Supporting Information for

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Number of pages- 22

Number of figures- 11

Number of tables- 6

Exposure assessment methods, QA/QC, and exposure descriptives.

Gravimetric sampling methods and elemental data compilation

Flow rates for gravimetric samples were measured at the beginning and end of each three-hour session and samples were stored in sealed bags between each session. Samples were deemed valid if they sampled successfully for 75% of the 30 hours with an average flow within 10% of the target 4 litres per minute (lpm). Field blanks were deployed (10% of field samples) and analysed with active samples. Teflon filters were weighed pre- and post- sampling after a 24-hour preconditioning period. After gravimetric analysis, Teflon filters were analysed for elemental composition using inductively coupled plasma-mass spectrometry (ICP-MS) for 36 elements. Median field blank concentrations found to be greater than the lab detection limit were used to blank-correct both gravimetric and elemental results. Microaethalometer data were integrated to a 1-week period to correspond to the integrated $PM_{2.5}$ samples and their associated elemental datasets. This was done to include black carbon into the source appointment analysis.

Continuous data QA/QC

Quality control for continuous measurements was ensured by factory calibration before each seasonal campaign. After each seasonal campaign, co-located inter-comparative sampling sessions were completed (i.e., concurrent monitoring by all units for at least one hour of a small burning event) to permit quantification of instrument precision, bias, and overall limit of detection as per Wallace et al (2011)[1]. Table S1 provides the descriptive statistics of these co-location sessions along with the limit of detection, bias and bias-corrected precision estimates. Continuous DustTrak sampling data for $PM_{2.5}$ agreed well with the filter-based time-integrated PM samples in all three cities. While the DustTrak has been noted to over-predict PM exposures relative to gravimetric methods in ambient environments, the inter-method ratio was very close to unity (Figure S2) in the metro environments sampled. This could be on account of the larger fraction of traditionally crustal materials (in this case metallic materials) increasing the density of particles up to the Arizona road dust used in calibration. The DustTrak units were checked for zero drift before and after each three-hour sampling session.

Continuous data management

The internal clocks of all continuous instruments, including digital voice recorders (DVRs), were synchronized to the technicians' computers on a daily basis. Continuous data were merged at one-second intervals and assigned to a rail segment or station based on DVR recordings of station names and boarding/disembarking activities. One-second GPS latitude and longitude values were converted into Universal Transverse Mercator (UTM) values to allow averaging of UTM *x* and UTM *y* coordinate values. Missing UTM data due to lost satellite signal while riding in subterranean portions of the metro systems were supplemented by inputting the known metro station coordinates while waiting on platforms. For riding periods, the linear extrapolation of the departed and destination station coordinates was used. All continuous data were then averaged by rail segment/station and sampling week.

Continuous data coverage

Sampling covered the entirety of each system with the exception of the 'yellow line' of the Montreal metro. This line comprises two stations and two rail segments and was omitted for logistical reasons. The stations and rail segments of each metro were sampled continually over the course of each three week sampling period. Since all routes were covered within each three hour sampling period, resulting in numerous visits to each station and rail segment (Table S2). On average, the study collected over two hours of continuous data at each station and rail segment of each metro.

Integrated sampling

Table S4 presents concentrations and percent of samples above detection for $PM_{2.5}$ samples collected with personal environment monitors along with the elemental results of $PM_{2.5}$ samples by ICP-MS. Gravimetric analyses found all samples to be above detection limit. Detection limits for $PM_{2.5}$-associated elements are species-specific. A high percentage of samples were found to be above detection for all elements. Gravimetric and elemental blank corrections were not conducted because median blank values were below detection in all cities and seasons. All $PM_{2.5}$ samples were valid in Toronto and Vancouver. In Montreal three PM samples were lost due to pump failures. This yielded a dataset of 18 $PM_{2.5}$ samples in Toronto and Vancouver and 15 samples in Montreal. All valid $PM_{2.5}$ samples were analysed for elemental composition.

System factors

Depth values for each platform and rail segment in the Toronto and Vancouver metros were estimated as the number of steps from surface to platform multiplied by step riser height. For Montreal, depth was obtained from the Montreal transit authority for each platform. Depths of rail segments were assigned the mean depth of the two adjoining platforms. ‘Elevation’ was defined as meters above sea level for each station and rail segment. This metric provided a measure for the vertical position of a segment of the metro, relative to the rest of the system. This metric was also independent of ‘depth’. ‘Percent coverage’ was defined as proportion of a station or rail segment covered or below grade. Stations were assigned 0% (above-grade) or 100% (below-grade). Rail segments were assigned a continuous value by review of metro network information using ArcGIS. ‘Distance to outside’ was calculated for each GPS point as the distance to the closest above-grade station or rail segment. Other design features such as year of construction were also considered.

Table S1: Descriptive statistics of co–location sampling conducted after each sampling session with limits of detection, bias and precision.

Instrument (pollutant & units)	Session*	N units	time (hours)	mean	limit of detection	Bias		Bias–Corrected Precision(%)	
						median	range	Median	Range
DustTrak ($PM_{2.5}$) (µg/m^3)	T&M summer	7	9.0	2.79	3.2	1	0.6 – 1.4	21	14-31
	T&M winter	6	22.7	38.5	10	1.1	1.0 – 1.2	27	16-38
	V winter	7	1.4	12.9	3	1.1	0.7 – 1.4	30	21-36
	V summer	7	1.4	6.83	3	1.2	0.3 – 1.9	27	23-45
CPC (UFP) (pts/cm^3)	T&M summer	7	8.9	4527	1573	1.1	0.5 – 1.6	19	8-28
	T&M winter	6	3.4	11096	1211	1	0.6 – 1.3	22	10-43.6
	V winter	5	1.5	35346	3547	1	0.9 – 1.1	14	3-22
	V summer	8	1.2	5249	2418	1	0.8 – 1.1	10	5-14
Micro Aeth (Black Carbon) (ng/m^3)	T&M summer	8	12.9	1579	208	1.1	0.9 – 1.1	6	4-33
	T&M winter	4	3.1	1584	100	1.1	0.8 – 1.4	5	2-24
	V winter	6	1.5	212	218	0.8	0.4 – 1.5	17	12-36
	V summer	5	0.5	370	302	1.4	0.3 – 1.8	25	11-31

*T&M = Toronto & Montreal, V = Vancouver

Table S2. Descriptive statistics for quantity of continuous exposure data collected by system and station/rail segment

City	Status	n sampled	minutes of data		n visits	
			mean	SD	mean	SD
Toronto, ON	stations	64	231	80	66	20
	rail segments	67	229	113	95	38
Montreal, QC	stations	66	256	88	57	21
	rail segments	67	152	54	78	22
Vancouver, BC	Stations	47	342	115	94	33
	rail segments	47	273	102	128	42

