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In-Vehicle Exposures to Particulate Air Pollution in Canadian Metropolitan Areas: The Urban Transportation Exposure Study

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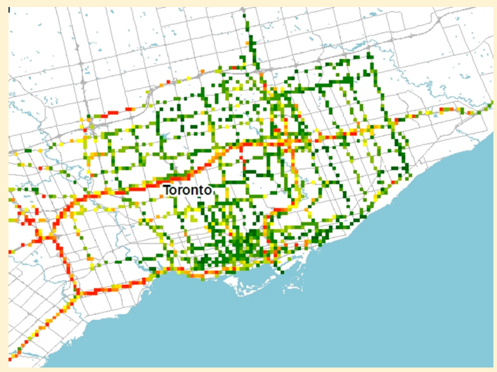
Supporting Information

ABSTRACT: Commuters may be exposed to increased levels of traffic-related air pollution owing to close proximity to traffic-emissions. We collected in-vehicle and roof-top air pollution measurements over 238 commutes in Montreal, Toronto, and Vancouver, Canada between 2010 and 2013. Voice recordings were used to collect real-time information on traffic density and the presence of diesel vehicles and multivariable linear regression models were used to estimate the impact of these factors on in-vehicle pollutant concentrations (and indoor/outdoor ratios) along with parameters for road type, land use, and meteorology. In-vehicle $PM_{2.5}$ and NO_2 concentrations consistently exceeded regional outdoor levels and each unit increase in the rate of encountering diesel vehicles (count/min) was associated with substantial increases (>100%) in in-vehicle concentrations of ultrafine particles (UFPs), black carbon, and $PM_{2.5}$ as well as strong increases (>15%) in indoor/outdoor ratios. A model based on meteorology and the length of highway roads within a 500 m buffer explained 53% of the variation in in-vehicle UFPs; however, models for $PM_{2.5}$ (R^2 = 0.24) and black carbon (R^2 = 0.30) did not perform as well. Our findings suggest that vehicle commuters experience increased exposure to air pollutants and that traffic characteristics, land use, road types, and meteorology are important determinants of these exposures.



■ INTRODUCTION

Traffic-related air pollution is known to contribute to cardiorespiratory morbidity.[1−7] In urban areas, traffic is a major source of ambient air pollution and may represent an important source of exposure for commuters owing to their close proximity to traffic emissions. Indeed, for some pollutants such as ultrafine particles (UFPs) (≤0.1 μm) and black carbon, exposures during daily commutes may represent a large portion of overall daily exposure levels despite relatively short time-periods spent in commuting environments.[8−11] Moreover, evidence from several short-term panel studies suggests that in-vehicle exposures may contribute to increased systemic inflammation, pulmonary inflammation, oxidative stress, and changes in cardiac autonomic modulation.[12−18] As a result, there is currently a need to understand determinants of these exposures in order to evaluate their potential health impacts in large-scale population-based studies.

The Urban Transportation Exposure Study was designed to characterize commuter exposures to traffic-related air pollutants in Canadian metropolitan areas including particulate air pollutants such as UFPs, black carbon, and fine particulate matter air pollution below a median aerodynamic diameter of 2.5 μm ($PM_{2.5}$), as well as nitrogen dioxide (NO_2) and volatile organic compounds (VOCs). In addition, models were developed to estimate the potential impacts of traffic characteristics, road types, land use, and meteorological factors on in-vehicle particulate air pollutant concentrations (and indoor/outdoor ratios) along various routes in these regions. This is the first national study of in-vehicle commuter exposures in Canada and to our knowledge is the first to use land use characteristics to predict in-vehicle concentrations along a given route.

■ MATERIALS AND METHODS

Study Design. The Urban Transportation Exposure Study (UTES) was conducted between 2010 and 2013 in Canada's three largest cities: Toronto, Ontario, Montreal, Quebec, and Vancouver, British Columbia. In Toronto, sampling was conducted for 2 weeks in September, 2010 (summer) and 1 week in March, 2011 (winter). Montreal monitoring was limited to 1 week in March, 2011 (winter) whereas monitoring in Vancouver was conducted for 2 weeks in May−June, 2013 (summer) and 2 weeks in December, 2012 (winter). All monitoring was conducted on weekdays. In all three cities, in-vehicle and roof-top monitoring of traffic-related air pollutants

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Table 1. Descriptive Data for In-Vehicle Air Pollution Concentrations during 3 h Commutes[a]

