Environmental Pollution 208 (2016) 241–248

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Characterizing the spatial distribution of ambient ultrafine particles in Toronto, Canada: A land use regression model

Scott Weichenthal [a, *], Keith Van Ryswyk [a], Alon Goldstein [b], Maryam Shekarrizfard [c], Marianne Hatzopoulou [c]

[a] Air Health Science Division, Health Canada, Ottawa, Canada
[b] School of Urban Planning, McGill University, Montreal, Canada
[c] Department of Civil Engineering, McGill University, Montreal, Canada

A R T I C L E I N F O

Article history:
Received 7 January 2015
Received in revised form 14 April 2015
Accepted 17 April 2015
Available online 29 April 2015

Keywords:
Ultrafine particles
Land use regression
Traffic
Built environment

A B S T R A C T

Exposure models are needed to evaluate the chronic health effects of ambient ultrafine particles (<0.1 μm) (UFPs). We developed a land use regression model for ambient UFPs in Toronto, Canada using mobile monitoring data collected during summer/winter 2010–2011. In total, 405 road segments were included in the analysis. The final model explained 67% of the spatial variation in mean UFPs and included terms for the logarithm of distances to highways, major roads, the central business district, Pearson airport, and bus routes as well as variables for the number of on-street trees, parks, open space, and the length of bus routes within a 100 m buffer. There was no systematic difference between measured and predicted values when the model was evaluated in an external dataset, although the R^2 value decreased ($R^2 = 50\%$). This model will be used to evaluate the chronic health effects of UFPs using population-based cohorts in the Toronto area.

1. Introduction

Traffic-related air pollution is known to contribute to cardiovascular morbidity including both acute and chronic health effects (Hoek et al., 2013; Mehta et al., 2013; Shah et al., 2013; Sun et al., 2010; Weichenthal, 2012). To date, population-based studies interested in the potential health effects of traffic-related air pollution have generally relied on NO_2 as a surrogate measure of exposure owing to the availability of existing land use regression models (Crouse et al., 2009, 2010; Jerrett et al., 2009). However, other air pollutants such as ultrafine particles (UFPs) (<0.1 um) may also contribute to adverse health effects. In particular, a number of studies have examined the acute health effects of UFPs and existing evidence suggests that these pollutants may contribute to acute changes in vascular function and cardiac autonomic modulation (Weichenthal, 2012) likely through pathways involving oxidative stress (Miller et al., 2012; Miller, 2014). Nevertheless, few studies have evaluated the chronic health effects of UFPs largely owing to the absence of exposure models suitable for use in large population-based studies. However, one recent study used a chemical transport model to estimate residential exposure to ambient UFPs and the findings suggest that UFP exposures may contribute to ischemic heart disease mortality (Ostro et al., 2015). To date, land use regression models have been developed for UFPs in Vancouver, Canada (Abernethy et al., 2013), Girona, Spain (Rivera et al., 2012), and Amsterdam, Netherlands (Hoek et al., 2011) but studies have yet to apply these models to examine associations between long-term exposure to UFPs and cardiovascular morbidity/mortality. In this study, we developed a land use regression model for UFPs in Toronto, Canada in order to characterize the spatial distribution of these pollutants in Canada's largest city. Sabaliauskas et al. (2015) recently described a land use regression model for Toronto based on afternoon monitoring data collected during summer 2008. Here we expand on this previous campaign by including more recent data collected during morning and afternoon periods in both the summer and winter months over a broader geographic region using mobile monitoring. Mobile monitoring has many advantages in conducting such studies as it offers a cost-effective means of characterizing spatial variations in ambient UFPs over large geographic areas that may otherwise be infeasible to capture given practical constraints.

* Corresponding author. 269 Laurier Ave West, Ottawa, Ontario, K1A 0K9, Canada.
E-mail address: scott.weichenthal@hc-sc.gc.ca (S. Weichenthal).

http://dx.doi.org/10.1016/j.envpol.2015.04.011
0269-7491/

2. Methods

2.1. Mobile monitoring of ultrafine particles

Ambient UFP data were collected during a mobile monitoring campaign conducted in Toronto, Canada for two weeks in September 2010 (summer) and one week in March 2011 (winter). These months were selected to capture the wide range of temperatures typical of Toronto, Canada. Details of this campaign have been described previously (Weichenthal et al., 2015). Briefly, each day three separate vehicles (Chevrolet Grand Caravans) equipped with roof-top monitoring devices (TSI Model 3007) monitored real-time ambient UFPs (<0.1 um) at 1-second resolution. Each vehicle collected UFP data for six hours each day: once in the morning (7:00–10:00) and once in the afternoon (15:00–18:00). All samples were collected on weekdays and ambient temperatures ranged from −9.2–24 °C (mean = 10.3 °C). Each vehicle focussed on specific portions of the city including downtown areas, major highways, and suburban areas. Dedicated routes were not assigned; instead, drivers focused on maximizing coverage of these regions during each sampling period with a different route taken each day. All vehicles carried a GlobalSat DG-100 monitor to log geographic coordinates which were subsequently matched to real-time UFP data at 1-second resolution.

