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Traffic-related air pollution near roadways: discerning local impacts from background

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S1 Data availability

The availability of pollutant data following quality assurance is displayed in Table S1, divided by site and season. The winter season is defined as containing the months of December, January, and February in full. Spring is March, April, and May; summer is June, July, and August; and lastly fall is September, October, and November.

Table S1: Percentage of valid data by site, pollutant, and season.

Pollutant	Site	2015		2016				2017	
		Summer	Fall	Winter	Spring	Summer	Fall	Winter	Spring
NO_x	NR-TOR-1	91	99	100	96	96	93	100	99
	BG-TOR-1	47	93	95	98	98	100	100	99
	NR-TOR-2	100	92	98	63	99	100	1	49
	BG-TOR-2	100	61	100	100	88	99	100	99
	NR-VAN	96	82	96	97	98	97	92	96
	BG-VAN	98	98	98	98	98	24	0	0
CO	NR-TOR-1	64	94	96	80	79	75	79	80
	BG-TOR-1	0	0	0	71	92	94	94	96
	NR-TOR-2	91	91	91	91	97	100	96	99
	BG-TOR-2	91	88	92	91	82	72	80	91
	NR-VAN	96	48	95	83	92	98	92	96
	BG-VAN	98	98	98	96	98	24	0	0
CO_2	NR-TOR-1	61	99	98	95	96	94	100	99
	BG-TOR-1	0	0	0	43	98	100	100	100
	NR-TOR-2	100	100	99	100	91	47	97	100
	BG-TOR-2	91	86	100	73	85	67	0	27
	NR-VAN	80	84	90	96	100	71	62	97
	BG-VAN	0	0	0	0	0	0	0	0
O_3	NR-TOR-1	91	97	100	92	95	91	97	92
	BG-TOR-1	47	88	95	98	98	100	99	99
	NR-TOR-2	100	94	99	100	100	100	97	99
	BG-TOR-2	96	98	79	100	95	99	100	99
	NR-VAN	96	82	96	95	98	97	91	95
	BG-VAN	98	97	97	98	98	24	0	0
$PM_{2.5}$	NR-TOR-1	89	97	100	97	96	98	100	89
	BG-TOR-1	44	93	95	100	97	100	100	99
	NR-TOR-2	97	100	99	99	100	95	95	99
	BG-TOR-2	97	97	87	100	95	99	100	99
	NR-VAN	94	83	98	99	81	98	92	97

	BG-VAN	99	99	100	100	100	24	0	0
UFP	NR-TOR-1	90	96	99	87	0	78	100	90
	BG-TOR-1	0	0	0	40	97	100	100	99
	NR-TOR-2	80	80	98	99	99	99	96	95
	BG-TOR-2	79	72	96	97	27	4	0	0
	NR-VAN	97	85	78	96	88	95	91	89
	BG-VAN	98	66	95	100	97	25	0	0
BC	NR-TOR-1	91	99	100	97	89	95	100	99
	BG-TOR-1	0	0	0	58	95	99	100	100
	NR-TOR-2	100	97	97	97	99	94	86	99
	BG-TOR-2	100	98	96	100	87	99	85	99
	NR-VAN	92	84	98	99	99	100	94	97
	BG-VAN	98	100	97	96	99	25	0	0

S2 Downwind-upwind measurements

Table S2: Average pollutant concentrations measured at the NR-TOR-1 site, aggregated based on whether an air mass originated upwind or downwind of the station, along with the downwind-upwind difference.

Pollutant	Downwind N	Downwind μ ± 95%CI	Upwind N	Upwind μ ± 95%CI	Δ (Downwind – Upwind)
NO [ppb]	2378	37.8 ± 1.1	1787	2.9 ± 0.3	34.9
NO_2 [ppb]	2303	21.2 ± 0.4	1748	10.7 ± 0.4	10.5
CO [ppb]	2015	364.4 ± 5.4	1577	226.6 ± 3.2	137.8
CO_2 [ppm]	2305	437.3 ± 1.0	1763	416.4 ± 1.1	20.9
O_3 [ppb]	2313	15.3 ± 0.4	1771	33.2 ± 0.8	-17.9
$PM_{2.5}$ [μg m^{-3}]	2377	7.68 ± 0.21	1801	9.01 ± 0.27	-1.33
UFP [cm^{-3}]	1839	56975 ± 1671	1313	15305 ± 513	41670
BC [μg m^{-3}]	2338	2.13 ± 0.06	1775	0.73 ± 0.03	1.40

Table S3: Average pollutant concentrations measured at the NR-TOR-2 site, aggregated based on whether an air mass originated from upwind or downwind of the station, along with the downwind-upwind difference.

Pollutant	Downwind N	Downwind μ ± 95%CI	Upwind N	Upwind μ ± 95%CI	Δ (Downwind – Upwind)
NO [ppb]	1970	6.0 ± 0.2	5242	3.2 ± 0.1	2.8
NO_2 [ppb]	1671	8.5 ± 0.2	4210	10.4 ± 0.2	-1.9
CO [ppb]	1990	247.9 ± 3.6	5165	246.8 ± 1.9	1.1
CO_2 [ppm]	1938	423.1 ± 0.7	4994	421.4 ± 0.5	1.7
O_3 [ppb]	2090	24.2 ± 0.3	5439	28.7 ± 0.3	-4.5
$PM_{2.5}$ [μg m^{-3}]	2036	3.80 ± 0.12	5435	9.01 ± 0.15	-5.21
UFP [cm^{-3}]	1974	12878 ± 398	5087	16676 ± 220	-3798
BC [μg m^{-3}]	2059	0.63 ± 0.02	5299	0.81 ± 0.02	-0.18

Table S4: Average pollutant concentrations measured at the NR-VAN site, aggregated based on whether an air mass originated from upwind or downwind of the station, along with the downwind-upwind difference.