location	total n	total median	total range	summer n	summer median	summer range	winter n	winter median	winter range
Toronto									
UFP (count/cm^3)	59	27 869	9502−133 629	32	28 589	14 321−54 774	27	25 504	9502−133 629
BC (ηg/m^3)	67	1404	337−6897	46	1938	337−6897	21	1050	526−1932
$PM_{2.5}$ (μg/m^3)	71	8.65	4.07−56.2	44	11.4	4.07−56.2	27	6.60	4.68−15.9
NO_2 (ppb)	83	49.5	6.7−153	53	51.3	6.7−104	30	49.3	23.7−153
CO (ppm)	74	3.94	0.802−16.8	44	3.85	1.73−10.0	30	4.03	0.802−16.8
BTEX (μg/m^3)	49	12.6	4.85−34.9	38	14.1	5.69−34.9	11	8.74	4.85−10.4
Vancouver									
UFP (count/cm^3)	114	24 401	2778−66 750	57	20 599	2778−57 922	57	30 857	9629−66 750
BC (ηg/m^3)	113	1995	159−7279	55	1810	159−5673	58	2009	200−7279
$PM_{2.5}$ (μg/m^3)	110	5.31	1.37−10.8	56	4.99	1.88−10.8	54	6.01	1.37−10.7
NO_2 (ppb)	119	25.9	1.2−196	59	14.6	1.2−97.0	57	28.3	4.3−196
CO (ppm)	52	3.03	2.16−16.5	52	3.03	2.16−16.5			
BTEX (μg/m^3)	103	10.8	2.81−146	54	10.9	5.17−146	49	10.6	2.81−68.1
Montreal									
UFP (count/cm^3)							23	29 650	8913−98 744
BC (ηg/m^3)							14	1526	398−2688
$PM_{2.5}$ (μg/m^3)							24	13.6	4.28−33.3
NO_2 (ppb)							29	45.5	8.4−117
CO (ppm)							26	6.28	0.031−28.1
BTEX (μg/m^3)							23	16.4	7.83−37.09
Overall									
UFP (count/cm^3)	196	26,321	2778−133 629	89	23,339	2778−57 922	107	29,650	8913−133 629
BC (ηg/m^3)	194	1779	159−7279	101	1874	159−6897	93	1779	159−7278
$PM_{2.5}$ (μg/m^3)	205	6.54	1.37−56.2	100	6.66	1.88−56.2	105	6.54	1.37−33.3
NO_2 (ppb)	231	39.0	1.2−196	112	35.5	1.2−104	116	41.5	4.3−196
CO (ppm)	152	3.86	0.031−28.1	96	3.45	1.73−16.5	56	3.86	0.031−28.1
BTEX (μg/m^3)	175	11.9	2.81−146	92	12.6	5.17−146	83	11.9	2.81−146

[a]n, Number of commutes.

took place twice each day during the morning (7:00−10:00) and evening (15:00−18:00) rush hour periods. Three separate vehicles (described below) were used to monitor air pollution concentrations during each route with each vehicle focusing on specific portions of the city: downtown areas, major highways, and suburban areas. Dedicated routes were not assigned; instead, drivers focused on maximizing coverage of these three specific regions during each sampling period. Moreover, drivers took a different path along their route each day in order to avoid encountering the same regions at the same time during each commute. Driving was not canceled due to rain or snow and roadways were not snow covered during winter monitoring. In general we experienced little precipitation during monitoring, although it did rain periodically in Vancouver during winter monitoring, which is typical of this time of year. Air pollution concentrations in public transit buses and light rail transit were also examined as part of UTES but these results will be reported separately.

Air Pollution Monitoring. All in-vehicle and roof-top monitoring was conducted using Chevrolet Grand Caravans (model year 2009−2012) with the windows closed and heating/cooling settings set to suit driver comfort. There were no discernible differences in vehicle size or cabin volume between model years. Recirculation settings were kept in the off position at all times and fan speeds were set to medium. Traffic-related air pollutants were simultaneously monitored both inside and outside vehicles during each route. In-vehicle monitors were positioned in the passenger seat with samples collected at breathing level. Outdoor measurements were collected using instruments mounted in cases strapped to roof racks with sampling tubes facing toward the rear of the vehicle. Real-time data were collected for UFPs (<0.1 μm) (TSI CPC model 3007), $PM_{2.5}$ (TSI Dustrak 8520), CO (Langan T15N CO monitors), temperature/relative humidity (HOBO data loggers), and black carbon (AethLabs MicroAeth-51) at 1 s sampling intervals and averaged over the duration of each route. Outdoor $PM_{2.5}$ samplers were not equipped with isokinetic inlets; however, indoor/outdoor ratios for $PM_{2.5}$ were not correlated with vehicle speed (R^2 = 0.02) suggesting that this did not have a dramatic impact on our results (Figure S1 Supporting Information (SI)). Weekly average gravimetric samples were also collected for $PM_{2.5}$ concentrations (for metals analysis to be presented separately) and the linear relationship between gravimetric and Dustrak measurements (Gravimetric $PM_{2.5}$ = 1.1 + 0.52(Dustrak $PM_{2.5}$); R^2 = 0.37) (Figure S2 SI) was used to convert Dustrak data to gravimetric equivalents to obtain descriptive data for each route (Table 1). Integrated samples for NO_2 and VOCs (reported as the sum of benzene, toluene, ethyl-benzene, and xylenes (BTEX)) were collected using Ogawa passive sampling badges and SUMMA canisters (6 L), respectively. NO_2 samples were analyzed using ion chromatography and VOC samples were analyzed using GC-MS as previously described.[38,39] Each vehicle carried a GlobalSat DG-100 monitor to log geographic coordinates for every second of each route. Photographs of the sampling setup are available in the SI (Figure S10)

Regional outdoor data for $PM_{2.5}$ and NO_2 were collected from Environment Canada's National Air Pollution Surveillance network in each city for the 3 h duration of each commute. Mean regional values were determined for each commuting period based on hourly data collected in each city. Specifically, 6 regional monitoring stations were available for Toronto and 13 stations were available in Montreal and Vancouver. Direct statistical comparisons were not made between regional outdoor data and in-vehicle concentrations owing to differences in measurement methods as well as the fact that regional data were not available for other air pollutants.

