	15	44	559	70	7	39	1937	34	73	2	543
SD2-3	0.0976	24625	1462	73	1451	43878	1546	809	1408	6971	14	29	523	38	8	50	1618	35	52	2	528
Average		24973	1302	78	1346	44489	1554	531	1257	6892	15	45	540	50	8	45	1708	30	58	2	555
Std deviation		2597	170	15	120	6653	43	244	146	678	1	17	18	18	0	5	200	7	14	0	33
% RSD (SD2)		10	13	19	9	15	3	46	12	10	7	37	3	35	6	12	12	23	24	13	6
SD3-1	0.0926	29683	1330	96	1836	67217	1546	1017	1518	7270	25	90	665	64	6	43	1550	38	71	14	863
SD3-2	0.1092	25346	1130	72	1444	47190	1539	846	1549	6004	19	43	588	33	7	29	1161	33	69	6	668
SD3-3	0.0958	26246	1358	97	1708	59733	1540	1238	1333	6158	20	69	652	110	7	48	1405	33	63	7	877
SD3-4	0.1185	30318	1544	85	1587	55774	1528	1038	1460	6377	15	47	591	49	5	28	1689	38	103	3	604
SD3-5	0.1100	26690	1285	87	1625	69301	1537	1651	1223	6288	16	49	583	49	5	35	1331	30	64	4	612
SD3-6	0.1064	25941	1220	83	1531	57311	1476	1393	1192	5731	16	33	496	41	5	36	1108	23	50	3	554
Average		27371	1311	86	1622	59421	1528	1197	1379	6305	19	55	596	58	6	37	1374	32	70	6	696
Std deviation		2093	140	9	137	8073	26	292	152	525	4	21	60	27	1	8	223	5	18	4	139
% RSD (SD3)		8	11	11	8	14	2	24	11	8	20	37	10	47	15	21	16	17	25	67	20

Table S3. Effect of acid rinsing on blank concentrations

	Before acid clean up PUF disks	After acid clean up of PUF disk
Be ng/g	1.1±1.1	ND
Al µg/g	4.2±2.3	1.1±0.5
Ti ng/g	181.7±66.4	128.2±1.5
V ng/g	9.9±3.3	ND
Cr ng/g	528.7±252.7	16.6±9.5
Mn ng/g	191.3±118.1	21.4±12.6
Fe µg/g	12.4±4.0	1.1±0.2
Co µg/g	1.8±0.3	1.3±0.1
Ni ng/g	229.9±62.3	31.7±13.1
Cu µg/g	2.3±1.8	ND
Zn µg/g	6.8±1.8	7.2±1.8
As ng/g	6.1±1.8	0.9±0.7
Se ng/g	27.3±14.7	0.3±1.0
Sr µg/g	1.2±1.5	ND
Mo ng/g	41.3±58.1	ND
Ag ng/g	4.0±1.9	1.3±2.3
Cd ng/g	6.9±5.3	0.3±0.6
Sn µg/g	120.9±56.8	85.6±20.6
Sb ng/g	10.3±2.5	0.2±1.2
Ba ng/g	290.1±216.5	ND
La ng/g	3.8±1.2	1.2±0.2
Ce ng/g	6.8±2.6	2.9±1.2
Tl ng/g	7.1±5.8	1.1±0.6
Pb ng/g	327.7±283.2	ND
U ng/g	5.4±4.5	ND