Pollutant	Downwind N	Downwind μ ± 95%CI	Upwind N	Upwind μ ± 95%CI	Δ (Downwind – Upwind)
NO [ppb]	2472	56.6 ± 2.5	1887	9.7 ± 0.7	46.8
NO_2 [ppb]	2475	21.9 ± 0.4	1890	11.5 ± 0.3	10.4
CO [ppb]	2222	414.3 ± 12.8	1615	210.1 ± 4.5	204.2
CO_2 [ppm]	2338	461.6 ± 3.3	1829	414.5 ± 1.2	47.1
O_3 [ppb]	2454	9.4 ± 0.4	1861	19.7 ± 0.5	-10.3
$PM_{2.5}$ [μg m^{-3}]	2460	8.81 ± 0.26	1742	5.57 ± 0.19	3.23
UFP [cm^{-3}]	2314	29960 ± 776	1784	14060 ± 381	15900
BC [μg m^{-3}]	2547	2.48 ± 0.07	1909	0.84 ± 0.04	1.64

S2.1 Site meteorology and downwind/upwind diurnal patterns

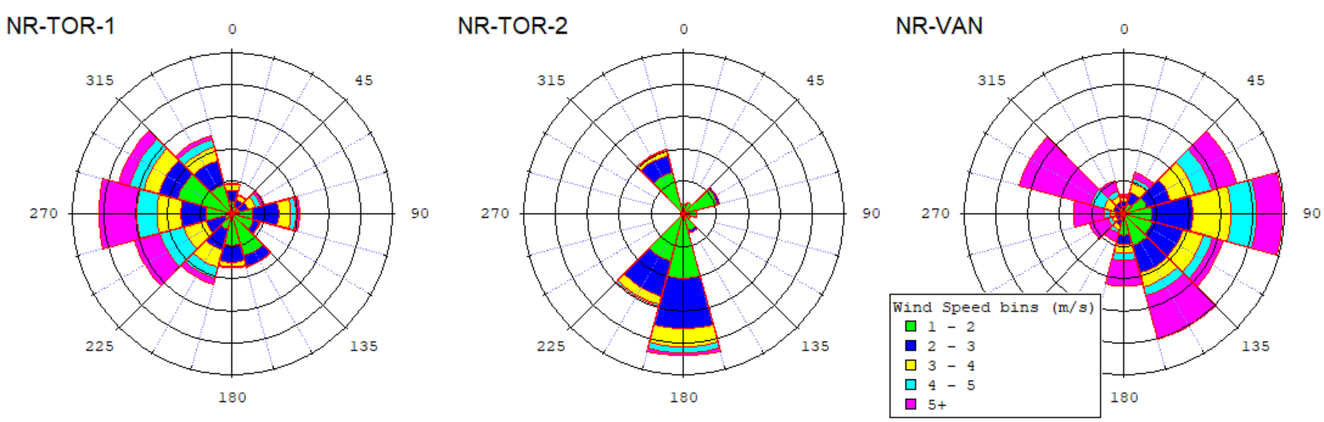


Figure S1: Frequency of wind measurements made at each near-road site: NR-TOR-1 (left), NR-TOR-2 (middle), and NR-VAN (right) throughout the two year measurement campaign.

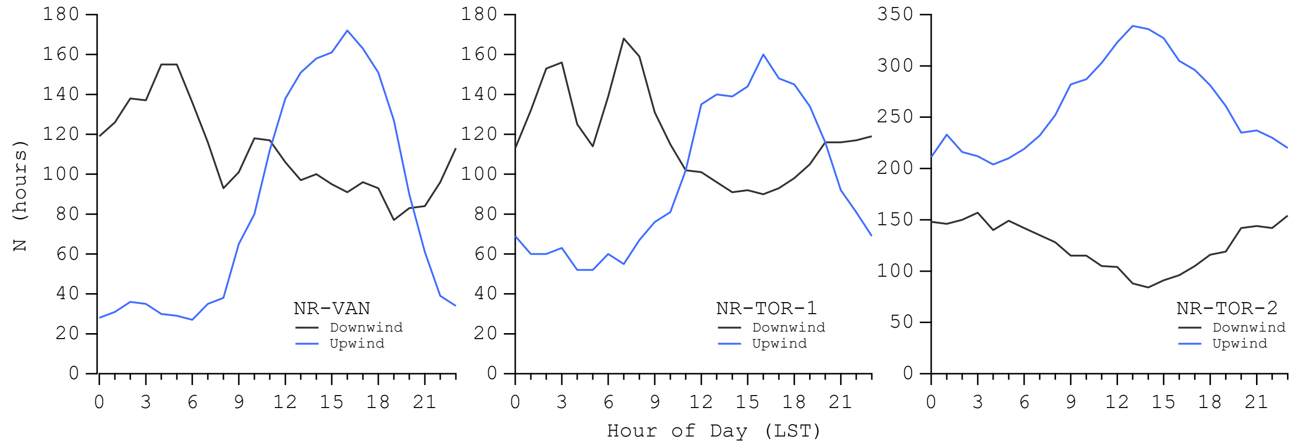


Figure S2: Frequency of hourly measurements originating from downwind and upwind of the major roadways upon which NR-VAN (left), NR-TOR-1(middle), and NR-TOR-2(right) are stationed.

As can be seen in Figure S2, each near-road site exhibited non-uniform diurnal patterns in frequency of downwind and upwind samples, meaning $C_{L,2}$ may be biased by these diurnal effects. Tables S5 and S6 show the differences between using all collected data for $C_{L,2}$, and what the average local concentration would be if downwind/upwind sampling was uniform (i.e. $C_{L,2,uniform}$). These uniform values were calculated by randomly sampling 'N' values per hour of day, where N is based on the diurnal minima in Figure S2. This process was repeated 100 times for each pollutant at NR-TOR-1 (Table S5) and NR-VAN (Table S6), and the average downwind and upwind values from this are reported as $DW_{uniform}$ and $UW_{uniform}$, respectively.

Table S5: Downwind and upwind pollutant averages at NR-TOR-1. $DW_{uniform}$ and $UW_{uniform}$ denote downwind and upwind pollutant averages using an equivalent number of samples from each hour of day so as to not be biased by diurnal effects.

Pollutant	DW	$DW_{uniform}$	UW	$UW_{uniform}$	$C_{L,2}$	$C_{L,2,uniform}$	% diff
NO [ppb]	37.8	37.3	2.9	2.9	34.9	34.4	1
NO_2 [ppb]	21.2	20.9	10.7	12.1	10.5	8.8	16
CO [ppb]	364.4	361.1	222.6	230.4	141.8	130.7	8
CO_2 [ppm]	437.3	436.8	416.4	420.8	20.9	16	23
O_3 [ppb]	15.3	15.5	33.2	28.3	-17.9	-12.8	28
$PM_{2.5}$ [µg m^{-3}]	7.68	7.6	9.01	9.37	-1.33	-1.77	-33
UFP [cm^{-3}	57000	56400	15300	14600	41700	41800	0
BC [µg m^{-3}]	2.13	2.11	0.73	0.71	1.4	1.4	0

Table S6: Downwind and upwind pollutant averages at NR-VAN. $DW_{uniform}$ and $UW_{uniform}$ denote downwind and upwind pollutant averages using an equivalent number of samples from each hour of day so as to not be biased by diurnal effects.

