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Assessing the transferability of landuse regression models for ultrafine particles across two Canadian cities

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HIGHLIGHTS

- Spatial transferability of LUR models between cities poorly characterized
- Transferability of UFP LUR models assessed across two Canadian metropolises
- Results support transferability between the 2 cities with a minor drop in model fit.
- Incorporating city-specific predictors improved the transferability of the models.
- Errors introduced by transferability lower than those introduced by sampling design

GRAPHICAL ABSTRACT

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ABSTRACT

Land use regression (LUR) models have been increasingly used to predict intra-city variations in the concentrations of different air pollutants. However, limited research assessing the transferability of these models between cities has been published to date. In this study, LUR models were generated for Ultra-Fine Particles (UFP) (<0.1 um) using data collected from mobile monitoring campaigns in two Canadian cities, Montreal and Toronto. City-specific models were first generated for each city before the models were transferred to the second city with and without recalibration. The calibrated transferred models showed only a slight decrease in performance, with the coefficient of determination (R^2), dropping from 0.49 to 0.36 for Toronto and from 0.41 to 0.38 for Montreal. Transferring models between cities with no calibration resulted in low R^2; 0.11 in Toronto and 0.18 in Montreal. Moreover, two additional models were generated by combining data from the two cities. The first combined model (CM1) assumed a spatially invariant effect of the predictors, while the second (CM2) relaxed the assumption of spatial invariance for some of the model coefficients. The performance of both combined models (R^2 ranged between 0.41 for CM1 and 0.43 for CM2; root mean squared error (RMSE) ranged between 0.34 for CM1 and 0.33 for CM2) was found to be on par with the Toronto city-specific model and outperformed the Montreal model. The results of this study highlight that the UFP LUR models appear to support transferability of model structures between cities with similar geographical characteristics, with a minor drop in model fit and predictive skill.

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1. Introduction

Elevated particulate matter (PM) concentrations of different sizes have been found to highly correlate with increased incidence of cardiovascular (Dehbi et al., 2017; Delfino et al., 2010; Franchini and Mannucci, 2012; Gan et al., 2012; Vanos et al., 2014) and respiratory (Beelen et al., 2008; Iii et al., 2002; Int Panis et al., 2010; Vanos et al., 2014) diseases. Recently, exposure to high concentrations of ultra-fine particles (UFPs, with diameter < 100 nm) has gained attention due to the ability of UFPs to pass through the lungs and affect other organs (Int Panis et al., 2010). UFPs have also been associated with elevated levels of inflammatory blood biomarkers in people living near major highways (Lane et al., 2015). More recently, exposure to UFPs has been associated with the incidence of various chronic illnesses (Bai et al., 2018; Goldberg et al., 2018; Goldberg et al., 2017; Weichenthal et al., 2017). The main source of UFPs has been linked to mobile combustion sources, with UFPs typically constituting around 90% or more of the particle number count (PNC) in areas highly influenced by vehicle emissions (Morawska et al., 2008). As daily commutes have become longer due to traffic congestion and as more people reside in the vicinity of major roads with high traffic densities, the rate of human exposure to UFPs has increased drastically, with the bulk of exposure happening during commuting hours (de Kluizenaar et al., 2017; Ham et al., 2017; Peters et al., 2014). This brings about the importance of developing robust UFP predictive models for urban areas.

The performance of land use regression (LUR) models has been well established in the literature to be robust for modeling and predicting the levels of traffic related air pollutants (Hoek et al., 2008). LURs have also been found to perform as well as, and even outperform, other air pollution modeling methods such as dispersion models (de Hoogh et al., 2014; Jerrett et al., 2005). They provide the advantage of being able to predict intra-urban variability of air pollutant concentrations at a micro scale level without the need for detailed spatio-temporal data inputs (Jerrett et al., 2005). Several LUR models have been developed for predicting UFP concentrations in North American (Abernethy et al., 2013; Sabaliauskas et al., 2015; Weichenthal et al., 2016a; Weichenthal et al., 2016b) and European (Cattani et al., 2017; Hoek et al., 2011; Kerckhoffs et al., 2017; Rivera et al., 2012; van Nunen et al., 2017; Wolf et al., 2017) cities.

Assessing the transferability of calibrated air pollution LUR models between cities is a crucial research question, given the logistical difficulties and costs associated with establishing sampling programs for each city. To date, only a few studies have looked at the potential transferability of LUR models for different air pollutants (Table 1). Conclusions from these studies have differed in their assessment of the adequacy of model transfer between cities. Several studies have shown that transferability only resulted in a small drop in the performance of the air quality LUR models. In a study by Allen et al. (2011), the performance of LUR models for nitrogen dioxide (NO_2) and nitrogen oxide (NO) dropped when transferred to another city, but the transferred models still explained more variability than simple binary or continuous road proximity models, which are commonly used in epidemiological studies. This showed that transferring NO_2 and NO LUR models between cities could provide a cost-effective and informative approach for assessing exposure to traffic related air pollutants in poorly monitored areas with different geographic conditions. Another study showed that the transferability of a NO_2 LUR model between cities could be significantly improved if the model coefficients were recalibrated for each city (Poplawski et al., 2009). Other studies have also reported a good performance of LUR models when the model transfer was between localities with similar geographic conditions, with only a slight drop in the prediction capability of the models (Vienneau et al., 2010; Wang et al., 2014). Nevertheless, some studies have concluded that LUR models are intrinsically area specific and as such tend to have low predictive skills when they are applied in regions with different geographic conditions (Briggs et al., 2000; Jerrett et al., 2005; Marcon et al., 2015).