Table S4. Duplicate averages of sample to blank ratios (S/B) of trace metals

		Al	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn	As	Se	Sr	Mo	Sb	Ba	La	Ce	Tl	Pb
1 week deployment	PAS-DD	4.4	3.8	4.4	1.4	6.6	4.6	2.3	1.0	1.2	1.6	0.8	1.1	1.2	1.6	2.3	7.1	9.6	1.0	1.5
	PUF-PAS	1.3	1.3	1.3	0.8	2.1	1.7	1.1	0.2	0.9	1.3	0.7	0.3	1.2	1.4	0.6	2.0	3.0	0.0	0.4
2 weeks deployment	PAS-DD	17.6	5.2	12.1	4.5	24.2	11.8	3.2	1.6	1.5	2.9	1.1	1.5	2.3	3.3	3.9	17.8	28.4	1.5	2.9
	PUF-PAS	3.4	2.6	4.4	1.5	9.7	5.8	4.3	0.7	1.0	3.0	1.0	0.5	1.8	2.7	1.8	8.8	10.6	1.1	1.3
4 weeks deployment	PAS-DD	22.6	9.9	22.9	7.0	45.0	25.2	3.2	1.7	2.2	6.8	2.3	2.0	3.1	6.0	7.1	40.4	61.4	3.2	6.3
	PUF-PAS	5.9	3.6	8.1	1.5	14.1	7.9	2.4	1.0	1.6	3.5	2.3	0.8	2.0	3.6	3.1	15.8	20.9	1.0	2.3
6 weeks deployment	PAS-DD	26.4	11.7	28.6	3.4	55.7	29.0	3.8	3.1	2.6	6.5	1.7	3.2	2.9	7.4	11.3	44.9	65.4	2.1	7.4
	PUF-PAS	11.3	5.5	12.5	3.1	24.1	15.0	3.5	1.7	1.9	3.9	1.8	1.5	2.3	5.0	6.3	34.6	47.8	2.5	4.1
8 weeks deployment	PAS-DD	75.1	33.2	81.7	4.0	158.8	79.4	4.1	6.3	4.9	22.0	4.8	8.0	7.6	19.3	24.3	147.8	230.3	8.1	20.1
	PUF-PAS	17.3	6.9	18.2	2.3	34.0	18.0	2.8	2.2	1.9	5.9	2.2	1.6	2.7	7.1	6.7	33.5	54.2	5.1	4.5

Table S5. Typical parameters for the Agilent 7900 ICP-MS operation.

Sample Introduction and Plasma	
RF Power (W)	1450 to 1550
Sampling Depth (mm)	6.5 to 8.5
Carrier Gas (L/min)	0.9 to 1.1
Dilution Gas (L/min)	0.1 to 0.2 (Makeup gas 0.60 for HMI mode)
Nebulizer pump (rps)	0.10
Interface	Nickel sampling cone and Platinum skimmer cone
Ion lens voltages	Optimized for sensitivity in 10 µg L^{-1} tuning solution (Li, Y, Ce, Tl)
Sample uptake rate	0.35 mL min^{-1}
Data acquisition	
Integration time	0.1–0.3 s /point
Peak pattern	3 points/peak
Repetition	4
Collision/Reaction Cell Parameters	
Cell gas flow	5.0 mL min^{-1} H_2
Octopole bias	-14 V
Quadrupole bias	3 – 5 more positive than Octopole bias

Table S6. Limits of detection and average trace metal concentrations in PUF blank.

	MDL, ppb	MDL ng/g	Average blank, ng/g	Calculated. ppb
Be	0.01	8	ND	0.002
Al	3	2250	1729	2
Ti	0.3	225	343	0.2
V	0.07	53	6	0.01
Cr	0.3	225	183	0.1
Mn	0.1	53	13	0.1
Fe	4	3000	3246	3
Co	0.02	15	1707	0.004
Ni	0.2	150	ND	0.1
Cu	0.3	225	ND	0.6
Zn	1	750	3461	0.5
As	0.03	23	2	0.01
Se	0.05	38	3	0.01
Sr	0.2	150	106	0.3
Mo	0.02	15	0.1	0.03
Ag	0.04	30	ND	0.01
Cd	0.02	15	0.4	0.002
Sn	0.05	38	92308	12
Sb	0.04	30	7	0.01
Ba	0.3	225	91	0.3
La	0.003	2	0.7	0.002
Ce	0.004	3	2	0.005
Tl	0.03	23	ND	0.004
Pb	0.1	75	ND	0.1
U	0.01	6	ND	0.01

(1) Calculated for 0.1000 g of PUF

(2) Average of field and travel blanks submitted with samples calculated for 0.1000 g of PUF

Table S7. Average recoveries of digested standard and certified reference materials (NIST SRM 1648 and method spikes, respectively).