Pollutant	DW	$DW_{uniform}$	UW	$UW_{uniform}$	$C_{L,2}$	$C_{L,2,uniform}$	% diff
NO [ppb]	56.6	56	9.7	11.5	46.9	44.5	5
NO_2 [ppb]	21.9	22.2	11.5	11.9	10.4	10.3	1
CO [ppb]	414.3	416.7	210.1	216.1	204.2	200.6	2
CO_2 [ppm]	461.6	459.6	414.5	417	47.1	42.6	10
O_3 [ppb]	9.4	9.9	19.7	17.1	-10.3	-7.2	30
$PM_{2.5}$ [μg m^{-3}]	8.81	8.87	5.57	5.42	3.24	3.45	-6
UFP [cm^{-3}	30000	30800	14000	13500	16000	17300	-8
BC [μg m^{-3}]	2.48	2.11	0.84	0.71	1.64	1.4	15

S3 Implications for using downwind-upwind analysis for estimating local TRAP concentrations

For the stations positioned on flat terrain (NR-VAN and NR-TOR-1), the average difference between downwind and upwind pollutant concentrations, Method 2, has yielded larger local concentrations for all pollutants (with the exception of $PM_{2.5}$) when compared with methods 1 and 3. Recall that Method 1 generates local concentrations, $C_{L,1}$ via:

$$\overline{C}_{L,1} = \overline{C}_{NR} - \overline{C}_{BG} \ , \qquad (1)$$

where C_{NR} and C_{BG} are concentrations explicitly measured at near-road and background locations, respectively. Whereas Method 2 determines local concentrations, $C_{L,2}$, from:

$$\overline{C}_{L,2} = \overline{C}_{DW} - \overline{C}_{UW} \ , \qquad (2)$$

where C_{DW} and C_{UW} are pollutant concentrations measured when air masses are originating downwind and upwind from the roadway at a near-road receptor, respectively. Presumably, average concentrations measured at near-road locations during upwind conditions are similar to those at nearby background locations, as neither receptor is impacted significantly by local sources during these times. Given this, the average difference between local concentrations generated using methods 1 and 2 is approximated with the following equality:

$$\overline{C}_{UW} \approx \overline{C}_{BG} \ \Rightarrow \ \overline{C}_{L,2} - \overline{C}_{L,1} \approx \overline{C}_{DW} - \overline{C}_{NR} \ , \qquad (3)$$

The above equalities state, in other words, that if average upwind concentrations at a near-road location are roughly equivalent to average background concentrations, then the difference between local TRAP concentrations inferred through methods 2 and 1 should be similar to the difference between average downwind and total near-road concentrations.

Firstly, to test the assumption $\overline{C}_{UW} \approx \overline{C}_{BG}$, these concentrations were calculated at NR-VAN, BG-VAN, NR-TOR-1, and BG-TOR-1 and are reported in Table S7.

Table S7: Average upwind concentrations at NR-VAN and NR-TOR-1, compared with average pollutant concentrations measured at BG-VAN and BG-TOR-1.

Pollutant	$\bar{C}_{UW}$ NR-VAN	$\bar{C}_{BG}$ BG-VAN	$\bar{C}_{UW}$ NR-TOR-1	$\bar{C}_{BG}$ BG-TOR-1
NO [ppb]	9.7	9.2	2.9	3.5
NO_2 [ppb]	11.5	14.2	10.7	10.8
CO [ppb]	210.1	228.9	226.6	210.6
CO_2 [ppm]	414.5		416.4	420.3
O_3 [ppb]	19.7	15.9	33.2	24.7
$PM_{2.5}$ [μg m^{-3}]	5.57	5.41	9.01	7.86
UFP [cm^{-3}]	14060	12880	15305	11968
BC [μg m^{-3}]	0.84	0.66	0.73	0.58

The differences in background pollutant quantities measured through these two methods agree fairly well with one another, with maximum differences of ~20%. Hence, the assumption that these two average quantities are approximately equivalent appears to be valid. The differences in Table S7 are not large enough to explain the differences observed between methods 1 and 2 in Tables 2-4. Table S8 shows the differences between $\bar{C}_{DW}$ and $\bar{C}_{NR}$ at NR-VAN and NR-TOR-1, as well as differences between methods 2 and 1 at these sites, and the similarities are evident. Therefore, the aforementioned equality in Eq. (3) appears valid. Furthermore, Method 2 appears to over-predict average local concentrations by factors of ~1.7 and ~1.4 (neglecting $PM_{2.5}$) at NR-VAN and NR-TOR-1, respectively.

Table S8: Average near-roar road and downwind concentrations at NR-VAN and NR-TOR-1, along with differences between these two average quantities, and differences between average local quantities inferred through methods 2 and 1.

Pollutant	NR-VAN					NR-TOR-1				
	$\bar{C}_{NR}$	$\bar{C}_{DW}$	$\bar{C}_{DW} - \bar{C}_{NR}$	$\bar{C}_{L,2} - \bar{C}_{L,1}$	$\bar{C}_{L,2}/\bar{C}_{L,1}$	$\bar{C}_{NR}$	$\bar{C}_{DW}$	$\bar{C}_{DW} - \bar{C}_{NR}$	$\bar{C}_{L,2} - \bar{C}_{L,1}$	$\bar{C}_{L,2}/\bar{C}_{L,1}$
NO [ppb]	36.9	56.6	19.7	23.8	2.0	24.6	37.8	13.2	13.4	1.6
NO_2 [ppb]	21.5	21.9	0.4	5.3	2.0	19.3	21.2	1.9	1.8	1.2
CO [ppb]	349.7	414.3	64.6	108.5	2.1	328.4	364.4	36.0	34.7	1.3
CO_2 [ppm]	439.8	461.6	21.8	-	-	436.8	437.3	0.5	6.5	1.5
$PM_{2.5}$ [μg m^{-3}]	7.79	8.81	1.02	0.97	1.4	9.39	7.68	-1.71	-2.82	-0.9
UFP [cm^{-3}]	27570	29956	2386	4334	1.4	39987	56975	16988	12065	1.4
BC [μg m^{-3}]	1.88	2.48	0.60	0.46	1.4	1.68	2.13	0.45	0.37	1.4

S4. Sensitivity of method 3 to window duration

The choice of input parameters α and W play a large role on the magnitude of average local concentrations determined using method 3. Here a sensitivity analysis shows the range of average local concentrations observed for each pollutant and near-road site when W is varied between 6 [hr] and 14 [hr]. The smoothing parameter, α, is constrained at α = 4 for the purposes of comparison.