Table S3. Metro exposures by season, riding/waiting, and above/below grade.

pollutant	value	Toronto		Montreal[α]		Vancouver	
		n	median (Q1-Q3)	n	median (Q1-Q3)	n	median (Q1-Q3)
$PM_{2.5}$ ($\mu g/m^3$)	winter	381	82.3 (57.8-126.1)	399	46.9 (37.2-56.1)	282	24.4 (19.6-31.1)
	summer	337	119.6 (78.4-165.9)†	396	26.3 (21.9-34.4)†	282	11.3 (9-18.6)†
	riding	361	80.8 (61.2-106.8)	402	36.3 (27.3-48.3)	282	18.6 (11.7-24.9)
	waiting	357	140 (78.9-183.5)†	393	35.2 (25.2-46.5)	282	20.6 (10.9-32)‡
	above-grade	102	46.7 (31.2-74.5)	-	-	423	15.1 (10.1-23.5)
	below-grade	616	106.4 (73-157.7)†	795	35.6 (25.8-47.4)	141	39.7 (26.1-61.2)†
UFP (10^3pts/cm^3)	winter	381	10.5 (7.3-15.8)	399	23.5 (17.9-31.7)	282	18.3 (11.4-28.2)
	summer	287	7.2 (5.6-9.6)†	376	11.6 (9.1-15.9)†	282	3.8 (3.1-5.2)†
	riding	337	6.7 (5.2-9.8)	393	17.2 (12.3-24.5)	282	6 (3.4-16.5)
	waiting	331	11 (8.7-16.6)†	382	17.1 (11.3-24.1)	282	8.3 (4.9-21.4)†
	above-grade	100	12 (7.2-16.3)	-	-	422	6.8 (3.6-15)
	below-grade	568	8.8 (5.9-11.6)†	775	17.1 (11.6-24.4)	142	11 (4.6-24.2)†
BC (ng/m^3)	winter	267	2.6 (1.4-5.2)	399	4.8 (3.6-7)	282	2.8 (1.9-3.6)
	summer	324	9.6 (6.4-14.4)†	396	3.5 (2.6-4.5)†	282	1.4 (1-2.6)†
	riding	304	4.6 (2.3-7.9)	402	4.3 (3.2-5.5)	282	2 (1.3-3.1)
	waiting	287	8 (3.7-14.8)†	393	3.9 (2.8-5.3)‡	282	2.1 (1.2-4)
	above-grade	97	3.2 (2-6.5)	-	-	423	1.6 (1.1-2.6)
	below-grade	494	6.9 (3.4-11.6)†	795	4.1 (3-5.4)	141	4.4 (3.2-7.2)†

[α] metro system entirely below grade; [‡] $p<0.05$; [†] $p<0.0001$

Table S4. Ambient and metro $PM_{2.5}$ elemental concentrations and abundance ratios (ARs) in all three cities.

		'Metro $PM_{2.5}$* n = (T=18;M=15;V=18)			Ambient $PM_{2.5}$* n = (T=17;M=23;V=19)		
City	element	% detected	median (IQR) (ng/m^3)*	median abundance ratio	% detected	median (IQR) (ng/m^3)	median abundance ratio
Toronto, Ontario	Al	100	441.3 (262.5-513.7)	0.00464	100	8.8 (6.5-15.7)	0.00070
	As	100	4.2 (2.7-5.3)	0.00005	100	0.4 (0.3-0.7)	0.00003
	Ba	100	1609.6 (899.3-1843.7)	0.01654	100	2.5 (1.6-3.0)	0.00018
	Cd	100	0.33 (0.23-0.45)	0.00000	100	0.10 (0.06-0.15)	0.00001
	Cr	100	132.46 (90.82-173.19)	0.00156	100	0.42 (0.38-0.58)	0.00003
	Cu	100	381.6 (319.2-442.0)	0.00443	100	2.4 (1.7-3.2)	0.00018
	Fe	100	52191 (31501-61540)	0.54844	100	17 (10-34)	0.00162
	Mn	100	431.0 (241.8-513.7)	0.00431	100	1.3 (1.0-2.2)	0.00011
	Mo	100	8.2 (6.2-10.5)	0.00010	100	0.2 (0.1-0.3)	0.00001
	Ni	100	19.8 (14.4-27.0)	0.00025	88	0.3 (0.2-0.3)	0.00002
	Pb	100	53.3 (48.8-69.5)	0.00069	100	2.2 (1.3-2.5)	0.00012
	Sr	100	6.3 (5.0-8.1)	0.00010	100	0.5 (0.4-0.7)	0.00004
	V	100	2.9 (1.6-3.2)	0.00003	94	0.3 (0.1-0.4)	0.00002
	Zn	100	131 (85-167)	0.00154	100	18 (10-33)	0.00120
Montreal, Quebec	Al	100	235.1 (92.5-270.6)	0.00681	100	7.5 (4.2-12.5)	0.00076
	As	100	0.7 (0.6-1.1)	0.00003	100	0.3 (0.2-0.6)	0.00004
	Ba	100	2.9 (2.5-3.1)	0.00010	100	1.0 (0.5-2.0)	0.00011
	Cd	100	0.45 (0.24-0.58)	0.00001	100	0.08 (0.06-0.15)	0.00001
	Cr	100	4.04 (2.06-5.36)	0.00015	48	0.20 (0.15-0.32)	0.00002

	Cu	100	1646.9 (680.8-2126.3)	0.05605	91	1.4 (0.7-3.1)	0.00014
	Fe	100	1416.1 (651.9-1503.4)	0.04314	100	7.8 (4.1-26.1)	0.00113
	Mn	100	19.8 (9.4-24.0)	0.00067	100	0.9 (0.6-2.1)	0.00010
	Mo	100	0.76 (0.46-0.96)	0.00003	57	0.08 (0.04-0.23)	0.00001
	Ni	100	5.8 (2.7-7.5)	0.00018	70	0.2 (0.1-0.4)	0.00002
	Pb	100	15.0 (9.4-21.0)	0.00048	100	1.3 (0.6-2.6)	0.00015
	Sr	100	1.8 (0.6-2.3)	0.00005	100	0.4 (0.2-0.6)	0.00004
	V	100	0.9 (0.6-1.1)	0.00003	87	0.4 (0.1-1.0)	0.00003
	Zn	100	60 (33-90)	0.00206	100	14 (7-29)	0.00120
Vancouver, British Columbia	Al	100	34.7 (26.0-58.8)	0.00208	100	5.9 (3.9-10.1)	0.00110
	As	100	1.0 (0.8-1.6)	0.00007	100	0.3 (0.2-0.4)	0.00005
	Ba	100	5.9 (3.8-7.3)	0.00034	100	1.3 (1.0-2.0)	0.00026
	Cd	100	0.16 (0.08-0.22)	0.00001	58	0.03 (0.02-0.06)	0.00001
	Cr	100	22.78 (7.72-45.29)	0.00154	11	0.19 (0.18-0.25)	0.00003
	Cu	100	40.8 (35.6-57.8)	0.00221	100	1.5 (1.2-2.1)	0.00033
	Fe	100	1954 (526-6029)	0.17091	100	6 (4-10)	0.00123
	Mn	100	24.0 (11.9-63.4)	0.00226	100	1.9 (0.7-3.3)	0.00042
	Mo	100	5.9 (1.6-78.3)	0.00063	74	0.1 (0.1-0.2)	0.00003
	Ni	100	9.1 (6.3-19.8)	0.00078	95	0.5 (0.2-0.8)	0.00008
	Pb	100	4.7 (2.1-9.8)	0.00032	100	0.9 (0.6-1.4)	0.00015
	Sr	100	0.6 (0.4-0.9)	0.00004	100	0.2 (0.2-0.3)	0.00003
	V	100	5.5 (1.9-7.7)	0.00031	100	1.0 (0.4-1.7)	0.00016
	Zn	100	29 (22-47)	0.00245	100	8 (7-13)	0.00161