Element	NIST 1648a (N=4)			Digested spike (N = 12)	
	Certified concentrations mg/kg	Average measured concentrations mg/kg	Average recovery %	Target concentration ng/mL	Average recovery (blank subtracted) %
Be	n/a	2	n/a	0.1	106
Al	34300	16139	47	30.0	105
Ti	4021	969	24	2.0	103
V	127	109	86	1.0	102
Cr	402	92	23	3.0	107
Mn	790	774	98	0.4	105
Fe	39200	29119	74	60.0	106
Co	18	14	76	0.2	102
Ni	81	72	89	2.0	104
Cu	610	583	96	3.0	101
Zn	4800	4940	103	8.0	101
As	116	144	125	0.3	97
Se	28	32	n/a	0.4	101
Sr	215	173	80	2.0	104
Mo	n/a	18	n/a	0.2	102
Ag	6	7	n/a	0.1	98
Cd	74	84	114	0.2	103
Sn	n/a	513	n/a	n/a	n/a
Sb	45	42	93	0.2	100
Ba	n/a	670	n/a	2.0	104
La	39	30	76	0.0	116
Ce	55	40	73	0.0	112
Tl	n/a	2	n/a	0.3	98
Pb	6550	6882	105	1.0	103
U	n/a	4	n/a	0.1	102

Table S8. Variability in duplicate PAS-DD and PUF-PAS samplers expressed as a ratio of sample 1 / sample 2 collected over 8 sampling periods at the Downsview field site.

Sample ID	Mass (g)	Al	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn
		ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g
PASDD T1-1	0.12271	6756.6	467.4	18.6	283.6	353.0	12466.1	355.3	458.0	3413.2
PASDD T1-2	0.12409	7826.4	930.2	15.9	399.5	203.5	8597.1	162.7	255.8	2539.1
PASDD T1-2/ PASDD T1-1		1.2	2.0	0.9	0.7	1.7	0.7	0.5	0.6	0.8
PASDD T2-1	0.11808	12790.1	781.7	35.5	365.9	808.0	21419.9	292.3	440.2	3418.1
PASDD T2-2	0.10778	45747.2	1146.8	60.0	1795.0	1247.7	33115.9	415.3	763.9	3840.4
PASDD T2-2/ PASDD T2-1		3.6	1.5	1.7	4.9	1.5	1.5	1.4	1.7	1.1
PASDD T4-1	0.11958	31510.5	1926.1	95.9	2018.1	2008.8	65950.3	296.0	564.3	5554.2
PASDD T4-2	0.11121	43813.6	1773.1	84.6	1343.0	1807.8	50036.5	423.6	718.1	5334.5
PASDD T4-2/ PASDD T4-1		1.4	0.9	0.9	0.7	0.9	0.8	1.4	1.3	1.0
PASDD T6-1	0.11904	33285.0	2022.7	91.7	743.2	1902.1	59136.2	420.4	783.9	5047.9
PASDD T6-2	0.10935	54908.0	2329.2	133.4	878.4	2828.5	74324.1	424.7	1499.9	7936.3
PASDD T6-2/ PASDD T6-1		1.6	1.2	1.5	1.2	1.5	1.3	1.0	1.9	1.6
PASDD T8-1	0.11960	111662.8	5490.8	300.3	726.7	6159.9	158453.5	350.0	2204.4	10672.7
PASDD T8-2	0.11755	138734.7	6846.9	343.4	1214.9	7313.4	207372.5	565.7	2459.5	13524.0
PASDD T8-2/ PASDD T8-1		1.2	1.2	1.1	1.7	1.2	1.3	1.6	1.1	1.3
PUF T1-1	0.11850	2577.3	291.8	5.0	171.3	79.4	3756.6	101.7	67.7	1902.8
PUF T1-2	0.11418	1806.5	182.6	5.1	210.4	100.6	4266.0	146.9	71.5	2343.4
PUF T1-2/ PUF T1-1		0.7	0.6	1.0	1.2	1.3	1.1	1.4	1.1	1.2
PUF T2-1	0.12332	6678.2	575.0	18.9	358.0	343.5	12239.1	270.2	311.5	2642.5
PUF T2-2	0.12665	4665.3	381.4	15.4	339.7	475.9	14376.2	695.7	168.7	2125.2
PUF T2-2/ PUF T2-1		0.7	0.7	0.8	0.9	1.4	1.2	2.6	0.5	0.8
PUF T4-1	0.09315	8546.3	580.5	30.5	357.3	536.1	16207.7	209.5	229.6	3685.5
PUF T4-2	0.11718	11245.4	761.9	33.3	371.6	662.1	20321.3	320.5	486.8	4066.4
PUF T4-2/ PUF T4-1		1.3	1.3	1.1	1.0	1.2	1.3	1.5	2.1	1.1
PUF T6-1	0.11678	18181.0	1032.9	48.9	1012.9	1030.3	34630.7	445.7	707.2	4640.7
PUF T6-2	0.10852	19447.8	1009.1	49.7	490.8	1017.5	34654.0	342.7	566.1	4860.4
PUF T6-2/ PUF T6-1		1.1	1.0	1.0	0.5	1.0	1.0	0.8	0.8	1.0
PUF T8-1	0.11195	36940.2	1649.2	93.0	704.1	1786.0	51807.7	428.3	815.5	5634.4
PUF T8-2	0.10679	20698.3	920.3	50.2	395.0	1095.5	30965.8	194.4	800.4	3851.7
PUF T8-2/ PUF T8-1		0.6	0.6	0.5	0.6	0.6	0.6	0.5	1.0	0.7