Table S9: Average $C_{L,3}$ values by site and pollutant for W = 6 and W = 14 [hr]. The smoothing parameter, α, is set to 4.

Pollutant	NR-VAN		NR-TOR-1		NR-TOR-2	
	W = 6	W = 14	W = 6	W = 14	W = 6	W = 14
NO [ppb]	24.0	30.9	15.4	20.8	3.4	4.2
NO_2 [ppb]	8.0	11.4	7.4	11.1	4.5	6.3
CO [ppb]	132.3	172.7	95.7	132.9	57.4	81.0
CO_2 [ppm]	31.4	47.7	16.6	22.7	11.1	15.6
O_3 [ppb]	-8.4	-13.3	-9.7	-15.7	-7.1	-11.4
$PM_{2.5}$ [μg m^{-3}]	3.34	4.59	3.52	5.18	2.33	3.63
UFP [cm^{-3}]	13057	17265	18843	26520	6031	8251
BC [μg m^{-3}]	1.09	1.41	0.84	1.15	0.35	0.48

S5 Regression of near-road data with respect to wind speed

Table S10: Regression parameters for the wind-speed dependence of each TRAP measured at the near-road sites.

Pollutant	NR-VAN		NR-TOR-1	
	c1	c2	c1	c2
NO	2.56	0.83	1.56	0.51
NO_2	1.62	0.40	1.50	0.46
CO	2.53	0.81	1.54	0.50
CO_2	2.36	0.76	2.05	0.88
UFP	1.58	0.37	1.01	0.01
BC	1.76	0.47	1.62	0.56
Average Values	2.02	0.59	1.55	0.49

S5.1 Regression differentiated by weekday and weekend

The mechanics of dispersion should be unaffected by day of week, and thus local pollutant concentrations should exhibit similar wind speed relationships between weekdays and weekends. One reason why dispersion in the near-road environment would inherently differ between weekdays and weekends is the greater traffic densities seen on weekdays may result in greater vehicular-induced turbulence. Figure S1 shows the relationship between normalized local concentrations and wind speed at NR-VAN and NR-TOR-1; it is important to note here that the concentrations are normalized with respect to a mean calculated for all days. Thus, this relationship will differ in the sense that lower local concentrations were seen on weekends.

Table S11: Regression parameters for the wind-speed dependence of each TRAP measured at the near-road sites separated by weekdays and weekends.

Pollutant	NR-VAN				NR-TOR-1			
	c1		c2		c1		c2	
	Weekdays	Weekends	Weekdays	Weekends	Weekdays	Weekends	Weekdays	Weekends
NO	2.74	2.08	0.74	1.17	1.81	0.78	0.46	0.68
NO2	1.69	1.40	0.33	0.57	1.56	1.33	0.37	0.80
CO	2.68	2.12	0.79	0.85	1.60	1.36	0.51	0.46
CO2	2.35	2.36	0.68	0.98	2.15	1.73	0.84	1.00
UFP	1.77	1.04	0.35	0.41	1.11	0.72	-0.03	0.15
BC	1.95	1.20	0.41	0.72	1.86	0.86	0.49	0.77
Average Values	2.20	1.70	0.55	0.79	1.68	1.01	0.44	0.64

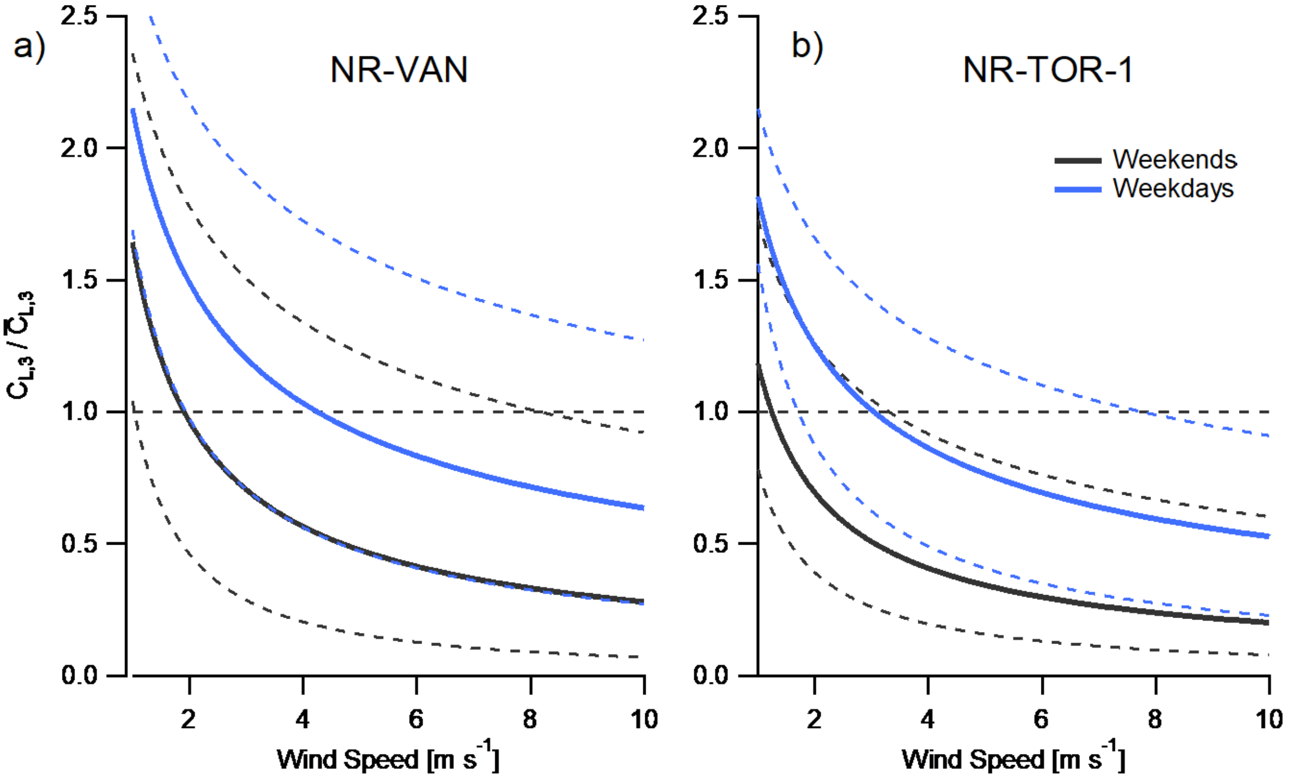


Figure S3: Normalized local pollutant concentrations determined using method 3 with respect to wind speed at NR-VAN (a) and NR-TOR-1 (b). Solid blue lines indicate the average trend amongst all TRAPs on weekdays and solid black lines on weekends. Dashed lines indicate the range of variability between pollutants.