Table S5. Spearman's correlation analysis for $PM_{2.5}$, UFP, and black carbon exposures measured by continuous samplers in below-grade sections of metros.

city	pollutant	n	Spearman's correlation		
			$PM_{2.5}$	UFP	BC
Toronto, ON	$PM_{2.5}$	616	1.00	0.12	0.52
	UFP	564		1.00	0.20
	BC	493			1.00
Montreal, QC	$PM_{2.5}$	795	1.00	0.49	0.51
	UFP	775		1.00	0.09
	BC	795			1.00
Vancouver, BC	$PM_{2.5}$	141	1.00	0.14	0.90
	UFP	140		1.00	0.11
	BC	141			1.00

Table S6. Spearman's correlation analysis for $PM_{2.5}$, UFP, and black carbon exposures measured by continuous samplers in above-grade sections of metros.

city	pollutant	n	Spearman's correlation		
			$PM_{2.5}$	UFP	BC
Toronto, ON	$PM_{2.5}$	102	1.00	-0.34	0.57
	UFP	96		1.00	-0.20
	BC	91			1.00
Vancouver, BC	$PM_{2.5}$	423	1.00	0.72	0.73
	UFP	421		1.00	0.59
	BC	423			1.00

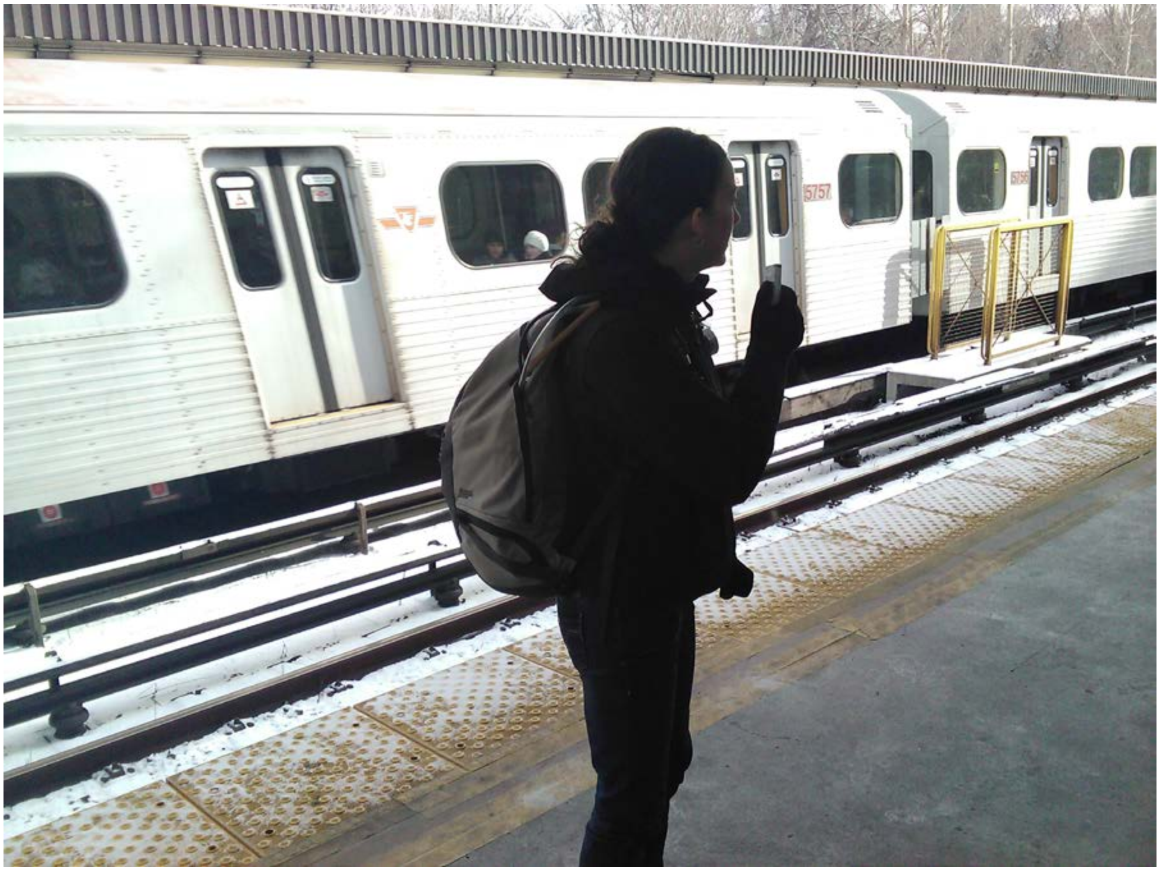

Figure S1. Field technician sampling setup**.**

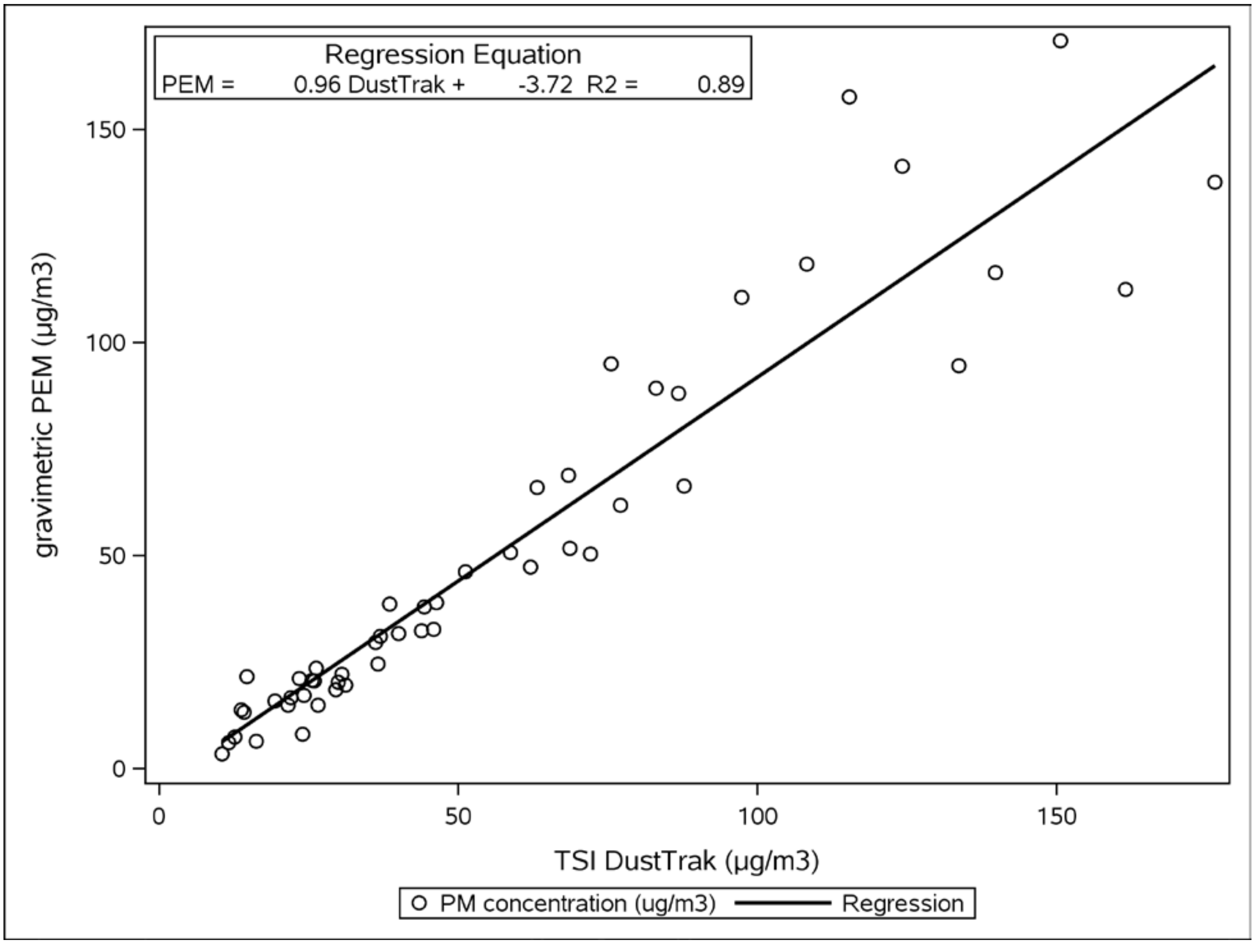

Figure S2. Inter-instrument agreement for $PM_{2.5}$ continuous monitor (DustTrak) and integrated filter-based samplers.

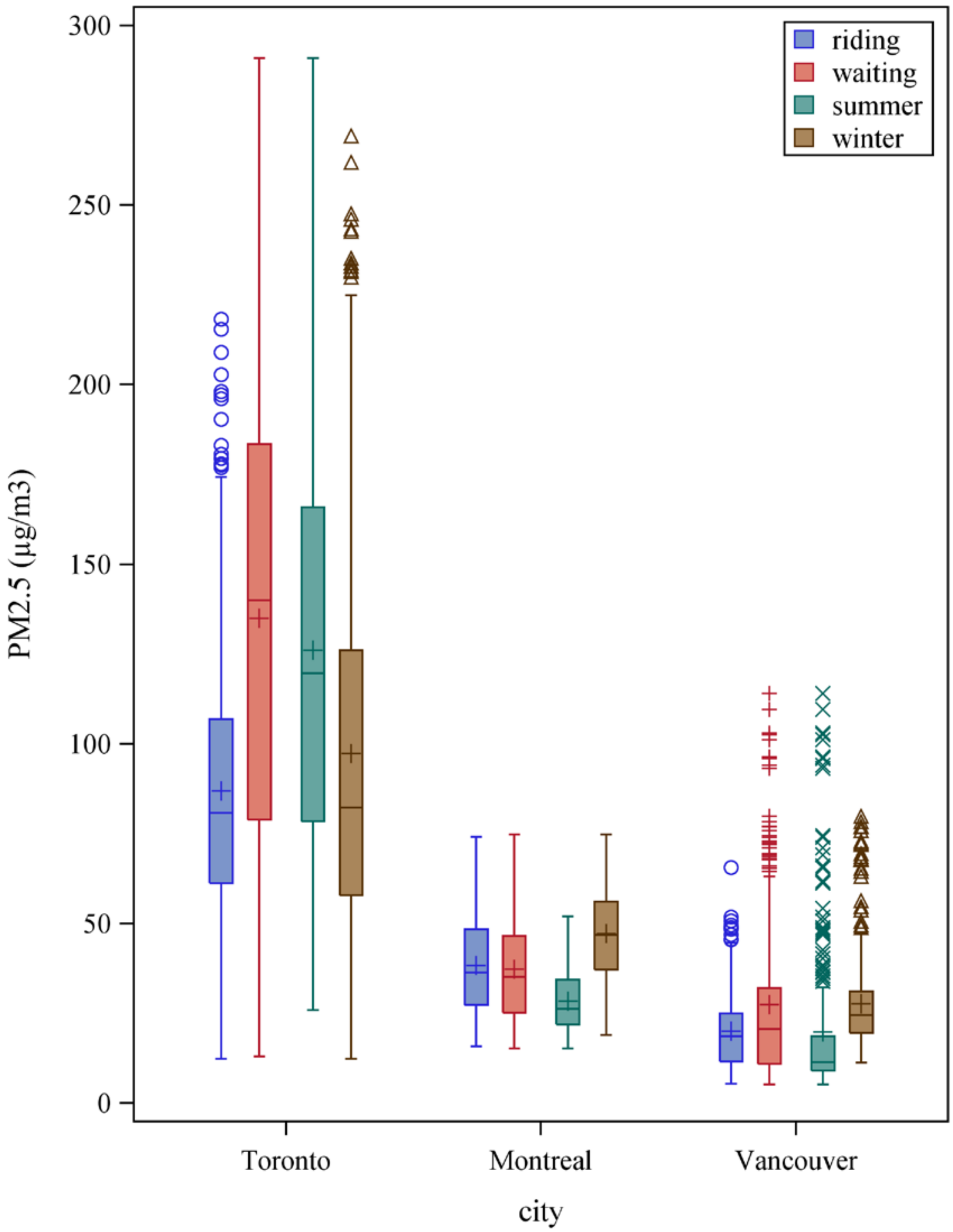

Figure S3: PM2.5 exposures sampled in Toronto, Montreal and Vancouver metros.