Digital Voice Recordings. Digital voice recordings were used to collect real-time information on traffic density and the presence of diesel vehicles encountered along each route. Specifically, observations were made concerning the occupied lane and each neighboring same-direction lane in the immediate vicinity of the vehicle. Drivers used an ordinal scale (0−4) to rank traffic-density throughout each commute with 0 indicating no cars in the referenced area and 4 indicating complete occupancy of the referenced area. Counts of diesel vehicles were also recorded and updated whenever conditions of the preceding recording changed. This information was subsequently matched to real-time air pollution data at 1 s resolution using the time stamp on audio files. Large trucks, construction equipment, school buses, or other vehicles clearly labeled as diesel vehicles were included in diesel counts. These counts were averaged over the duration of each trip to estimate the rate of encountering diesel vehicles along a given route.

Land Use and Road Network Data. Road network and land use data were obtained from the DMTI CanMap Content Suite (DMTI Spatial, Markham, ON). DMTI road network and land use data were available in a vector format which was then converted to a raster format with a resolution of 5 × 5 m. A raster based method was used to calculate predictors for each GPS point as raster based analysis is often preferable when the number of points is large.

Roadways were labeled as local roads (i.e., neighborhood roads) or highway roads (including expressways, principal highways and secondary highways) and land use categories included commercial, government/institutional, open areas, park land, residential, industrial, and water. An intermediate class of roadways (i.e., major roads) was also considered but was ultimately excluded from analyses as this measure was strongly correlated with government/institutional land use (r = 0.73) and was not correlated with in-vehicle particulate air pollution concentrations (0.01< r <0.11). Candidate predictor variables for road network and land use data were generated by first converting road way and land use classifications to a raster format at a 5 × 5 m resolution. Vehicle GPS data were then used to estimate mean values for land use and road types encountered along each route using ArcGIS (ESRI, Redlands, California). Specifically, median vehicle position was determined for each minute of every commute and the length of highway/local roads and land use area at each point was calculated for circular buffers of 500, 750, and 1000 m using the focal statistics function in ArcGIS 10.0 (ESRI, Redlands, California). These values were then averaged over the duration of each commute to arrive at mean values for each 3 h trip.

Statistical Analysis. Our objective in conducting statistical analysis was to first evaluate single predictor models to evaluate the potential impact of candidate predictors on in-vehicle air pollution concentrations. Next, multivariable models were constructed focusing on predictors that could be easily measured in a population-based study of air pollution health effects. Given the hierarchical structure of the data (i.e., air pollution measurements nested within cities), we examined within and between-city standard deviations in air pollutant concentrations to evaluate potential clustering of air pollution measurements within cities. Next, hierarchical linear regression models with random intercepts for city were used to estimate the impact of each candidate predictor (described below) on log-transformed air pollution concentrations adjusting for continuous measures of mean outdoor temperature and wind speed which are known to influence in-vehicle air pollution concentrations.[23,28,29] In these analyses, within-city standard deviations in air pollution concentrations were greater than between-city standard deviations and hierarchical models suggested that between-city variance was small compared to within-city variance (data not shown). As a result, standard multivariable linear regression models were selected for the main analyses as little evidence of within-city clustering of air pollution concentrations was apparent in the data. Regression modeling was limited to particulate air pollutants (UFPs, black carbon, and $PM_{2.5}$) as these were the primary pollutants of interest. All air pollution data for in-vehicle concentrations were log-transformed to improve normality of model residuals. The general structure of multivariable regression models used in this study is presented below where α is the constant term, ε is the random error term, and $\beta_{1\text{-}i}$ are slopes values for predictors $X_{1\text{-}i}$.

$$\ln(\text{pollutant concentration}) = \alpha + \beta_1 X_1 + \beta_2 X_2 + \ldots\ldots + \beta_i X_i + \varepsilon$$

A number of parameters were evaluated as potential determinants of in-vehicle air pollution concentrations and indoor/outdoor ratios (calculated by dividing indoor concentrations by outdoor concentrations). These parameters included mean traffic density (on a scale of 0−4) and diesel vehicle counts (mean count/min) obtained through digital voice recordings as well as continuous variables for the land use and road network data described above. Vehicle speed was also considered but was ultimately excluded owing to strong correlations with highway road length (r = 0.74) and local road length (r = −0.78). In addition, information on vehicle speed would be difficult to collect in large population based studies whereas data for highway road length and local road length are readily available. Each determinant was first evaluated separately in multivariable linear regression models to estimate its impact on in-vehicle air pollution concentrations (and indoor/outdoor ratios) adjusted for continuous measures of mean outdoor temperature and wind speed. A continuous variable for relative humidity and an indicator variable for city were also explored but were not included in multivariable models as their inclusion had little impact (<10% change) on model coefficients for traffic and land use variables.