S6 Fraction of pollution attributable to local and background sources

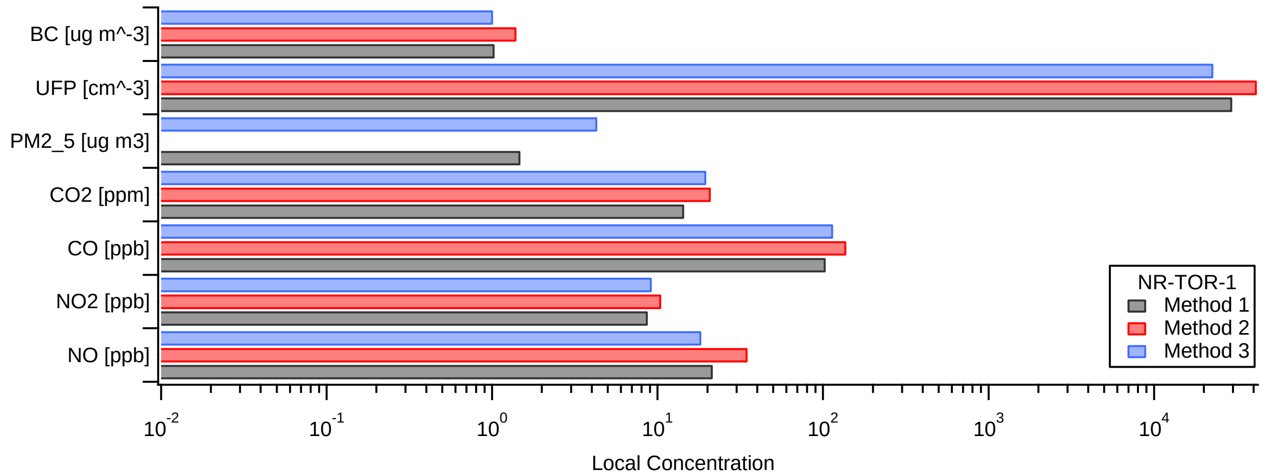


Figure S4: Local pollutant concentrations determined using each method at NR-TOR-1.

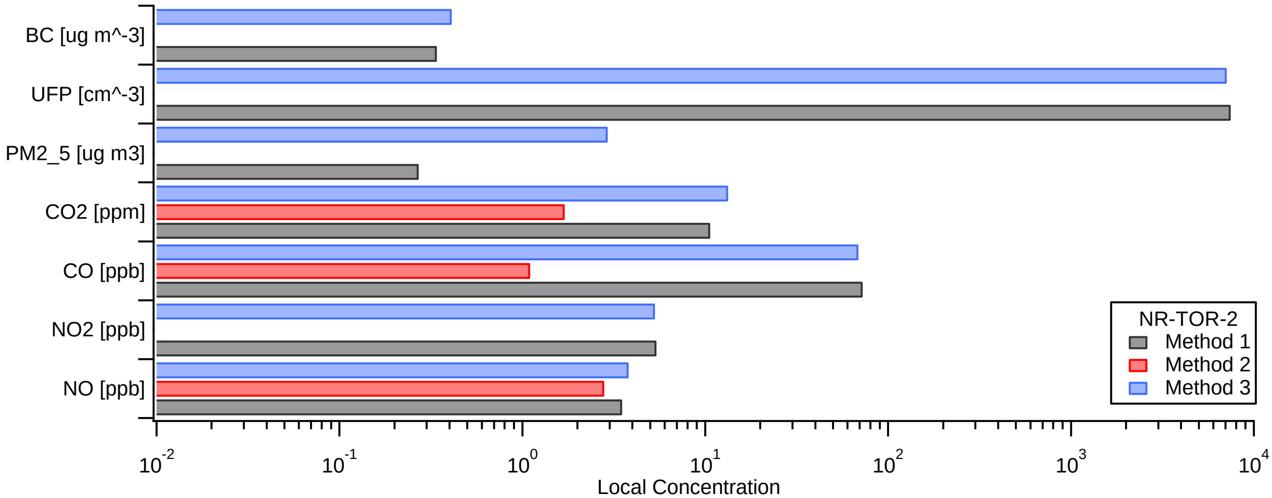


Figure S5: Local pollutant concentrations determined using each method at NR-TOR-2.

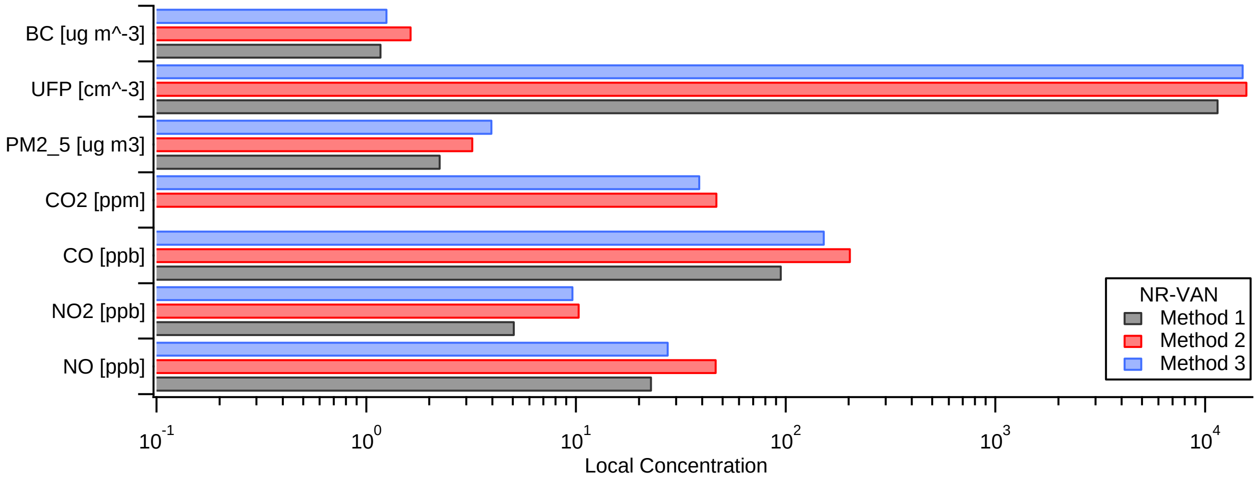


Figure S6: Local pollutant concentrations determined using each method at NR-VAN.