2.2. Assigning ultrafine particle concentrations to road segments

The mid-point of each road segment (mean length: 162 m; interquartile range: 74–201 m) was assigned a mean UFP concentration based on data collected throughout the monitoring campaign over both seasons (Supplemental Material Fig. S1). The number of data points available for each road segment varied depending on the number of times it was traversed throughout the monitoring period. Our model is based on road segments with at least 250 UFP data points (mean: 595 points/segment; interquartile range: 312–690) as this threshold provided the best balance of spatial coverage and points per segment for model development. In preliminary analysis, we also examined models based on road segments with at least 400–600 data points (6.7–10 min per segment) to increase the duration of measurement data available for each segment; however, this resulted in decreased spatial coverage and points primarily reflected major highways (data not shown). Similarly, models based on road segments with at least 100–200 data points were examined but only small gains in spatial coverage were apparent and model RMSE (root mean square error) values increased owing to a decreased number of points per segment. Therefore, the final criteria of at least 250 points per segment was selected as this threshold provided the best balance of spatial coverage and air pollution data available for model development.

2.3. Derivation of land use and built environment data for model development

The midpoint of each road segment was geocoded in a geographic information systems (GIS) environment using ArcMAP10.2 and spatially intersected with a number of GIS layers describing land-use and built environment. We associated each point with a set of variables either by generating buffers around the point and calculating means or sums within the buffer or by computing distances between each point and potential sources. We generated several buffer sizes (50–300 m) and intersected these buffers with the following GIS layers: road network, bus network, restaurants, on-street trees, and land-use classes. We also generated five distance variables by computing the straight-line distance between every segment midpoint and the nearest highway, nearest major road, nearest bus route, the central business district, and Pearson International Airport. Air pollution maps were generated by first dividing the city of Toronto into 100 × 100 m grid cells. Buffers were drawn around the centroids of each grid cell and were intersected with land-use layers in order to derive predictors for each cell; final model coefficients were applied to each cell to generate a surface for UFPs at a resolution of 100 × 100 m.

2.4. Statistical analysis

Land use regression models were developed for mean UFP concentrations as well as ln-transformed UFP concentrations. Single-predictor linear regression models were first examined to evaluate the impact of each candidate predictor on ambient UFPs; in total, 44 predictors were evaluated. Candidate predictors included distances to potentially important sources including highways/major roads, bus routes, Pearson International airport (the major international airport in Toronto), and the central business district along with other factors integrated within circular buffers (100–300 m) including total road length, land use variables (e.g. residential, commercial, industrial, parks, open space), total restaurants/bars, total number of on-street trees, total bus stops, and total length of bus routes. Open space reflects undeveloped land not including parks or recreational areas. A 50 m buffer was also examined for total restaurants/bars. Variables for the natural logarithms of distance variables were also evaluated to account for non-linear decreases in UFPs with distance from traffic sources (Zhu et al., 2002). We did not place any *a priori* restrictions on the direction of coefficients for inclusion in multivariable models as the primary purpose of modeling was prediction.

Variables that were associated with ambient UFP concentrations in single-predictor models (i.e. 95% confidence intervals excluded the null) were retained for evaluation in multivariable models. If more than one buffer size was examined for a given variable, the buffer size with the strongest association (i.e. largest R^2 value and lowest RMSE) was retained for analysis. Spearman correlations were also examined between candidate predictors; if two variables were highly correlated ($r > 0.80$) the variable with the strongest association with UFP was retained for analysis. The remaining variables were included in multi-variable linear regression models. Variables that were not statistically significant in multivariable models were only removed if doing so decreased (or did not substantially change (i.e. <1%)) the RMSE of the model.

Although monitoring was conducted during the same portion of each day (i.e. morning and evening rush hour), individual road segments were monitored at different times on different days throughout the monitoring period and as a result temporal variations might have contributed to differences in UFP concentrations between road segments. Previous studies have used correction factors derived from fixed site monitoring data for UFPs (Abernethy et al., 2013; Hoek et al., 2011) or NO_x (Rivera et al., 2012) to adjust for temporal variations between samples collected at different times. Since fixed-site UFP data were not available in Toronto, we used ambient temperature to adjust for temporal variability between sampling periods as temperature is known to be an important determinant of day-to-day fluctuations in ambient UFPs (Alm et al., 1999; Kaur and Nieuwenhuijsen, 2009; Weichenthal et al., 2008, 2014; 2015). Specifically, each road segment was assigned a value for mean ambient temperature using real-time data (1-second resolution) collected from vehicle rooftop monitors (HOBO Datalogger) at the same time as UFP measurements. Both linear and quadratic terms for ambient temperature were included in all regression models to account for potential non-linearity in the relationship between temperature and UFPs. Wind speed was also

Table 1
Descriptive statistics for UFP concentrations (count/cm^3).

Statistic	UFP
Minimum	6334
10th Percentile	17,670
First quartile	25,060
Mean (SD)	44,419 (27,299)
Median	35,843
Third quartile	56,747
90th Percentile	89,297
Maximum	125,279

Data reflects a total of 440 road segments with at least 250 points/segment.

evaluated as a possible covariate to account for temporal variations in ambient UFPs using hourly data available from Environment Canada.