Table S8. cont'd

Sample ID	Mass (g)	As	Se	Sr	Mo	Sb	Ba	La	Ce	Tl	Pb
		ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g	ng/g
PASDD T1-1	0.12271	4.5	2.8	1027.5	21.0	11.8	365.1	6.0	13.1	0.4	50.5
PASDD T1-2	0.12409	3.1	3.0	130.8	7.6	5.1	256.3	3.9	5.8	0.1	32.5
PASDD T1-2/ PASDD T1-1		0.7	1.1	0.1	0.4	0.4	0.7	0.7	0.4	0.3	0.6
PASDD T2-1	0.11808	6.4	3.9	695.2	30.2	12.1	487.1	11.8	21.8	0.4	62.6
PASDD T2-2	0.10778	7.5	4.0	860.3	25.2	22.1	578.4	13.2	33.9	0.4	104.0
PASDD T2-2/ PASDD T2-1		1.2	1.0	1.2	0.8	1.8	1.2	1.1	1.6	0.8	1.7
PASDD T4-1	0.11958	13.5	9.4	1132.9	40.3	33.3	938.5	29.2	64.5	0.8	197.2
PASDD T4-2	0.11121	19.2	7.5	916.7	35.8	29.0	1000.9	27.5	55.8	0.8	160.4
PASDD T4-2/ PASDD T4-1		1.4	0.8	0.8	0.9	0.9	1.1	0.9	0.9	1.0	0.8
PASDD T6-1	0.11904	14.8	6.1	1774.6	28.9	33.2	1045.0	24.6	51.2	0.5	195.4
PASDD T6-2	0.10935	16.3	6.3	1473.4	40.1	44.4	2033.2	38.3	76.8	0.5	226.8
PASDD T6-2/ PASDD T6-1		1.1	1.0	0.8	1.4	1.3	1.9	1.6	1.5	1.0	1.2
PASDD T8-1	0.11960	43.7	15.7	2269.6	61.8	81.8	2825.9	97.1	215.3	1.5	579.8
PASDD T8-2	0.11755	61.9	19.4	5962.9	121.4	119.3	3827.7	110.1	235.7	2.7	562.4
PASDD T8-2/ PASDD T8-1		1.4	1.2	2.6	2.0	1.5	1.4	1.1	1.1	1.9	1.0
PUF T1-1	0.11850	3.7	2.7	141.5	13.0	8.6	81.9	1.6	3.3	ND	9.4
PUF T1-2	0.11418	2.7	2.3	152.4	15.1	6.1	95.3	1.2	2.5	ND	13.6
PUF T1-2/ PUF T1-1		0.7	0.8	1.1	1.2	0.7	1.2	0.7	0.8		1.4
PUF T2-1	0.12332	10.2	5.2	295.3	20.3	15.5	309.6	8.6	12.1	0.3	33.8
PUF T2-2	0.12665	4.0	2.1	196.1	24.0	12.5	179.7	3.7	8.7	0.3	38.0
PUF T2-2/ PUF T2-1		0.4	0.4	0.7	1.2	0.8	0.6	0.4	0.7	1.0	1.1
PUF T4-1	0.09315	6.0	6.2	242.8	31.2	13.9	327.6	12.4	22.2	0.3	43.8
PUF T4-2	0.11718	10.8	10.4	615.0	16.3	24.1	518.4	9.8	18.8	0.2	86.0
PUF T4-2/ PUF T4-1		1.8	1.7	2.5	0.5	1.7	1.6	0.8	0.8	0.7	2.0
PUF T6-1	0.11678	10.4	6.2	940.5	27.4	24.3	805.9	15.1	32.8	0.5	113.0
PUF T6-2	0.10852	8.2	6.9	621.6	29.2	27.5	928.8	33.4	60.8	0.8	118.8
PUF T6-2/ PUF T6-1		0.8	1.1	0.7	1.1	1.1	1.2	2.2	1.9	1.6	1.1
PUF T8-1	0.11195	13.3	11.6	755.2	32.8	36.7	1035.7	28.9	68.3	2.1	157.7
PUF T8-2	0.10679	15.2	4.2	919.8	31.4	37.1	791.8	18.1	37.9	0.5	98.5
PUF T8-2/ PUF T8-1		1.1	0.4	1.2	1.0	1.0	0.8	0.6	0.6	0.2	0.6