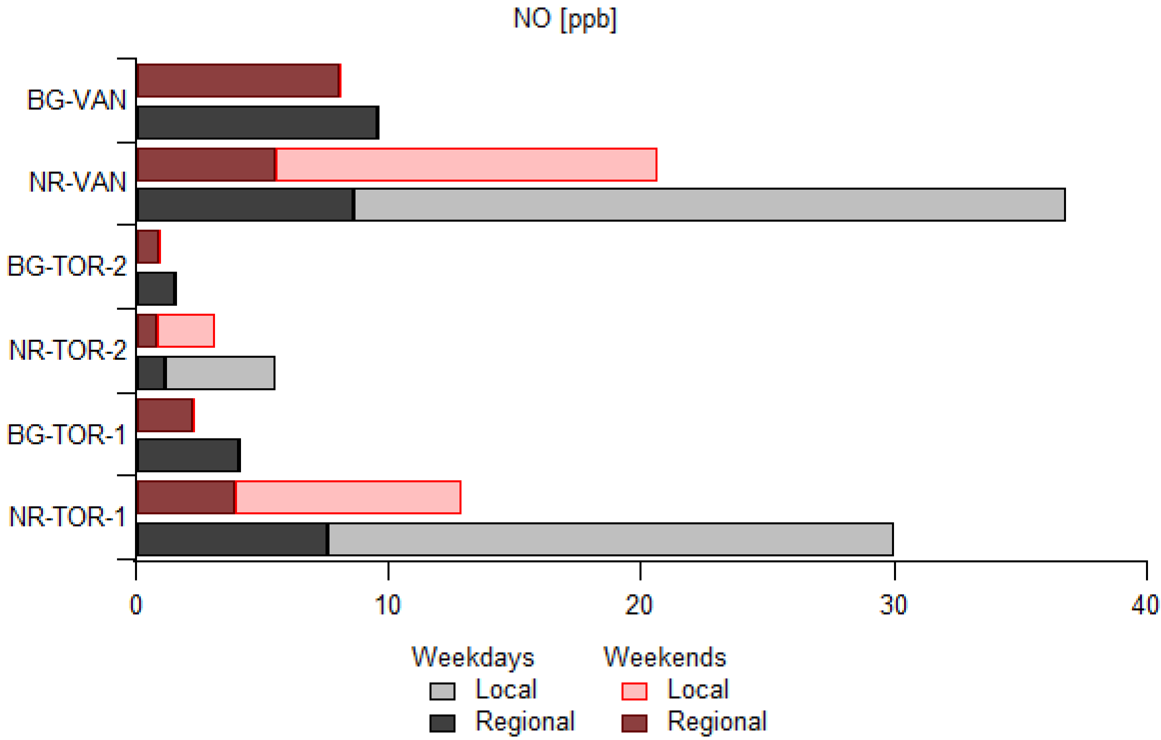


Figure S7: Nitric oxide concentrations measured at each monitoring location in this study. Each site is separated by weekday and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources. Background stations are presumed fully regional and therefore contain no local component.

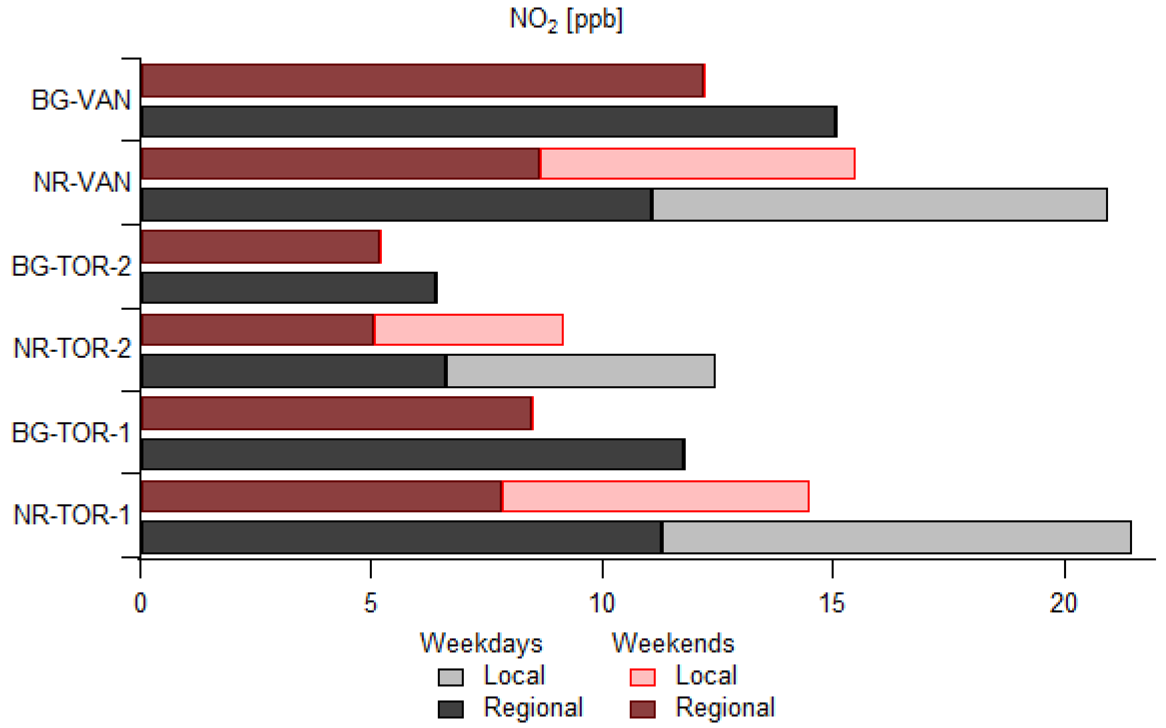


Figure S8: Nitrogen dioxide concentrations measured at each monitoring location in this study. Each site is separated by weekday and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources. Background stations are presumed fully regional and therefore contain no local component.