Potential bias and precision of final model estimates were evaluated by applying models to an external dataset of road segments that fell slightly below the threshold of 250 points per segment for inclusion in model development. Specifically, final models were used to predict UFP concentrations for road segments with 200–249 points per segment and the mean difference (95% CI) between measured and predicted values was used to evaluate potential bias in model estimates. Precision was evaluated using R^2 and RMSE values from linear models relating measured and predicted UFP concentrations and the slopes of these models were used to estimate the strength of the linear relationships between measured and predicted values.

3. Results

In total, 440 road segments had at least 250 data points covering a distance of approximately 170 km of roadway. On average, each road segment contained 10 min of UFP data (interquartile range: 5.2–11.5 min) and segments were distributed across a large portion of the greater Toronto area (Supplemental Material Fig. S2). Ambient UFP concentrations ranged substantially across road segments with a mean value of 44,419/cm^3 and a range of 6334–125,279/cm^3 (Table 1).

3.1. Single variable models

Descriptive data for candidate predictors are shown in Table 2 and a number of these factors were identified as potentially important predictors of ambient UFPs (Table 3) (models for ln(UFP)

Table 2
Descriptive statistics for candidate predictor variables.

Independent variable	Buffer size (m)	Mean (SD)	Minimum	Maximum
Distance to nearest highway		1266 (1394)	0	7032
Distance to nearest major road		22.1 (87.95)	0	1193
Distance to CBD		10,090 (6652)	195	38,190
Distance to Pearson Airport		16,670 (7133)	0	51,441
Distance to nearest bus route		74.7 (120)	0	646
Ambient temperature (°C)		10.3 (5.8)	−9.2	24
Wind speed (m/s)		17.0 (5.4)	0.03	43
Total road length	100	364 (173)	0	948
	200	1354 (571)	400	2886
	300	2931 (1148)	600	5706
Total intersections				
	100	0.39 (0.57)	0	2
	200	1.26 (1.34)	0	7
	300	2.52 (2.40)	0	13
Land use (%)				
Residential	100	31 (31)	0	100
	200	35 (29)	0	100
	300	37 (28)	0	100
Commercial	100	16 (23)	0	100
	200	15 (19)	0	88
	300	15 (17)	0	78
Industrial	100	25 (30)	0	100
	200	26 (28)	0	100
	300	26 (26)	0	99
Parks	100	6.3 (16)	0	100
	200	7.5 (15)	0	81
	300	8.1 (13)	0	66
Open space	100	22 (28)	0	100
	200	16 (20)	0	100
	300	13 (16)	0	100
Total restaurants	50	1.0 (2.8)	0	21
	100	3.1 (6.4)	0	42
	200	9.8 (19)	0	114
	300	19.6 (37)	0	215
Total on street trees	100	15 (20)	0	94
	200	71 (71)	0	331
	300	206 (158)	0	665
Total bus stops	100	1.4 (2.2)	0	12
	200	4.0 (4.4)	0	27
	300	4.8 (5.4)	0	28
Total length of bus routes	100	1812 (2854)	0	22,986
	200	4348 (6471)	0	44,463
	300	7920 (8398)	0	65,898

All distances are in meters; CBD, central business district.

Table 3
Single-predictor linear regression models for UFPs.

Independent variable	Buffer size (m)	β (95% CI)	Adjusted R^2	RMSE
ln(distance to highway)		−4061 (−4527, −3596)	0.46	20,074
Distance to highway		−8.93 (−10.51, −7.34)	0.29	22,940
ln(distance to major road)		428.9 (−1067, 1924)	0.09	25,987
Distance to major road		−49.05 (−76.69, −21.41)	0.12	25,640
Distance to CBD		1.04 (0.645, 1.44)	0.15	25,234
ln(distance to CBD)		6574 (4068, 9082)	0.15	25,236
Distance to Pearson Airport		−1.37 (−1.69, −1.04)	0.22	24,186
ln(distance to Pearson Airport)		−12,683 (−15,290, −10,077)	0.26	23,440
Distance to bus route		52.68 (32.53, 72.83)	0.15	25,284
ln(distance to bus route)		4209 (3183, 5234)	0.21	24,278
Total road length	100	12.75 (−1.59, 27.10)	0.10	25,906
	200	−0.374 (−4.75, 4.00)	0.09	25,996
	300	−0.597 (−2.78, 1.59)	0.09	25,988
Total intersections	100	−12,909 (−17,293, −8525)	0.19	24,513
	200	−6250 (−8203, −4297)	0.20	24,343
	300	−3550 (−4649, −2452)	0.20	24,321
Land use				
Residential	100	−18,850 (−26,475, −11,224)	0.14	25,317
	200	−13,704 (−21,970, −5439)	0.12	25,684
	300	−11,328 (−20,115, −2542)	0.11	25,807
Commercial	100	−20,803 (−31,814, −9793)	0.12	25,593
	200	−18,238 (−31,481, −4995)	0.11	25,780
	300	−16,815 (−31,875, −1754)	0.10	25,854
Industrial	100	−6296 (−14,442, 1849)	0.09	25,928
	200	−3015 (−11,738, 5709)	0.09	25,983
	300	−2340 (−11,780, 7099)	0.09	25,990
Parks	100	−16,157 (−31,059, −1256)	0.10	25,862
	200	−15,165 (−31,778, 1447)	0.10	25,901
	300	−17,743 (−35,967, 480)	0.10	25,888
open space	100	53,374 (45,724, 61,024)	0.37	21,715
	200	61,617 (50,565, 72, 670)	0.29	23,008
	300	68,619 (55,050, 82,188)	0.26	23,463
Total restaurants and bars	50	−1869 (−2731, −1007)	0.13	25,469
	100	−1052 (−1429, −675)	0.15	25,140
	200	−351 (−481, −221)	0.15	25, 194
	300	−173 (−241, −105)	0.14	25, 274
Total on street trees	100	−404 (−528, −280)	0.20	24,299
	200	−88.2 (−122, −54.1)	0.18	24,732
	300	−35.1 (−50.6, −19.6)	0.16	24,902
Total bus stops	100	−3783 (−4873, −2692)	0.18	24,746
	200	−2124 (−2659, −1590)	0.21	24,379
	300	−379 (−845, 87.1)	0.10	25,982
Total length of bus routes	100	−1.93 (−2.78, −1.08)	0.13	25,463
	200	−0.564 (−0.940, −0.189)	0.11	25,787
	300	−0.195 (−0.491, 0.101)	0.10	26,009
Meteorology[a]				
Temperature		2515 (1512, 3518)	0.10	25,967
Temperature^2		−165 (−215, −116)		