+ = mean, line=median, box=p25 & p75, tails=min & max (of values within 1.5IQR), emarkers=obs>1.5IQR

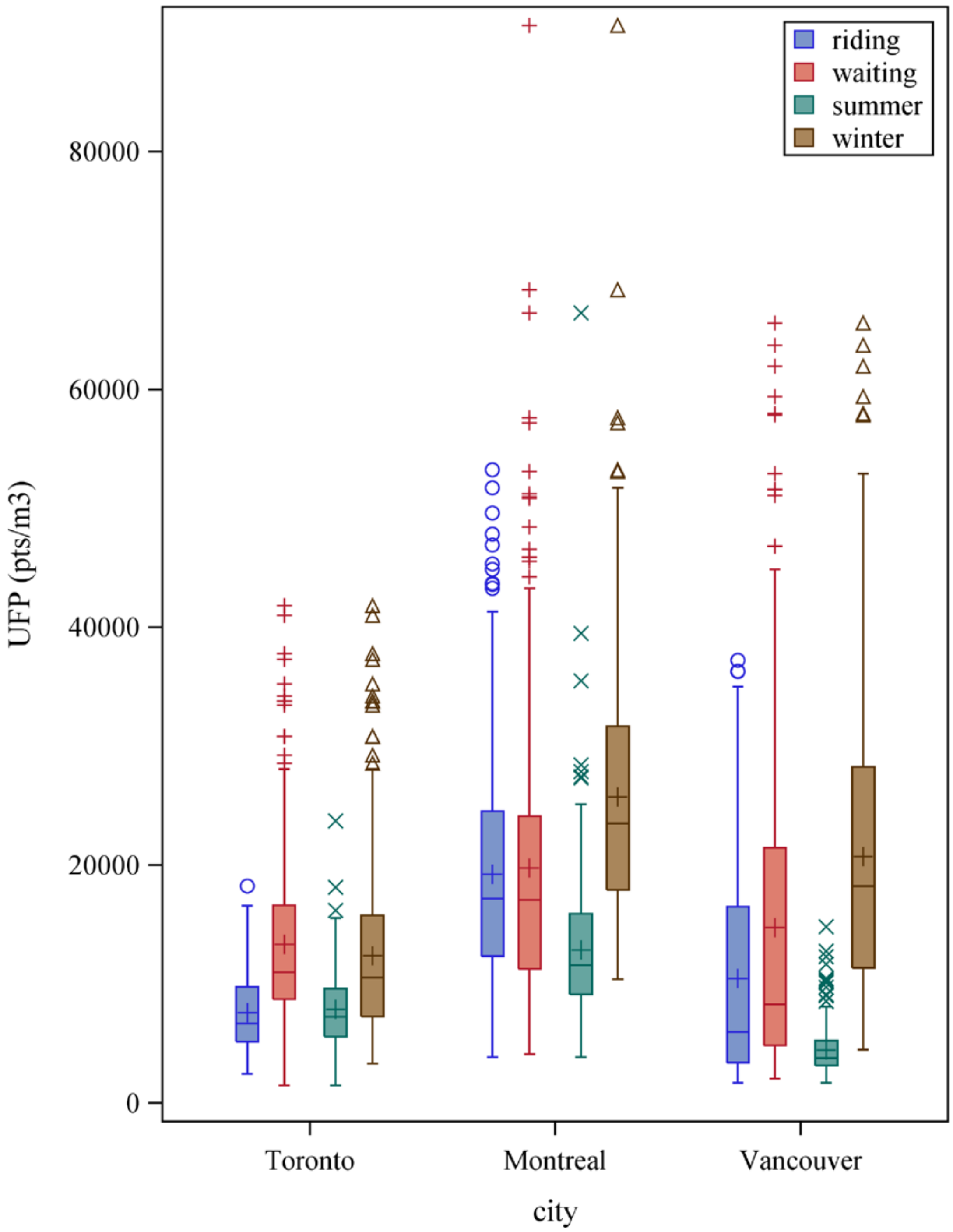

Figure S4: UFP exposures sampled in Toronto, Montreal and Vancouver metros.

+ = mean, line=median, box=p25 & p75, tails=min & max (of values within 1.5IQR), emarkers=obs>1.5IQR