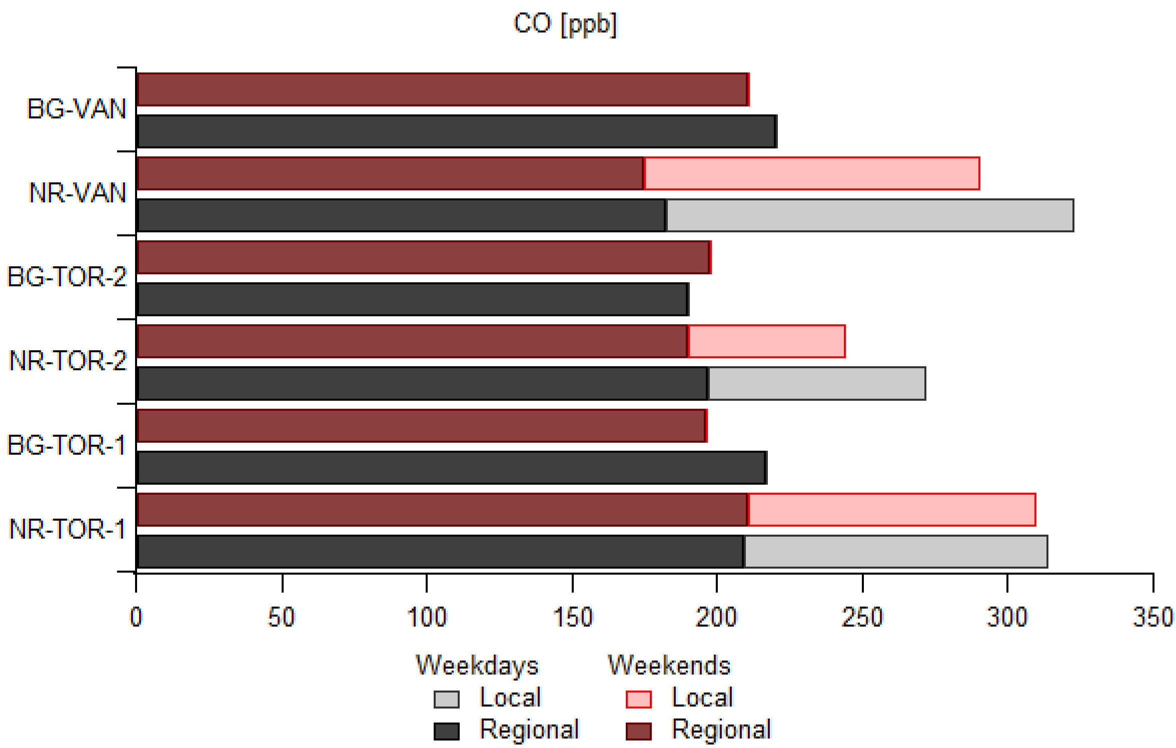


Figure S9: Carbon monoxide concentrations measured at each monitoring location in this study. Each site is separated by weekday and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources. Background stations are presumed fully regional and therefore contain no local component.

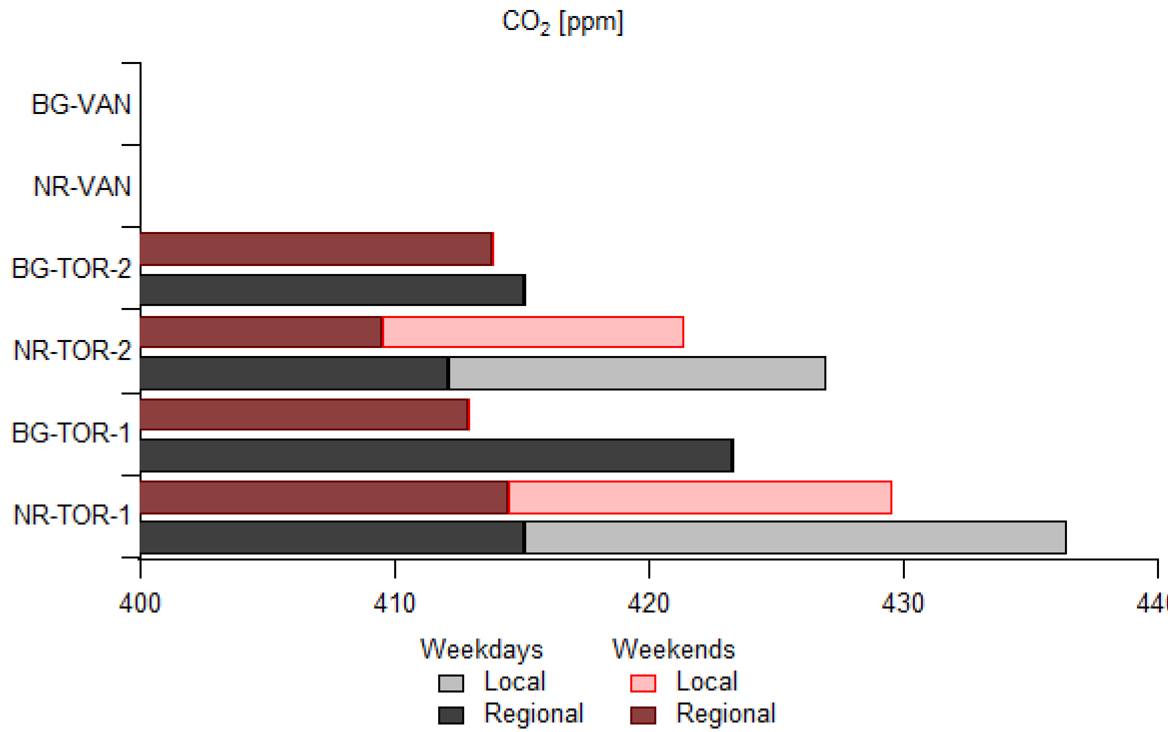


Figure S10: Carbon dioxide concentrations measured at each monitoring location in this study. Each site is separated by weekday and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources. Background stations are presumed fully regional and therefore contain no local component. Carbon dioxide data was not measured at BG-VAN, and so data from NR-VAN are omitted for clarity.