[a] Model includes both temperature variables. All models for candidate predictors include linear and quadratic terms for ambient temperature and all distances are in meters. CBD, central business district.

are available in the Supplemental Material Table S1). In general, variables for the natural logarithm of distances to the nearest highway, the nearest bus route, and Pearson airport explained the largest proportion of the variation in ambient UFPs along with predictors for the proportion of open space (100 m buffer) and the total number of on-street trees (100 m buffer) followed by various other factors. Spearman correlations between candidate predictors were less than 0.8 ($0.012 \leq r \leq -0.76$) and none of the predictors were eliminated because of co-linearity (Supplemental Material Table S2). Models including only linear and quadratic terms for ambient temperature explained approximately 10% of the variation in ambient UFP concentrations. Regional wind speed was not strongly associated with ambient UFPs assigned to each road segment ($\beta = 80.8$, 95% CI: −365, 527) ($R^2 < 0.01$) and as a result we did not use wind speed to account for temporal variability. As we were limited to regional wind speed data during the hour of UFP monitoring, measurement error may have limited our ability to detect a relationship between wind speed and ambient UFPs.

3.2. Multivariable models

Complete covariate data were available for 405 road segments and final land use regression models are shown in Table 4. The model for mean UFP concentrations explained 67% of the spatial variation in ambient UFP concentrations and included variables for the natural logarithm of distances to the nearest highway, major roads, the central business district, bus routes, and Pearson airport, along with variables for the proportion of open space and park land use (100 m buffer), the number of on-street trees (100 m buffer), and the length of bus routes (100 m buffer). Variables for park land use, the number of on-street trees, and the length of bus routes were not statistically significant but removing these factors increased the RMSE value by 8.5% and decreased the adjusted R^2 values by 6% and thus these factors were retained. The model for ln(UFP) explained a slightly smaller proportion of the variation in ambient UFPs (adjusted $R^2 = 0.60$) but included many of the same covariates with the exception of on-street trees and park land use

Table 4
Land use regression models for UFPs in Toronto, Canada (n = 405).

Dependent variable	Alpha	Independent variables	β (95% CI)	Adjusted R^2	RMSE
Mean UFP	133,042	ln(distance to highway[a])	−2975 (−3543, −2407)	0.67	15,680
		ln(distance to major road[a])	−2754 (−3806, −1704)		
		ln(distance to CBD[a])	5182 (3409, 6956)		
		ln(distance to Pearson Airport[a])	−12,434 (−15,515, −9355)		
		ln(distance to bus route)	1486 (586, 2385)		
		Park land[b]	−8160 (−17,630, 1310)		
		Open space[b]	13,145 (4467, 21,824)		
		Total on-street trees[b]	−31.47 (−125, 61.6)		
		Length of bus routes[b]	−0.341 (−0.969, 0.286)		
		Mean temperature	−47.91 (−759, 664)		
		Mean temperature^2	−17.46 (−53.5, 18.5)		
ln(UFP)	11.29	ln(distance to highway[a])	−0.0723 (−0.0859, −0.0586)	0.60	0.3855
		ln(distance to major road[a])	−0.0905 (−0.116, −0.0648)		
		ln(distance to CBD[a])	0.124 (0.0628, 0.184)		
		ln(distance to Pearson Airport[a])	−0.157 (−0.233, −0.0819)		
		ln(distance to bus route[a])	0.0183 (−0.00369, 0.0404)		
		Total intersections[c]	0.0138 (−0.0120, 0.0396)		
		Commercial land use[b]	0.107 (−0.0842, 0.299)		
		Open space[b]	0.379 (0.167, 0.591)		
		Length of bus routes[b]	−0.0000122 (−0.0000282, 0.00000389)		
		Mean temperature	−0.00106 (−0.0186, 0.0164)		
		Mean temperature^2	−0.000374 (−0.00127, 0.000517)		

[a] Distance in meters.
[b] 100 m buffer.
[c] 300 m buffer; CBD, central business district.