Covariates for final multivariable models were limited to parameters that could conceivably be measured for participants in a population-based study interested in capturing air pollution exposures during daily commutes; this limited candidate predictors in final models to land use parameters, road network data, and meteorological factors. In the context of a population-based study, land use and road network data could be estimated based on origin-destination data collected through a questionnaire. Variables for final multivariable models were selected using approximate Bayes factors calculated using the BMS package[19] in R (version 2.15.3) with uniform model priors (which assumes that all models are equally likely a priori) and noninformative "unit information" priors for model coefficients.

Table 2. Indoor/Outdoor Ratios for Air Pollution Concentrations during 3 h Commutes[a]

location	total			summer			winter		
	n	median	range	n	median	range	n	median	range
Toronto									
UFP (count/cm^3)	51	0.74	0.24−1.3	27	0.88	0.60−1.3	24	0.67	0.24−0.93
BC (ηg/m^3)	50	0.97	0.38−2.1	39	0.94	0.38−2.1	11	1.1	0.88−1.4
$PM_{2.5}$ (μg/m^3)	61	0.96	0.32−5.1	37	0.96	0.32−5.1	24	0.97	0.43−1.6
NO_2 (ppb)	75	1.05	0.29−10	47	0.99	0.29−4.2	28	1.08	0.35−10
CO (ppm)	68	1.1	0.12−2.7	40	1.0	0.22−2.2	28	1.6	0.12−2.7
BTEX (μg/m^3)	48	1.1	0.21−2.6	37	1.0	0.21−1.4	11	1.3	0.48−2.6
Vancouver									
UFP (count/cm^3)	101	0.55	0.14−1.5	48	0.56	0.14−1.0	53	0.53	0.29−1.5
BC (ηg/m^3)	103	0.72	0.085−2.2	50	0.73	0.085−1.9	53	0.70	0.21−2.2
$PM_{2.5}$ (μg/m^3)	105	0.77	0.11−1.6	53	0.81	0.27−1.2	52	0.72	0.11−1.6
NO_2 (ppb)	103	1.06	0.02−13	49	0.9	0.02−13	54	1.34	0.12−13
CO (ppm)	50	0.48	0.12−3.8	50	0.48	0.12−3.8			
BTEX (μg/m^3)	102	1.2	0.22−14	53	1.2	0.35−14	49	1.2	0.22−8.4
Montreal									
UFP (count/cm^3)							23	0.64	0.27−0.79
BC (ηg/m^3)							9	0.80	0.37−1.2
$PM_{2.5}$ (μg/m^3)							22	1.01	0.66−1.6
NO_2 (ppb)							29	1.03	0.29−3.2
CO (ppm)							19	0.66	0.010−2.2
BTEX (μg/m^3)							21	1.71	0.61−3.1
Overall									
UFP (count/cm^3)	175	0.62	0.14−1.5	75	0.65	0.14−1.3	100	0.60	0.24−1.5
BC (ηg/m^3)	162	0.78	0.085−2.2	89	0.81	0.085−2.1	73	0.75	0.21−2.2
$PM_{2.5}$ (μg/m^3)	188	0.88	0.11−5.1	90	0.87	0.27−5.1	98	0.89	0.11−1.6
NO_2 (ppb)	207	1.05	0.02−13	96	0.98	0.02−13	111	1.09	0.12−13
CO (ppm)	139	0.84	0.010−3.8	90	0.77	0.12−3.8	47	1.2	0.10−2.7
BTEX (μg/m^3)	171	1.2	0.21−14	90	1.1	0.21−14	81	1.4	0.22−8.4

[a]n, Number of commutes.

Bayes Factors lead to the probability of including a given parameter in the model (Posterior Inclusion Probability (PIP)) which is an indication of the overall utility of this variable in making future predictions. Bayes Factors have also been shown to avoid overfitting the data and lead to optimal future predictions in future samples.[20] Candidate predictors with the highest PIPs (>50%) were selected for final multivariable models along with ambient temperature and wind speed which were included in all final models. An interaction term between ambient temperature and wind speed was also considered for final models but was only included if the PIP was greater than 50%. In addition, the variable reflecting residential land use was excluded from the final model selection process owing to a strong correlation with local roadways (r = 0.81).

In general, the optimal model for making future predictions is a weighted model average of all coefficients with the weights reflecting model probability. However, this implies that all variables must be included even if their PIP is very small and their contribution to future predictions is minimal. Therefore, a reasonable compromise is to eliminate those variables with small PIPs, sacrificing what is likely a small amount of predictive ability for a model with fewer parameters that will be easier to use in practice.

■ RESULTS

In total, 238 commutes were monitored throughout the study period. Air pollution and covariate data were complete for the majority of these routes although a small number of samples were lost due to instrument failure or technician error. In particular, 23 missing values for outdoor temperature were replaced using the linear relationship between vehicle roof-top temperature measurements and regional temperature values (vehicle roof-top temperature = 4.11 + 0.96 (regional temperature); R^2 = 0.95). In addition, six negative values of black carbon were removed from analysis. All VOC samples were above the limits of detection (0.01−0.15 μg/m^3). Seven NO_2 samples were below the limit of detection (1.4−3.3 ppb) and were replaced with half the detection limit. Estimates of accuracy and precision are shown for air monitoring data in the SI (Table S1); NO_2 measurements were least precise likely owing to the use of passive sampling over a short duration. During monitoring, ambient temperatures ranged from 3.7−15 °C in Montreal (mean = 9.4 °C), −5.3−28 °C in Toronto (mean = 14 °C), and 6−29 °C in Vancouver (mean = 14 °C).