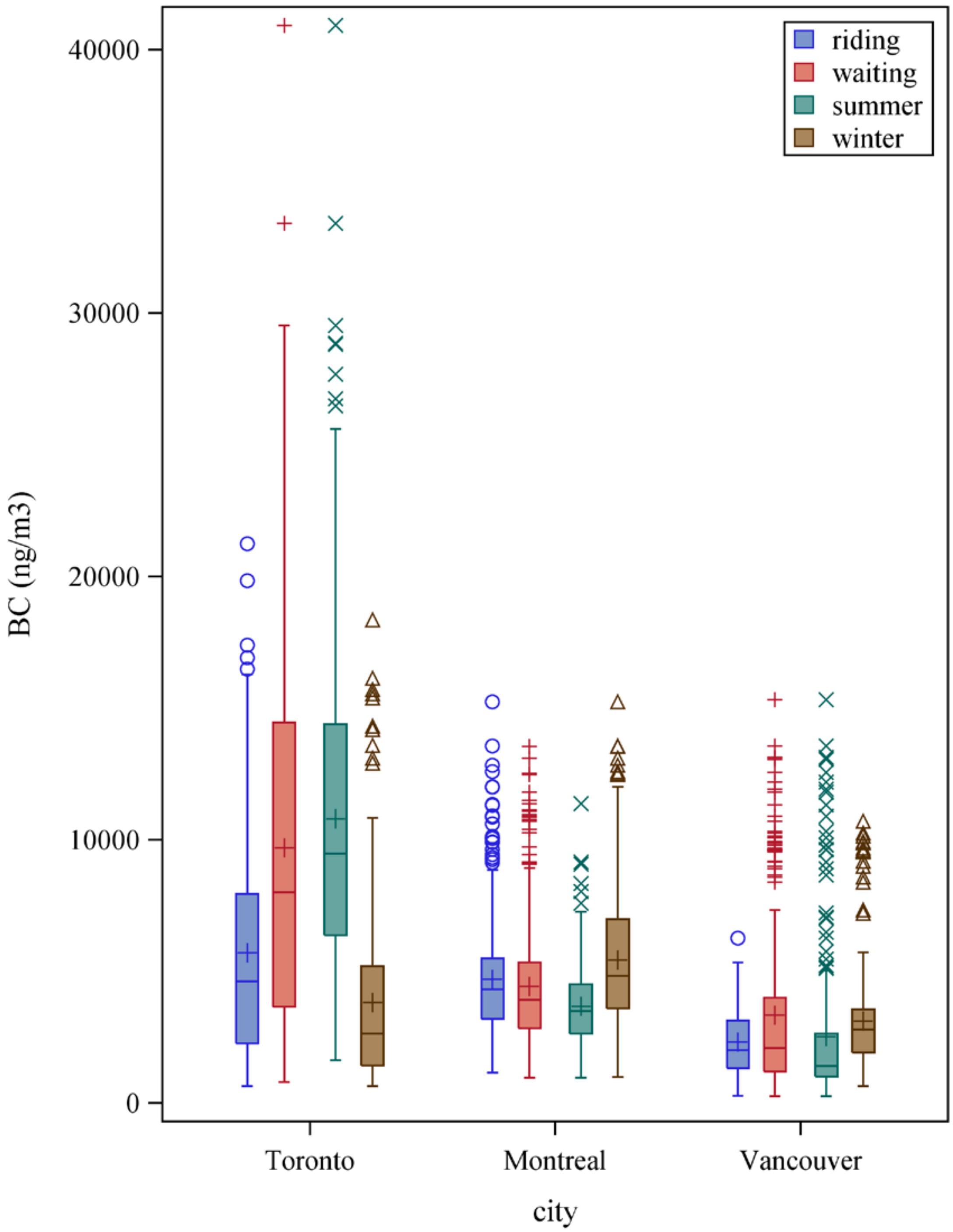

Figure S5: BC exposures sampled in Toronto, Montreal and Vancouver metros.

+ = mean, line=median, box=p25 & p75, tails=min & max (of values within 1.5IQR), emarkers=obs>1.5IQR