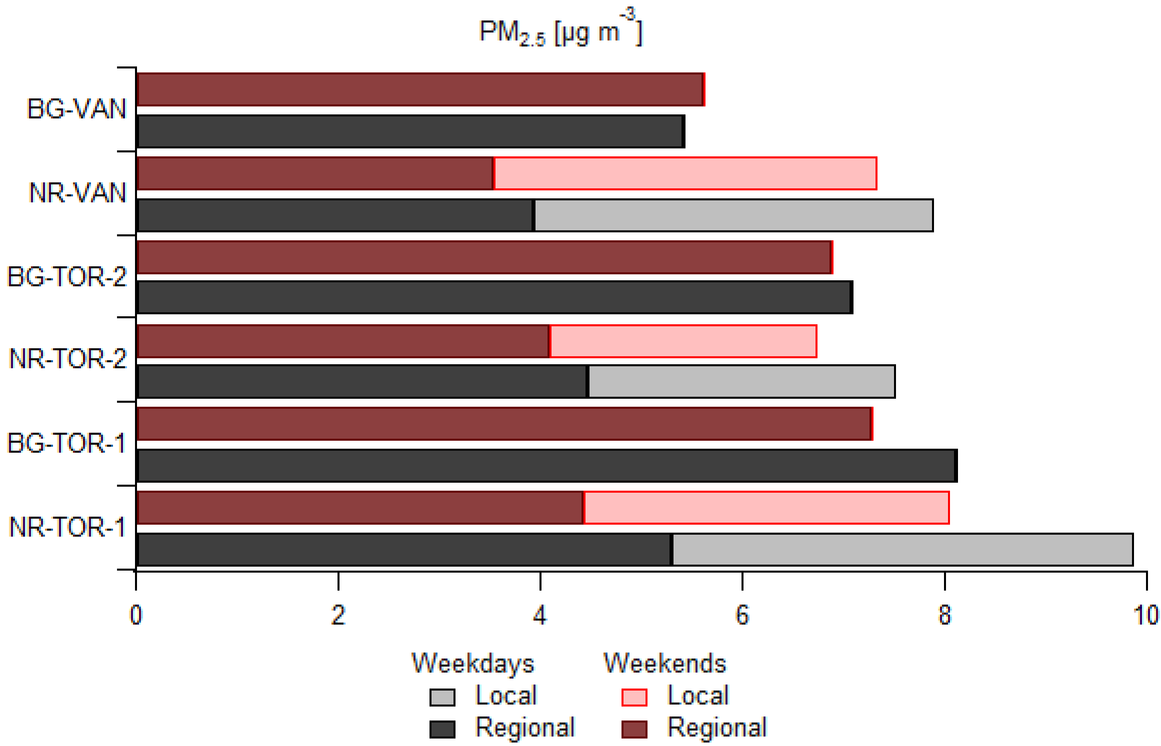
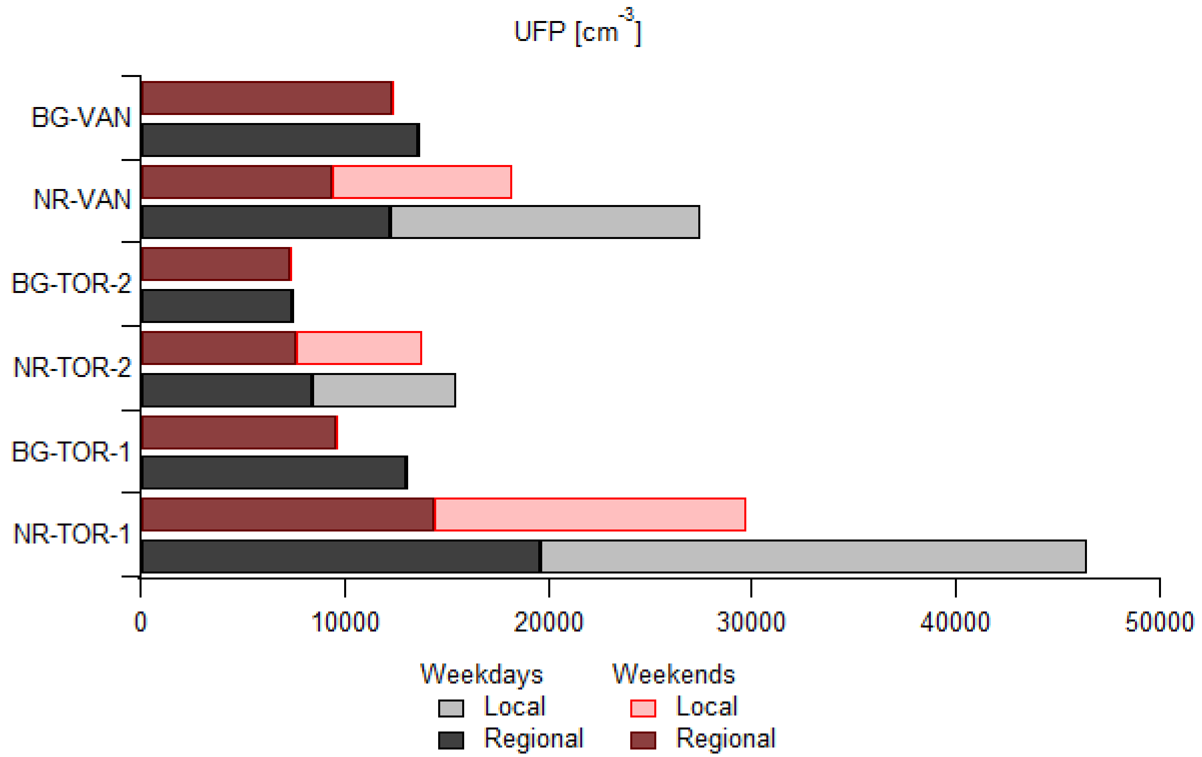


Figure S11: $PM_{2.5}$ concentrations measured at each monitoring location in this study. Each site is separated by weekday and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources. Background stations are presumed fully regional and therefore contain no local component. Large differences between regional contributions estimated at near-road stations and average concentrations at respective background stations is likely a reflection upon the poor performance of this methodology when applied to $PM_{2.5}$—local components appear to be largely overestimated, and so this method is not recommended for near-road particulate matter.



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317 **Figure S12: Ultrafine particle concentrations measured at each monitoring location in this study. Each site is separated by weekday**
318 **and weekend, and bars at near-road sites are stacked according to concentrations attributed to local and regional sources.**
319 **Background stations are presumed fully regional and therefore contain no local component.**

320