Regional outdoor concentrations of $PM_{2.5}$ (median: 4.02 μg/m^3, range: 0.663−31.3 μg/m^3) and NO_2 (median: 15.0 ppb, range: 5.63−28.2 ppb) were generally low during commuting periods and were consistently below in-vehicle concentrations. Descriptive data for in-vehicle air pollution concentrations are shown in Table 1 and indoor/outdoor ratios are presented in Table 2. In-vehicle air pollution concentrations varied substantially between commutes and correlations between pollutants were strongest for UFPs and black carbon (r = 0.69), $PM_{2.5}$ and UFPs (r = 0.52), and $PM_{2.5}$ and NO_2 (r = 0.58). As expected, in-vehicle pollutant concentrations were also highly correlated with outdoor levels (0.70 ≤ r ≤ 0.84). In-vehicle concentrations were similar for most pollutants between cities with the exception of $PM_{2.5}$ and NO_2 which were higher in

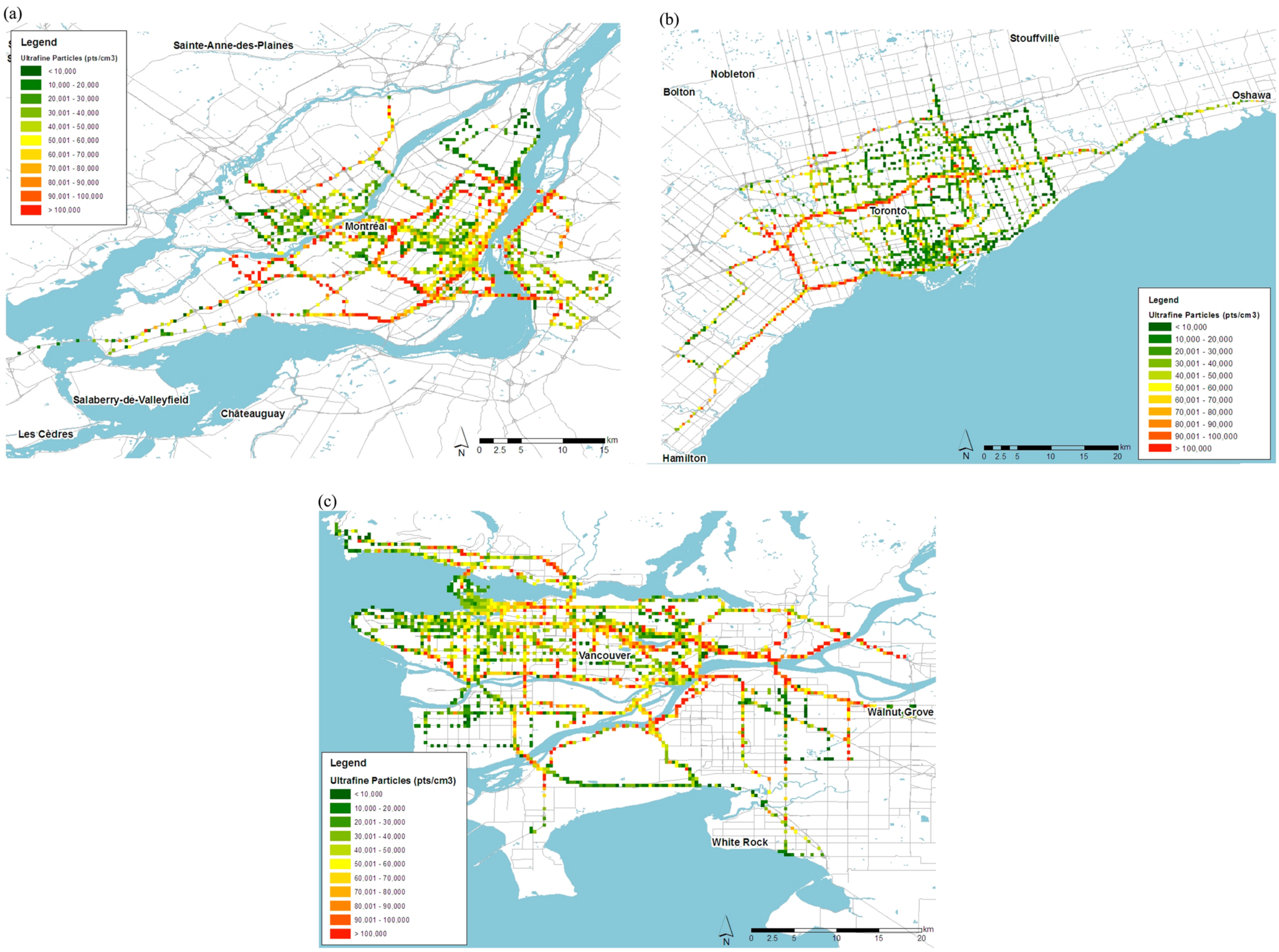

Figure 1. Spatial variability of in-vehicle UFP concentrations during winter for Montreal (a), Toronto (b), and Vancouver (c).