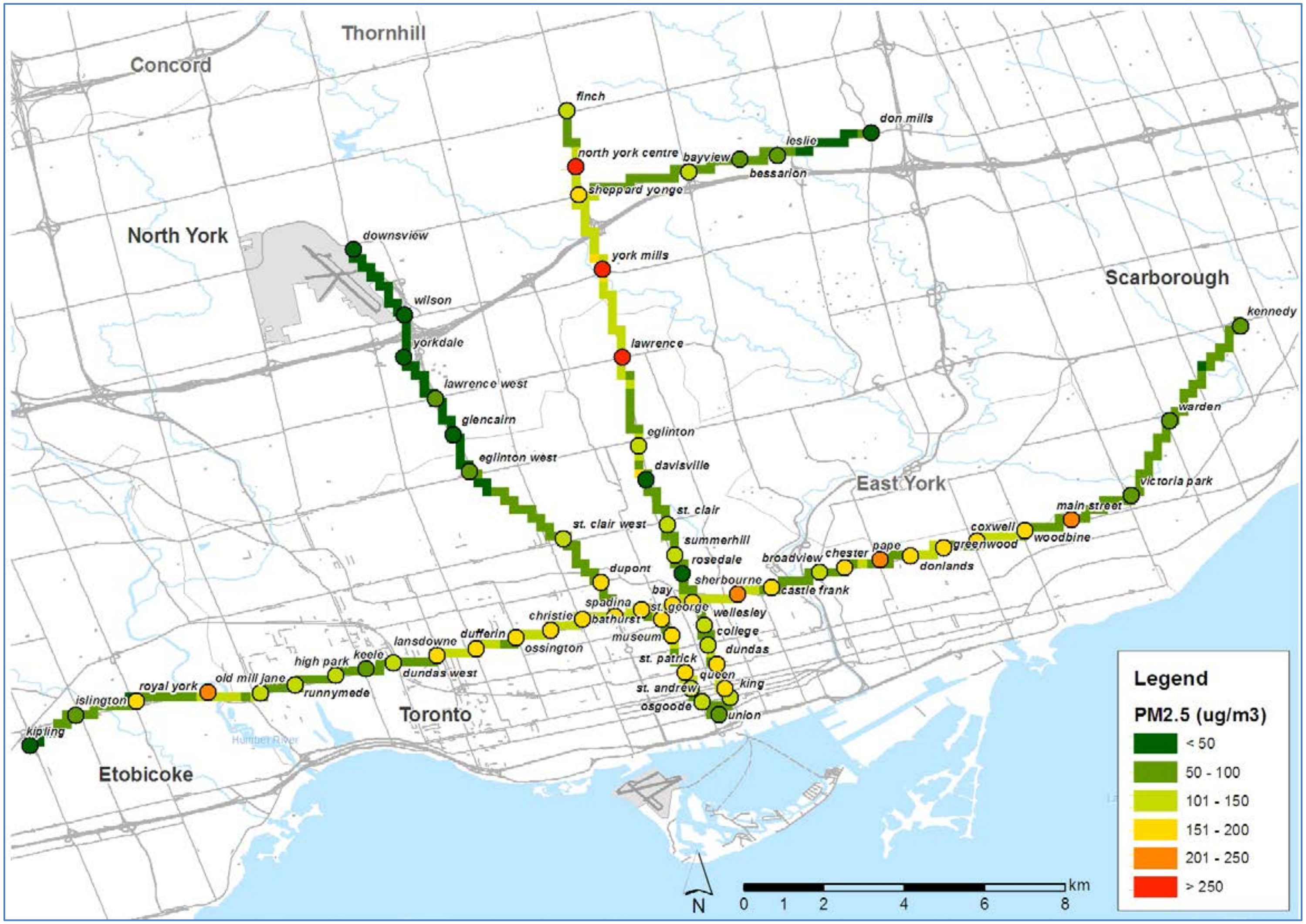

Figure S6. Spatial means of $PM_{2.5}$ across Toronto metro system

Figure S7. Spatial means of $PM_{2.5}$ across Montreal metro system

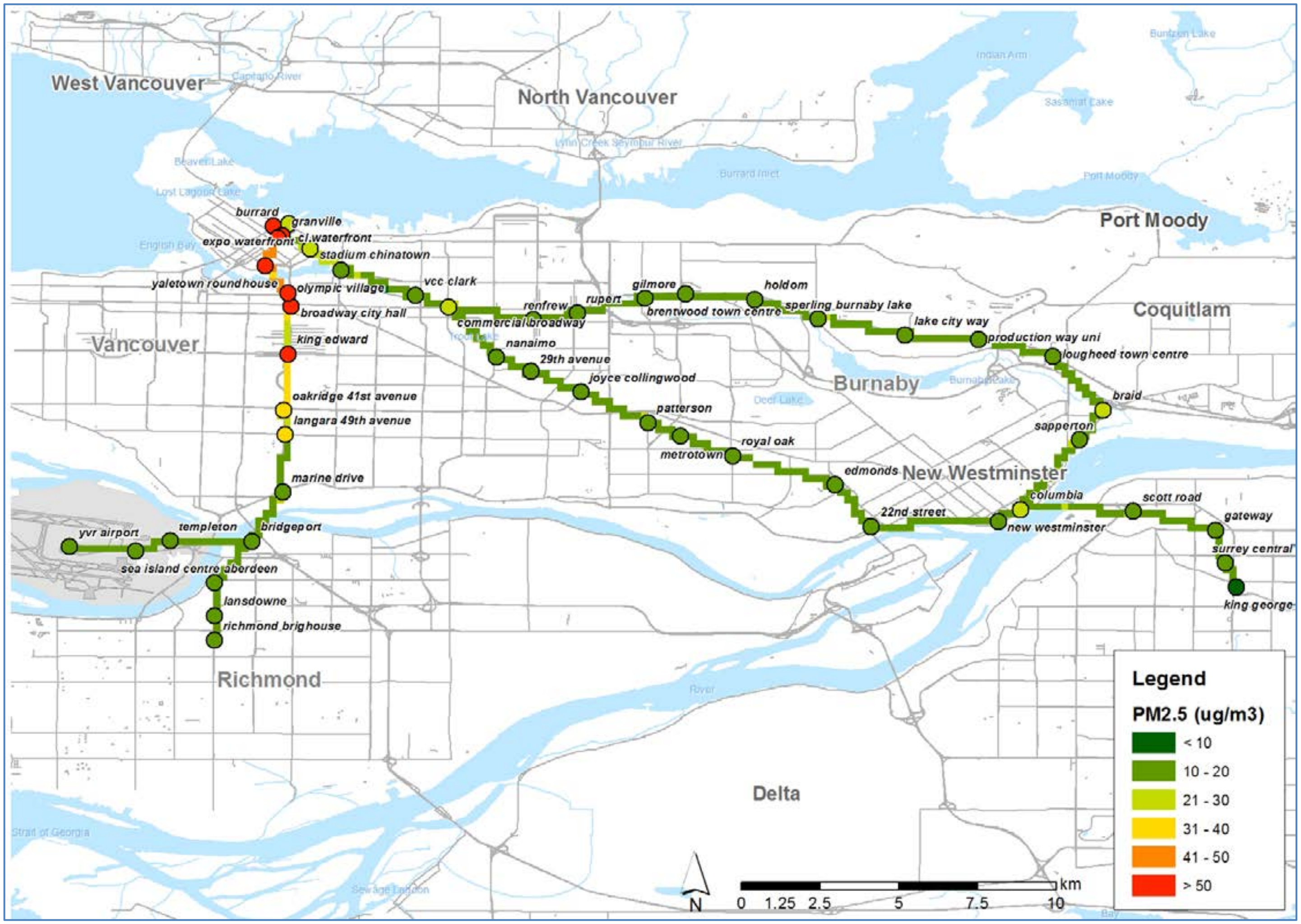

Figure S8. Spatial means of $PM_{2.5}$ across Vancouver metro system

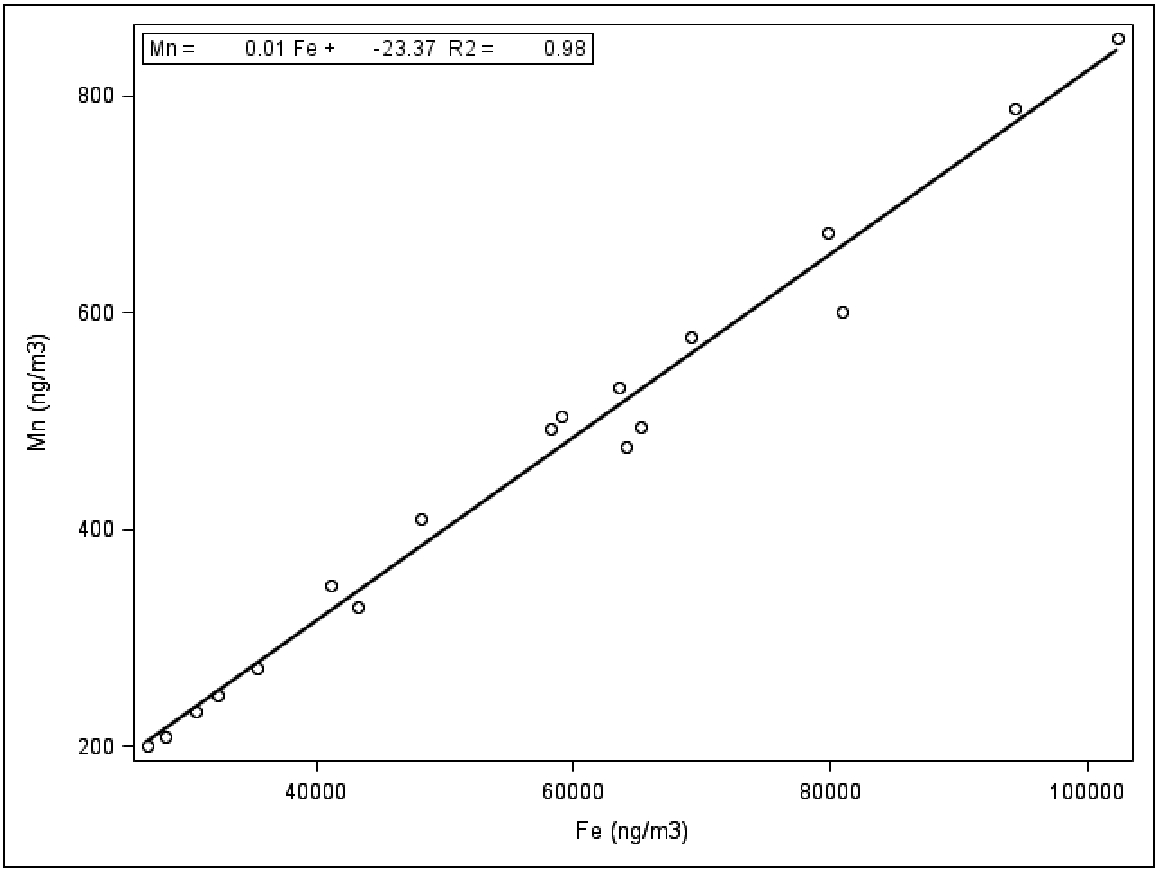

Figure S9. Correlation of Fe and Mn in Toronto metro $PM_{2.5}$ samples.