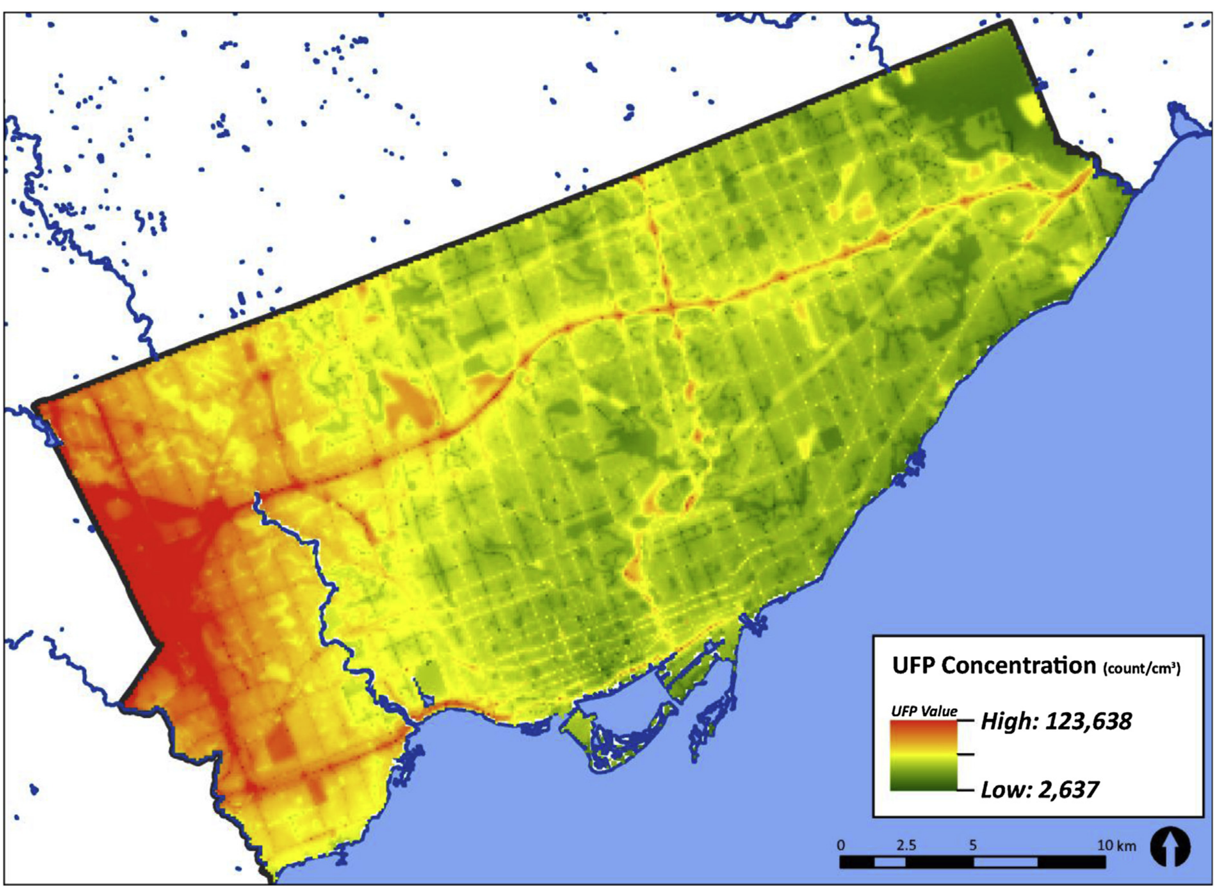

Fig. 1. Predicted spatial distribution of ambient UFPs in Toronto, Canada.

which were not included in the model. Variables for commercial land use (100 m buffer), total intersections (300 m buffer), the length of bus routes (100 m buffer), and the natural logarithm of the distance to the nearest bus route were not statistically significant but removing these variables increased the RMSE value by 5.9% and decreased the adjusted R^2 value by 5% and thus these factors were

Table 5
Measured and Predicted UFP Concentrations at model evaluation sites (count/cm^3).

Measured values mean (SD)		Predicted values mean (SD)		Mean difference (95% CI)		β[a] (95% CI)		R^{2a}		RMSE[a]	
UFP	ln(UFP)	UFP	ln(UFP)	UFP	ln(UFP)	UFP	ln(UFP)	UFP	ln(UFP)	UFP	ln(UFP)
35,938 (19,532)	10.360 (0.512)	37,323 (18,745)	10.369 (0.397)	−1385 (−3754, 982)	−0.00869 (−0.0739, 0.0565)	0.73 (0.61, 0.85)	0.81 (0.65, 0.98)	0.50	0.39	13,907	0.400

Model evaluation is based on 151 road segments with 200–249 points/segment.
[a] For linear regression model relating measured and predicted values.

retained. Variance inflation factors (VIF) were less than 3 (range: 1.15–2.89) for all predictor variables except for the temperature terms which had VIF values less than 7, thus suggesting limited collinearity among variables. Weak spatial autocorrelation was observed for UFP concentrations (Morans' $I = 0.22$) and for model residuals (Morans' $I = 0.050$). Fig. 1 illustrates the predicted spatial distribution of mean UFPs in Toronto, Canada at 10 °C (this is approximately the annual average temperature in Toronto).