Table 3. Descriptive Data for Candidate Predictor Variables[a]

candidate predictors	n	mean (SD)	median	range
ambient temperature (°C)	238	13 (7.5)	14	−5.3–29
wind speed (km/h)	238	14 (10)	14	0.02–49
perceived traffic density[b]	232	2.2 (0.63)	2.1	0.82–3.8
mean diesel count (min^{-1})[c]	232	0.43 (0.27)	0.37	0.012–1.2
Land Use Variables[d]				
commercial (m^2)	221	21,104 (21 718)	16 268	444–227 138
government (m^2)	221	40,572 (25 849)	35 238	5691–159 872
industrial (m^2)	221	150,383 (49 198)	151 554	52 925–325 169
park land (m^2)	221	73,460 (27 965)	72 253	17 455–177 614
residential (m^2)	221	372,329 (90 110)	375 686	139 328–562 813
water (m^2)	221	25,777 (22 617)	21 911	0–100 795
highway roads (m)	221	2291 (1697)	1805	0–7727
local roads (m)	221	7732 (1756)	7920	3993–12 162

[a] n, Number of commutes. [b] Driver's perceived traffic density on a scale of 0 (no traffic) to 4 (completely surrounded) based on digital voice recordings. [c] Rate (count/min) of encountering diesel vehicles during a commute based on digital voice recordings. [d] Within a 500 m buffer averaged over each commute.

Toronto than in Vancouver during the summer months. During winter, median $PM_{2.5}$ concentrations were greatest in Montreal. The spatial variability of in-vehicle UFP concentrations are shown in Figure 1 for the winter monitoring periods.

In general, indoor/outdoor ratios were less than 1 or very close to 1 and mean comparison tests between indoor and outdoor concentrations of NO_2 (mean difference = 4.4 ppb, 95% CI: −0.25, 9.12) and $PM_{2.5}$ (mean difference: −0.614 $\mu g/m^3$, 95% CI: −1.34, 0.117) did not suggest a systematic difference between indoor and outdoor concentrations. Alternatively, on average outdoor concentrations of UFPs (mean difference: −18, 416 cm^{-3}, 95% CI: −20, 919, −15,

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913), black carbon (mean difference: −544 ηg/m^3, 95% CI: −684, −405), and CO (mean difference = −3.92 ppm, 95% CI: −5.32, −2.53) were higher than indoor values, whereas indoor BTEX was higher than outdoor concentrations (mean difference: 3.99 μg/m^3, 95% CI: 2.00, 5.98). It is important to note that indoor BTEX may also be influenced in-vehicle sources of VOCs such as adhesives used inside the cabin in addition to outdoor sources.

Descriptive data for candidate predictor variables evaluated in multivariable linear regression models are shown in Table 3 and their estimated impacts on in-vehicle air pollution concentrations are shown in Table 4 (models for indoor/outdoor ratios are shown in SI Table S4). The 500 m buffer was retained for analyses of land use and road length variables as the magnitude of associations with air pollution concentrations were consistently strongest for this buffer size (data not shown). Of the candidate predictors examined, mean diesel counts were strongly associated with in-vehicle concentrations (and indoor/outdoor ratios) of all three particulate air pollutants whereas perceived traffic density was associated with increased in-vehicle UFPs and black carbon with a weaker relationship observed for $PM_{2.5}$. Highway road length was positively associated with in-vehicle concentrations (and indoor/outdoor ratios) of all three pollutants whereas residential land use was inversely associated with in-vehicle concentrations of these pollutants (and indoor/outdoor ratios). Similarly, increased use of local roads was associated with significant decreases in in-vehicle UFPs and black carbon as well as indoor/outdoor ratios for these pollutants. Other land use variables were not associated with UFPs but commercial and government land use were both positively associated with in-vehicle $PM_{2.5}$ concentrations and industrial land use was associated with a decrease in the indoor/outdoor ratio for black carbon. In addition, in-vehicle black carbon concentration were inversely associated with commercial land use and positively associated with water coverage. Scatter plots of in-vehicle air pollution concentrations are shown in the SI (Figures S15−S28) for the strongest associations reported in Table 4. Final multivariable models for in-vehicle pollutant concentrations are shown in Table 5 (models for indoor/outdoor ratios are shown in SI Table S5). The final model for UFPs had the largest coefficient of determination (R^2 = 0.53) and the lowest root-mean-square error (RMSE) and included highway road length, ambient temperature and wind speed, and an interaction term between ambient temperature and wind speed. For $PM_{2.5}$, highway road length and government/institutional land use were retained in final models whereas local road length and commercial land use were retained in models for in-vehicle black carbon concentrations. Posterior inclusion probabilities (PIPs) were close to 1 for all variables included in final models for in-vehicle pollutant concentrations with the exception of temperature and wind speed in the $PM_{2.5}$ model. Likewise, PIPs were close to 1 for land use/road length variables included in