Table S9. Comparison of PAS-DD and PUF-PAS sampling rates (m^3/day) calculated using air concentrations of $PM_{2.5}$ and PM_{10}.

Element	Sampling rates (m^3/day) and correlation coefficients (R^2)				Average dry deposition fluxes (µg/m^2/day)
	PUF-PAS ($PM_{2.5}$)	**PUF-PAS (PM_{10})**	**PAS-DD ($PM_{2.5}$)**	**PAS-DD (PM_{10})**	**PAS-DD**
Al**	8.2, (0.88)	0.72, (0.94)	32, (0.87)	3.5, (0.95)	1809
Ti**	2.3, (0.97)	0.42, (0.98)	12, (0.87)	2.1, (0.89)	84.1
Mn	1.8, (0.89)	0.31, (0.96)	6.1, (0.99)	1.4, (0.96)	24.4
Fe	1.9, (0.94)	0.40, (0.95)	8.1, (0.91)	1.7, (0.93)	6666
Zn	0.30, (0.72)	0.17 (0.66)	0.95, (0.77)	0.5, (0.79)	35.7
Mo*	0.33, (0.97)	NA	1.2, (0.94)	NA	0.312
Ba	0.35, (0.67)	0.080, (0.87)	1.2, (0.71)	0.3, (0.78)	11.6
La*	3.6, (0.89)	NA	6.1, (0.87)	NA	0.344
Ce*	5.1, (0.86)	NA	9.4, (0.89)	NA	0.746

* ICP-MS data was used.

** Since the recoveries of Al and Ti from SRM (Standard Reference Material) matrix were low, the amounts were corrected using SRM recoveries (Table SI.7) for flux calculations.

NA: Not available

Table S10. Concentrations of trace metals (water soluble) measured at sampling sites (ng/g PUF).

Water soluble concentrations of Al, Mn, Zn, Sr, Ba, Cu, As and Pb at the MECP site was almost 2 times higher than at other sampling sites. For the remaining trace metals the differences between sites was less pronounced.

	BU	DV	MECP	NT	NY	KE
Be	0.11	0.09	0.06	0.06	ND	0.02
Al	739.64	781.41	1233.81	661.34	159.42	517.69
Ti	12.81	13.31	34.67	7.16	2.63	5.11
V	22.25	7.67	25.71	7.16	3.85	4.97
Cr	15.40	10.53	31.36	5.71	3.86	8.62
Mn	747.70	540.49	1196.43	355.78	256.71	284.71
Fe	239.97	404.37	1173.70	209.18	66.32	170.36
Co	17.58	6.85	24.72	6.06	2.66	7.14
Ni	118.37	13.56	59.30	5.00	3.63	22.85
Cu	13.13	24.82	104.28	11.43	9.49	12.54
Zn	740.89	522.78	1457.03	154.69	66.32	263.61
As	5.75	3.21	11.08	2.74	3.66	7.14
Se	7.40	1.83	4.44	1.10	0.37	0.64
Sr	382.88	753.79	2194.25	397.22	295.26	465.56
Mo	4.26	4.24	34.37	2.29	1.89	2.47
Ag	0.45	0.22	0.49	ND	ND	0.01
Cd	1.10	0.38	1.34	0.19	0.19	0.43
Sn	51.29	12.13	58.96	30.10	17.36	11.45
Sb	7.32	13.70	110.43	5.86	4.08	5.31
Ba	115.50	245.52	2337.71	96.32	102.74	115.82
Tl	3.99	6.74	4.88	3.64	3.57	5.72
Pb	5.74	4.92	8.40	1.56	1.11	3.63
U	0.11	0.03	0.07	0.02	n	0.03

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