Coefficients for ambient temperature were not statistically significant in final multivariable models and excluding these terms increased RMSE values by 0.45–2% and decreased R^2 values by a similar magnitude. This suggests that temporal variations were perhaps not as important as spatial differences in explaining overall variability in ambient UFPs. To explore this notion further, we also calculated within-segment (i.e. day to day temporal variation) and between-segment (spatial variation) values for the coefficient of variation (COV) (standard deviation/mean). The median within-segment COV value was 0.45 compared to the overall between-site value of 0.61; thus suggesting that spatial contributions to overall variation in ambient UFPs exceeded the temporal contribution.

3.3. Model evaluation

In total, 151 road segments with 200–249 data points were available for model evaluation and the range of UFP concentrations (5328–110,663/cm^3) and ambient temperatures (−4.6–25.3 °C) on these segments were comparable to the range used for model development. There were no systematic differences between measured and predicted values for mean UFPs or ln(UFPs) and strong positive slopes (β > 0.70) were observed in linear models relating measured and predicted values; however, R^2 values were lower when the models were applied to the external dataset (Table 5). A scatter plot of measured and predicted mean UFP concentrations is shown in Fig. 2.

4. Discussion

Little is currently known about the long-term health effects of ambient UFPs owing to the absence of exposure models suitable for use in large population-based studies. We developed a land use regression model for ambient UFPs in Toronto, Canada to characterize the spatial distribution of UFPs in Canada's largest city. This model explained the majority of the spatial variation in ambient UFPs and is available to support future population-based cohort studies. In particular, studies of chronic cardiovascular outcomes are of interest owing to a number of short-term studies suggesting adverse cardiovascular health effects (Weichenthal, 2012).

In general, our model explained a similar or slightly larger proportion of the spatial variability in ambient UFPs compared to previous models developed for Vancouver, Canada (Abernethy et al., 2013), Girona, Spain (Rivera et al., 2012), and Amsterdam, Netherlands (Hoek et al., 2011). In particular, while the Vancouver model generally reported lower R^2 values (0.29–0.53), model RMSE vales were smaller perhaps owing in part to longer monitoring periods (60 min) or the inclusion of variables related to truck routes and truck counts that were not available for Toronto. Our model also explained a similar proportion of the spatial variability in ambient UFPs compared to a previous LUR model for Toronto based on afternoon data collected during the summer months

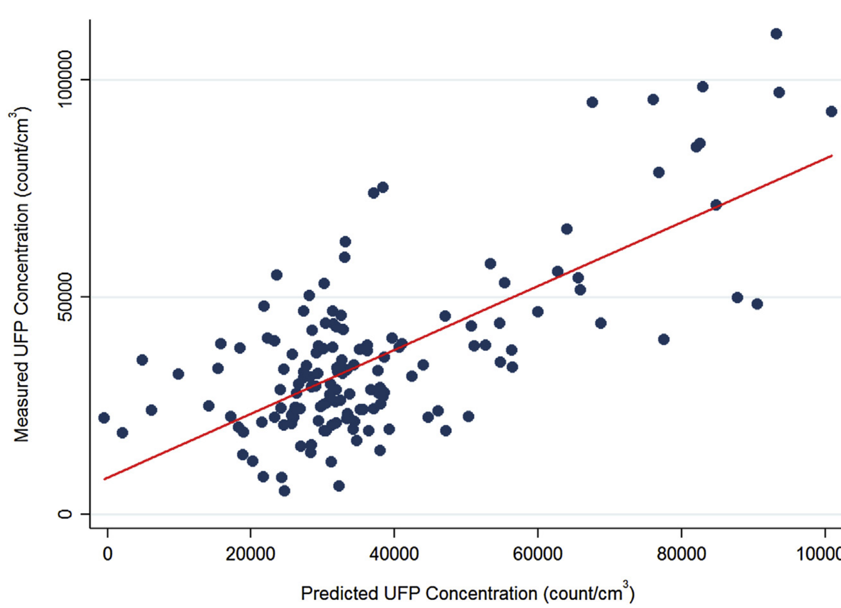

Fig. 2. Scatter plot of measured versus predicted mean UFP concentrations at model evaluation sites (count/cm^3).

(temperature range: 25–32 °C) in 2008 over a more limited geographical area (Sabaliauskas et al., 2015). However, our findings suggest that this model may underestimate long-term mean UFP concentrations owing to higher UFPs during the winter months and the fact that monitoring was limited to residential areas. In particular, the highest mean UFP concentration for road segments included in the previous study was approximately 20,000/cm^3 whereas values larger than 100,000/cm^3 were observed during our mobile monitoring campaign.

In general, while UFP models to date all contain similar terms for traffic intensity and/or distances to major roadways, several between-city differences are apparent. For example, in Vancouver restaurant density was positively associated with ambient UFPs but a similar relationship was not observed for Toronto in this study or previously (Sabaliauskas et al., 2015). Moreover, port proximity was identified as an important determinant of ambient UFPs in both Vancouver and Amsterdam but airport proximity was not specifically evaluated in previous studies. In this study, distance to Pearson International airport was identified as an important predictor of ambient UFPs and this is consistent with recent evidence suggesting that airports are important sources of these pollutants (Hudda et al., 2014). While the magnitude of this association was small, our model is the first to incorporate airport proximity and suggests that airports have a measurable impact on ambient UFPs after adjusting for other important factors including proximity to highways and major roads. Furthermore, some evidence suggests that green infrastructure may improve urban air quality (Pugh et al., 2012) and our model is the first to include a term for the number of on-street trees. While this term was not statistically significant in the final multivariable model, it had a meaningful impact on the accuracy and precision of model estimates and future models should also explore the potential impact of this variable on model performance as the relationship between urban vegetation and urban air quality is complex (Vos et al., 2013).