Table 4. Single-Predictor Models for In-Vehicle Particulate Air Pollution[a]

	percent change (95% CI)		
candidate predictors	UFPs	$PM_{2.5}$	black carbon
traffic density[b]	28% (14, 43)*	4.0% (−12, 24)	17% (−0.41, 38)
mean diesel count (min^{-1})[c]	106% (58, 168)*	210% (114, 348)*	123% (56, 218)*
Land Use Variables			
commercial (m^2)[d]	−2.3% (−5.8, 1.4)	7.8% (2.3, 14)*	−6.3% (−12, −0.43)*
government (m^2)[d]	0.42% (−2.8, 3.7)	11% (6.8, 16)*	−1.4% (−5.5, 2.8)
industrial (m^2)[d]	1.3% (−0.25, 2.9)	1.2% (−1.1, 3.6)	−0.62% (−2.7, 1.6)
park land (m^2)[d]	0.28% (−2.4, 3.0)	3.1% (−0.78, 7.2)	2.6% (−1.1, 6.5)
residential (m^2)[d]	−2.4% (−3.2, −1.7)*	−1.6% (−2.8, −0.43)*	−2.3% (−3.3, −1.3)*
water (m^2)[d]	2.3% (−1.0, 5.8)	0.068% (−4.6, 4.9)	4.7% (0.23, 9.4)*
highway roads (m)[e]	18% (13, 23)*	9.4% (2.9, 16)*	13% (7.1, 20)*
local roads (m)[e]	−9.9% (−13, −6.2)*	1.8% (−4.3, 8.2)	−12% (−17, −7.4)*

[a]All models are adjusted for mean ambient temperature and wind speed. [b]Traffic density on a scale of 0 (no traffic) to 4 (completely surrounded) based on digital voice recordings. [c]Coefficient reflects 1 unit change in the rate (count/min) of encountering diesel vehicles during a commute based on digital voice recordings. [d]Coefficient reflects a 10 000 m^2 increase within a 500 m buffer. [e]Coefficient reflects a 1000 m increase within a 500 m buffer.

Table 5. Final Multivariable Models for In-Vehicle Particulate Air Pollution Concentrations[a]

dependent variable	intercept	independent variables	coefficient (95% CI)	PIP	R^2	RMSE
log(UFP)	10.9	length of highway roads[a]	0.1819 (0.1450, 0.2188)	1.0	0.53	0.400
		temperature	−0.06715 (−0.08015, −0.05415)	1.0		
		wind speed	−0.04847 (−0.06099, −0.03594)	1.0		
		temperature × wind speed	0.002610 (0.001892, 0.003328)	1.0		
log($PM_{2.5}$)	1.78	length of highway roads[a]	0.159 (0.101, 0.218)	1.0	0.24	0.670
		government/institutional[b]	0.146 (0.105, 0.188)	1.0		
		temperature	−0.0113 (−0.0250, 0.00244)	0.49		
		wind speed	−0.0107 (−0.0206, −0.000840)	0.59		
log(BC)	9.61	length of local roads[a]	−0.1397 (−0.1886, −0.09083)	0.92	0.30	0.567
		commercial[b]	−0.08081 (−0.1346, −0.02704)	0.89		
		temperature	−0.04717 (−0.06640, −0.02794)	1.0		
		wind speed	−0.06118 (−0.08022, −0.04215)	1.0		
		temperature × wind speed	0.003011 (0.002027, 0.003994)	1.0		

[a]PIP, posterior inclusion probability. [a]Coefficient reflects a 1000 m increase within a 500 m buffer. [b]Coefficient reflects a 10,000 m^2 increase within a 500 m buffer.

final models for indoor/outdoor ratios although PIPs for ambient temperature and wind speed were lower and in general these models explained only a small portion of the variation in indoor/outdoor ratios ($0.10 \leq R^2 \leq 0.15$).

■ DISCUSSION

Air pollution is a known public health threat and is recognized as an important contributor to global disease burden.[21] To date, population-based studies interested in the potential health effects of air pollution have generally assigned exposures to residential locations; however, in-vehicle exposures may also be important as some evidence suggests that daily commutes can have a meaningful impact on overall exposure levels for some pollutants.[8−11] The Urban Transportation Exposure Study was designed to characterize Canadian commuter exposures to traffic-related air pollutants and to identify determinants of these exposures. In general, our findings suggest that vehicle commuters are repeatedly exposed to elevated levels of several air pollutants (relative to background concentrations) and that traffic characteristics, road types, meteorology, and land use characteristics have an important impact on in-vehicle concentrations.