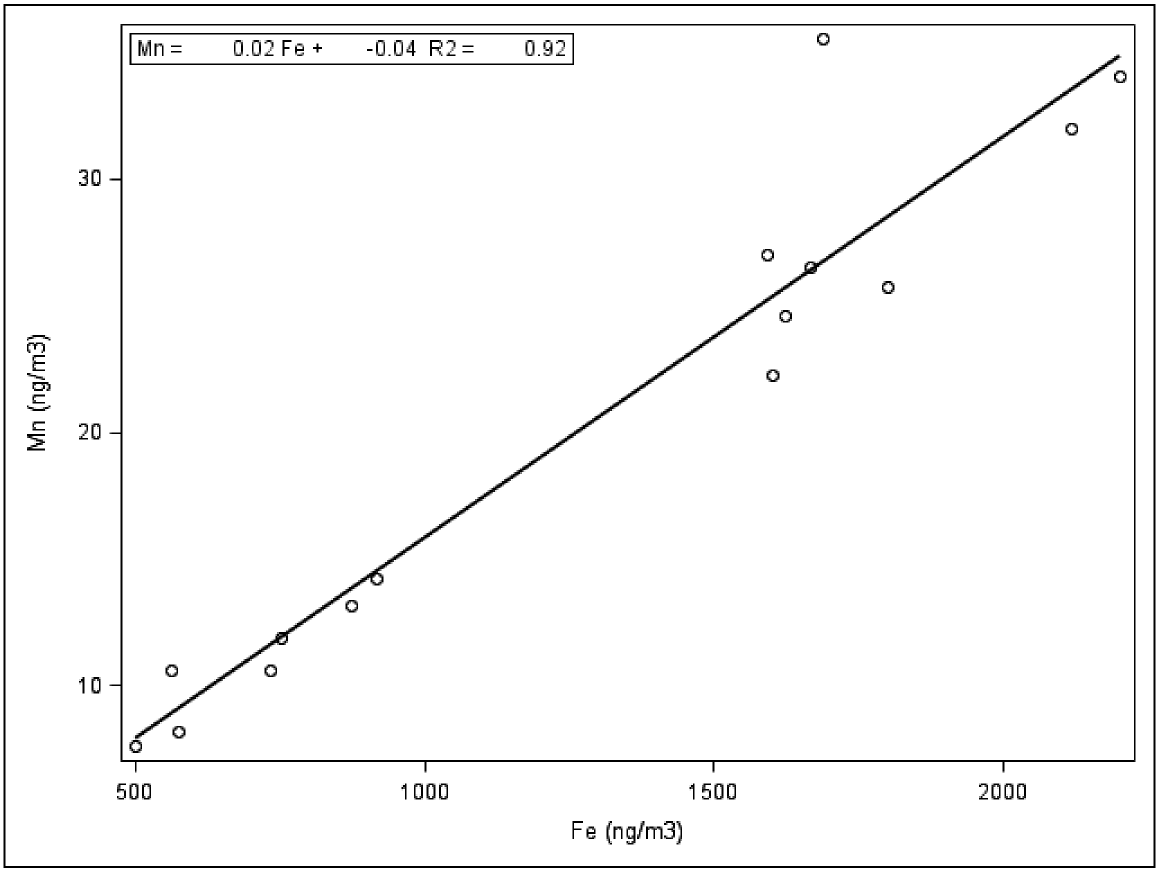

Figure S10. Correlation of Fe and Mn in Montreal metro $PM_{2.5}$ samples.

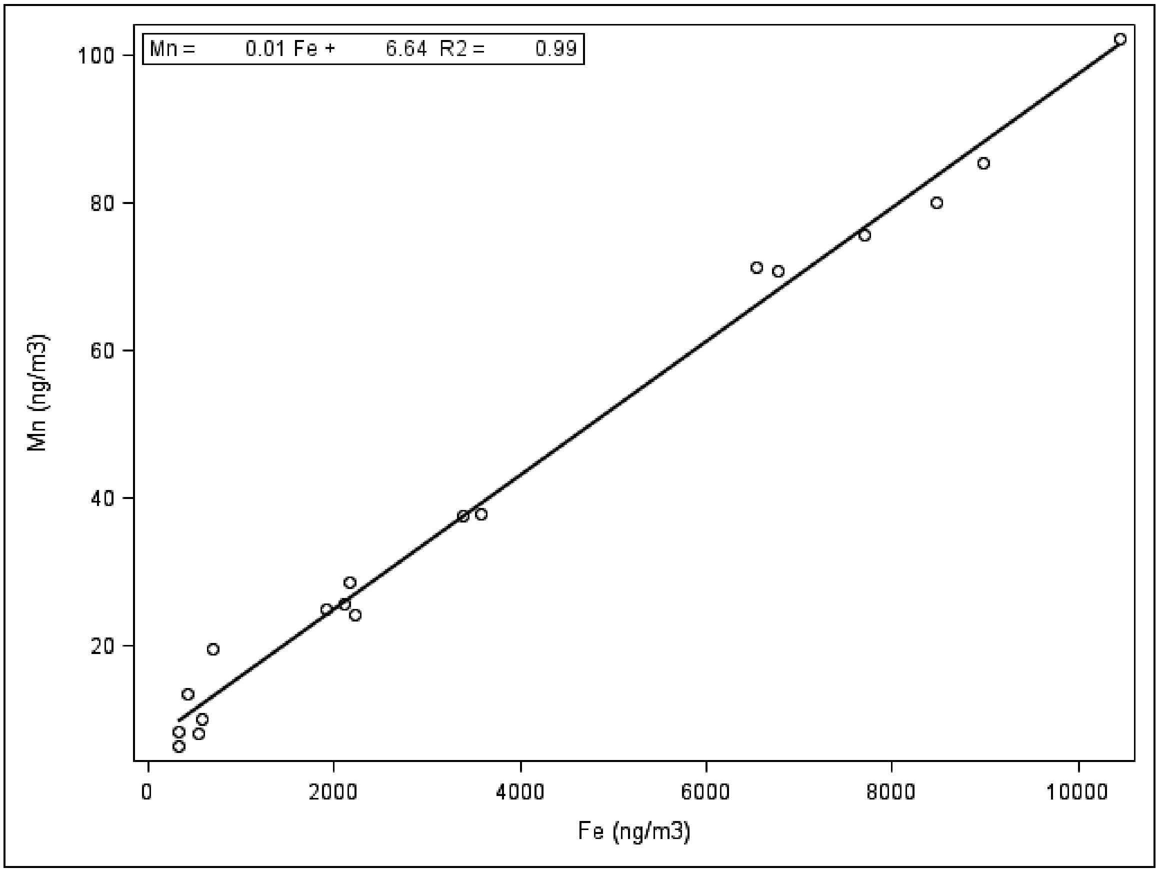

Figure S11. Correlation of Fe and Mn in Vancouver metro $PM_{2.5}$ samples.

References

1. Wallace, L. A., Wheeler, A. J., Kearney, J., Van Ryswyk, K., You, H., Kulka, R., Rasmussen, P., Brook, J., Xu, I. Validation of continuous particle monitors for personal, indoor, and outdoor exposures. *J Expos Sci Environ Epidemiol*. 2011;21(1):49-64. Accessed 12 July 2016. doi: 10.1038/jes.2010.15.