While the use of mobile monitoring data offers a cost-effective method of characterizing spatial variations in ambient UFPs over large geographic areas it is important to note several limitations. First, all of the data were collected on weekdays during morning and afternoon rush hour periods and as a result our model may overestimate ambient concentrations as evenings and weekends are expected to have lower levels. However, previous studies may underestimate UFP concentrations close to major highways as safety reasons often prohibit monitoring near these locations. Mobile monitoring is advantageous in this respect as it facilitates monitoring directly on roadways. In addition, our data were collected over a relatively short three-week period and may not completely capture seasonal variations in ambient UFP concentrations. Indeed, some evidence suggests ambient UFPs vary throughout the year with higher levels occurring during winter months and minimum values occurring during the summer (Johansson et al., 2007). This pattern is consistent with the known inverse association between ambient temperature and UFPs and the inclusion of temperature terms in our models may at least partially address this limitation. Finally, while we cannot rule out some contribution from "self-pollution" emitted by the mobile monitoring vehicles, these contributions likely do not explain the observed associations as all road segments were monitored with the same vehicle type and thus were likely similarly impacted by these emissions.

In general, road segments included in our analysis reflected a broad range of land use characteristics and ambient UFP/temperature levels varied substantially across segments; therefore, our model is likely generalizable to much of the greater Toronto area. However, one further limitation was the quantity of exposure data available for individual road segments and this might have decreased the precision of model estimates compared to longer monitoring campaigns (Klompmaker et al., 2015). This limitation could be addressed in future studies by increasing the number and duration of mobile monitoring campaigns conducted throughout the year; however, the mean duration of UFP monitoring for each road segment in this study was comparable to the 15-minute time period previously employed in Girona, Spain (Rivera et al., 2012). In addition, since model performance was comparable to those based on longer monitoring campaigns, our findings support the use of short-term repeated measurements in the development of similar models in the future.

Since all road segments were not monitored simultaneously, ambient temperature was used to account for temporal variations in ambient UFP concentrations between monitoring periods. This decision was based on existing evidence suggesting that ambient temperature is an important determinant of day-to-day changes in ambient UFPs (Alm et al., 1999; Kaur and Nieuwenhuijsen, 2009; Weichenthal et al., 2008, 2014; 2015). Regardless, this may be considered a limitation as previous studies have relied on fixed-site data for UFPs (Abernethy et al., 2013; Hoek et al., 2011) or NO_x (Rivera et al., 2012) to adjust for temporal variability and we cannot rule out a residual impact of temporal variation on our results. Indeed, while ambient temperature was a significant predictor of UFPs in simple linear models, including temperature terms in final multivariable models offered only modest improvements in R^2 and RMSE values. Nevertheless, analysis of within and between-segment variability suggested that spatial variability exceeded temporal variations in ambient UFPs and others have reported similar observations (Abernethy et al., 2013); therefore, we feel that this approach was justified given the absence of fixed-site UFP data and the known inverse relationship between ambient UFPs and ambient temperature. Despite this possible limitation, including temperature terms in the model is also advantageous as it adds a temporal component directly to the model allowing predictions to be made throughout the year as opposed to a single static surface. Indeed, historical ambient temperature data are readily available from Environment Canada and this feature may be particularly useful in large-scale studies interested in ambient UFPs during specific portions of the year.

5. Conclusions

We developed a land use regression model for ambient UFPs in Toronto, Canada. This model explained the majority of the spatial variability in ambient UFPs and provided unbiased estimates of ambient concentrations when applied to an external dataset. This model may be used to evaluate the chronic health effects of UFPs using population-based cohorts in the Toronto area.

Financial interests' declaration

None declared.

Acknowledgments

We would like to thank Dr. Daniel Crouse and Dr. Rick Burnett for their helpful comments in reviewing this manuscript. This work was supported by CHRP and Health Canada.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2015.04.011.

References

Abernethy, R.C., Allen, R.W., McKendry, I.G., Brauer, M., 2013. A land use regression model for ultrafine particles in Vancouver, Canada. Environ. Sci. Technol. 47, 5217–5225.

Alm, S., Jantunen, M.J., Vartiainen, M., 1999. Urban commuter exposure to particle matter and carbon monoxide inside and automobile. J. Expo. Anal. Environ. Epidemiol. 9, 237–244.

Crouse, D.L., Goldberg, M.S., Ross, N.A., 2009. A prediction-based approach to modelling temporal and spatial variability of traffic-related air pollution in Montreal, Canada. Atmos. Environ. 43, 5075–5084.

Crouse, D.L., Goldberg, M.S., Ross, N.A., Chen, H., Labreche, F., 2010. Postmenopausal breast cancer is associated with exposure to traffic-related air pollution in Montreal, Canada: a case-control study. Environ. Health Perspect. 118, 1578–1583.

Hoek, G., Beelen, R., Kos, G., Dijkema, M., Van Der Zee, S.C., Fischer, P.H., Brunekreef, B., 2011. Land use regression model for ultrafine particles in Amsterdam. Environ. Sci. Technol. 45, 622–628.