Similar studies in Europe and North America have generally reported higher in-vehicle exposures to traffic-related air pollutants than those observed in Canada.[17,22−27] For some pollutants, such as black carbon, differences between North America and Europe are likely explained in part by a higher prevalence of diesel vehicles in European countries.[22] In final multivariable models, highway roads were the most important determinants of in-vehicle exposures to UFPs and $PM_{2.5}$ after adjusting for meteorology whereas local roads were most important for black carbon likely owing to the general absence of large diesel vehicles on local roads. Previous studies also reported that road type and nearby diesel vehicles were important determinants of in-vehicle air pollution concentrations[9,30,31] but other studies using land use characteristics to predict in-vehicle air pollution concentrations were not identified. While the final model for UFPs explained 53% of the variability in in-vehicle exposures, models for $PM_{2.5}$ and black carbon did not perform as well and other parameters should be considered in future studies to improve all of the models presented. For example, one obvious candidate is a variable to explicitly capture diesel vehicles as the rate of encountering diesel vehicles was the strongest predictor of in-vehicle exposure to UFPs, black carbon, and $PM_{2.5}$ in single-variable models. However, as noted above, the rate of encountering diesel vehicles was not evaluated in final multivariable models as this information cannot be measured at the individual level in population-based studies. One alternative may be to catalogue the distribution of traffic characteristics at multiple points along roadways in major urban areas and use this information as a surrogate measure of the likelihood of encountering diesel vehicles along a given route. However, the development of such databases would require considerable time, effort, and resources and use of such surrogate data would almost certainly underestimate the impact of diesel vehicles on in-vehicle air pollution concentrations owing to exposure measurement error. Nevertheless, our evaluation of land use parameters suggests that this information is not captured by traditional GIS predictors as few were strongly associated with in-vehicle air pollution concentrations. Therefore, the development of such databases may be justified if we hope to refine exposure assessment in population-based studies beyond simply assigning exposures to the place of residence. Alternatively, future studies should examine the possibility of using exposure surfaces for outdoor air pollution in conjunction with individual-level origin/destination data to estimate in-vehicle exposures as in-vehicle concentrations were highly correlated with outdoor values. This approach is conceptually similar to methods currently used by route planning applications to estimate low-exposure routes for cyclists in urban areas.[40]

Cabin ventilation is also known to be an important predictor of in-vehicle air pollution concentrations[32,33] but was not specifically examined in this study as detailed information on vehicle air exchange rates would be difficult to obtain in large population-based studies. Recently, Hudda et al.[32] identified three factors that were important predictors of in-vehicle air exchange rates with ventilation settings set to outdoor air intake: fan strength, vehicle speed, and cabin volume. In this study, two of these three factors (fan speed and cabin volume) were kept constant throughout air monitoring and thus speed likely had a more important impact on between-route variations in cabin air exchange in our study. However, as noted above, speed was highly correlated with both highway and local road length and thus was at least partially captured the impact of speed on cabin air exchange through these land use predictors.

While this study had many important advantages including detailed monitoring of in-vehicle (and outdoor) air pollution concentrations along different road types during multiple commutes in Canadian metropolitan areas it is important to note several limitations. First, electric vehicles were not used in this study and thus we cannot rule out some contribution from study vehicles to indoor/outdoor concentrations. In addition, outdoor PM samplers were not equipped with isokinetic inlets and thus it is possible that particles of different sizes did not enter the sampling inlet proportionately. However, as the same vehicle type and monitoring design was used for each route these factors would not explain the observed associations for predictors of in-vehicle air pollution concentrations. Moreover, this study was limited to a single vehicle type and thus our findings may not be generalizable to the entire range of in-vehicle exposures experienced by commuters in the broader Canadian fleet owing to differences in factors such as vehicle age, cabin volume, and model type.[32,34,35] However, in-vehicle UFP concentrations observed in this study were comparable to values measured previously in a private vehicle in Montreal, Canada.[7] In addition, others studies have reported similar in-vehicle air pollutant concentrations between gasoline and diesel automobiles[36] and thus our findings may provide a reasonable range of values typically encountered by Canadian commuters. A second limitation is that our models did not explicitly include a term for vehicle ventilation and this likely contributed to that fact the models presented left a large portion of the variation in-vehicle concentrations and indoor/outdoor ratios unexplained. Indeed, air exchange rates are known to be important predictors of in-vehicle air pollution concentrations[32,33] and some evidence suggests that these values may be estimated under recirculation conditions based on vehicle age, mileage, speed, and manufacturer.[37] If possible, future studies aimed at estimating in-vehicle air pollution concentrations should incorporate parameters to specifically capture variations in cabin ventilation. Nevertheless, models based on the factors identified in this study may offer an incremental improvement to exposure assessment in population-based studies as the current approach of assigning values to residential locations ignores commuting exposure entirely. Finally, although our study was designed to include temperature conditions typical of Canadian values, winter temperatures during monitoring were rather mild. As a result, descriptive data for pollutants impacted by temperature, such as UFPs, may underestimate exposures during colder periods.

In general, our findings suggest that Canadian vehicle commuters may be repeatedly exposed to elevated levels of traffic-related air pollutants and that traffic characteristics, land use, road type, and meteorology are important determinants of these exposures. Models based on these factors may be useful in population-based studies interested in capturing in-vehicle air pollution exposures as a compliment to residential exposure estimates.

■ ASSOCIATED CONTENT

Supporting Information

Summary of data cleaning procedures, precision, accuracy, and instrument sensitivity (S1), scatter plot of indoor/outdoor $PM_{2.5}$ and vehicle speed (Figure S1), scatter plot of gravimetric versus Dustrak $PM_{2.5}$ (Figure S2), box plots of indoor/outdoor air pollution data and indoor/outdoor ratios (Figures S3−S9), photograph of monitoring setup (Figure S10), correlation matrix (Table S3), models for indoor/outdoor ratios (Tables S4−S5), scatter plots of untransformed air pollution data for the strongest associations in Table 4 (Figures S11−S24). This material is available free of charge via the Internet at http://pubs.acs.org/.

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Notes

The authors declare no competing financial interest.

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