Hoek, G., Krishnan, R.M., Beelen, R., Peters, A., Ostro, B., Brunekreef, B., Kaufman, J.D., 2013. Long-term air pollution exposure and cardio-respiratory mortality: a review. Environ. Health 12, 43.

Hudda, N., Gould, T., Hartin, K., Larson, T.V., Fruin, S.A., 2014. Emissions from an international airport increase particle number concentrations 4-fold at 10 km downwind. Environ. Sci. Technol. 48, 6628–6635.

Jerrett, M., Finkelstein, M.M., Brook, J.R., Arain, M.A., Kanaroglou, P., Stieb, D., Gilbert, N.L., Verma, D., Finkelstein, N., Chapman, K.R., Sears, M.R., 2009. A cohort study of traffic-related air pollution and mortality in Toronto, Ontario, Canada. Environ. Health Perspect. 117, 772–777.

Johansson, C., Norman, M., Gidhagen, L., 2007. Spatial & temporal variations of PM10 and particle number concentrations in urban air. Environ. Monit. Assess. 127, 477–487.

Kaur, S., Nieuwenhuijsen, M., 2009. Determinants of personal exposure to $PM_{2.5}$, ultrafine particle counts, and CO in a transport microenvironment. Environ. Sci. Technol. 43, 4737.

Klompmaker, J.O., Montagne, D.R., Meliefste, K., Hoek, G., Brunekreef, B., 2015. Spatial variation of ultrafine particles and black carbon in two cities: results from a short-term measurement campaign. Sci. Total Environ. 508, 266–275.

Mehta, S., Shin, H., Burnett, R., North, T., Cohen, A.J., 2013. Ambient particulate air pollution and acute lower respiratory infections: a systematic review and implications for estimating the global burden of disease. Air Qual. Atmos. Health 6, 69–83.

Miller, M.R., Shaw, C.A., Langrish, J.P., 2012. From particles to patients: oxidative stress and the cardiovascular effects of air pollution. Future Cardiol. 8, 577–602.

Miller, M., 2014. The role of oxidative stress in the cardiovascular actions of particulate air pollution. Biochem. Soc. Trans. 42, 1006–1011.

Ostro, B., Hu, J., Goldberg, D., Reynolds, P., Hertz, A., Bernstein, L., Kleeman, M.J., 2015. Associations of mortality with long-term exposures to fine and ultrafine particles, species and sources: results from the California Teachers Study Cohort. Environ. Health Perspect. http://dx.doi.org/10.1289/ehp.1408565.

Pugh, T.A.M., Mackenzie, A.R., Whyatt, J.D., Hewitt, C.N., 2012. Effectiveness of green infrastructure for improvement of air quality in urban street canyons. Environ. Sci. Technol. 46, 7692–7699.

Rivera, M., Basagana, X., Aguilera, I., Agis, D., Bouso, L., Foraster, M., Medina-Ramon, M., Pey, J., Kunzli, N., Hoek, G., 2012. Spatial distribution of ultrafine particles in urban settings: a land use regression model. Atmos. Environ. 54, 657–666.

Sabaliauskas, K., Jeong, C.H., Yao, X., Reali, C., Sun, T., Evans, G., 2015. Development of a land-use regression model for ultrafine particles in Toronto, Canada. Atmos. Environ. 110, 84–92.

Shah, A.S.V., Langrish, J.P., Nair, H., McAllister, D.A., Hunter, A.L., Donaldson, K., Newby, D.E., Mills, N.L., 2013. Global association of air pollution and heart failure: a systematic review and meta-analysis. Lancet 382, 1039–1048.

Sun, Q., Hong, X., Wold, L.E., 2010. Cardiovascular effects of ambient particulate air pollution exposure. Circulation 121, 2755–2765.

Vos, P.E., Maiheu, B., Vankerkom, J., Janssen, S., 2013. Improving local air quality in cities: to tree or not to tree? Environ. Pollut. 183, 113–122.

Weichenthal, S., Dufresne, A., Infante-Rivard, C., Joseph, L., 2008. Determinants of ultrafine particle exposures in transportation environments: findings of an 8-month survey conducted in Montreal, Canada. J. Expo. Sci. Environ. Epidemiol. 18, 551–563.

Weichenthal, S., 2012. Selected physiological effects of ultrafine particle in acute cardiovascular morbidity. Environ. Res. 115, 26–36.

Weichenthal, S., Farrell, W., Goldberg, M., Joseph, L., Hatzopoulou, M., 2014. Characterizing the impact of traffic and the built environment on near-road ultrafine particle and black carbon concentrations. Environ. Res. 132, 305–310.

Weichenthal, S., Van Ryswyk, K., Kulka, R., Sun, L., Wallace, L., Joseph, L., 2015 Jan 6. In-vehicle exposures to particulate air pollution in Canadian metropolitan areas: the urban transportation exposure study. Environ. Sci. Technol. 49 (1), 597–605. http://dx.doi.org/10.1021/es504043a. Online Dec 3.

Zhu, Y., Hinds, W., Kinm, S., Shen, S., Sioutas, C., 2002. Study of ultrafine particles near a major highway with heavy-duty diesel traffic. Atmos. Environ. 36, 4233–4235.