Table 1
Previous LUR transferability studies.

Source	Pollutant	Location	Method	Conclusions
Briggs et al. (2000)	NO_2	4 cities in the UK	• Developed and calibrated a model for Huddersfield • Transferred model structure to 3 other UK cities. • Model coefficients were re-calibrated for each city using a subset of the collected data	R^2 dropped from 0.82 to 0.60, 0.61 and 0.76 (calibration done based on 10 sites only)
Marcon et al. (2015)	NO_2	Veneto, Italy	Developed and calibrated a model for Veneto	R^2 of predicted versus observed dropped from 0.75 to 0.52 (in non-urban areas), and to 0.18 in urban areas
Vienneau et al. (2010)	NO_2 and PM_{10}	UK and Netherlands	• Model used for predictions in the city of Verona without calibration (model transferred both in terms of structure and coefficients) • Developed and calibrated a pooled model for the UK and the Netherlands • Developed and calibrated country-specific models • Tested the transferability of the model structures between the two countries while calibrating the model coefficients based on country data	R^2 for the NO_2 dropped from 0.86 to 0.44 when the Netherlands model was recalibrated to the UK data R^2 for the NO_2 remained the same when the UK model was transferred and calibrated with the Netherlands data UK model calibrated with 156 sites for NO_2, and 93 sites for PM_{10}, while Netherlands model was less robust with 44 sites for NO_2, and 19 sites for PM_{10} The drop was more significant for both the UK and the Netherlands PM_{10} models
Allen et al. (2011)	NO and NO_2	Edmonton and Winnipeg, Canada	• Developed city specific models • Tested transferability of the models between cities without recalibration	R^2 of predicted versus observed dropped from (0.84–0.81) to (0.37–0.52) for NO_2 and from (0.55–0.56) to (0.24–0.41) for NO
Poplawski et al. (2009)	NO_2	Vancouver, Victoria and Seattle	• Tested the transferability of a calibrated model for Vancouver on predicting NO2 levels in Victoria and Seattle • Transferability was tested with and without city-level calibration	R^2 of predicted versus observed ranged from 0.51 to 0.33 for Victoria and Seattle respectively R^2 for models with local calibration was (0.58–0.65) for Victoria and Seattle respectively

In this study, we look at the transferability of LUR models developed to model UFP levels in two Canadian cities, Toronto and Montreal. The importance of this study is in its ability to limit biases that have affected previous air quality LUR studies, such as differences in geographic conditions (Briggs et al., 2000; Marcon et al., 2015), missing predictors between cities (Allen et al., 2011), differences in the scale of the regions (Marcon et al., 2015; Wang et al., 2014) and differences in the sampling protocols (Poplawski et al., 2009). This study also limits any bias caused by different fleet compositions or urban planning since both cities are located in the same country and are similar in that regard. In this paper, city specific, as well as pooled models were developed and calibrated before they were used to assess changes in the model coefficients between cities as well as to monitor the overall performance of the transferred models in predicting the spatial variability of UFP levels within a city.

2. Methods

2.1. Study area and data collection

Toronto and Montreal have a relatively similar land use given that their areas consist of dense urban regions, with population densities of 4000 and 2700 inhabitants/km^2 respectively (Statistics Canada, 2016). A main highway system borders each city with major North-South and East-West arterial roads crossing them. Moreover, both cities have a long waterfront; Toronto's waterfront is along Lake Ontario, south of the city while Montreal is an island, surrounded by the St Lawrence River. Both cities have a humid continental climate, with warm humid summers and cold winters.

Data were collected in both cities through an intensive monitoring campaign that made use of mobile measurements conducted on bicycles. Data collection in Montreal took place between June and July in 2012, covering 23 days. Measurements were taken along 25 predefined cycling routes using mobile air pollution monitoring instruments placed in backpacks that were carried by the cyclists. In total, around 475 km of unique roadways were covered, amounting to 4058 road segments (Fig. 1). Due to missing site-specific information on key model predictors, 1512 road segments in Montreal were excluded; as such 2279 segments were potentially useable in the development of the LUR models. Measurements were divided into 2 sampling periods, morning sampling (starting 8 a.m.) and afternoon sampling (starting 2:30 p.m.), and were mainly focused during traffic rush hours. Project bicycles were mounted with condensation particles counters (TSI, CPC Model 3007) that measured UFP. GPS units (Garmin, Edge 800) were also used to geolocate each sampling point. UFP data were measured in particles per cubic centimeter (#/cm^3). In addition, ambient temperature was measured concurrently. All instruments had a measurement frequency of 1 Hz. The Toronto monitoring campaign followed the same protocol as that for Montreal. It extended between May and August 2016, covering 39 days in total. In total, 3791 different road segments were visited (Fig. 1). Instead of the condensation particle counters (TSI, Model 3007), the Toronto campaign used DiSCminis (Testo Instruments). All instruments were collocated with reference monitoring stations and correction factors were applied. Nevertheless, the fact that the two studies were conducted 4 years apart and that the mobile equipment used were different may weaken our conclusions about the transferability of the UFP models. Yet, the relative importance of these differences in the monitoring campaigns is expected to be minimal when compared to inherent differences between the two cities.

Concentrations of UFP for each road segment were averaged across the multiple visits. By averaging the concentrations, we minimize the effect that temporal predictors (such as day of the week and time of day) might have had on the measured UFP readings. Averaging also minimizes the impacts of meteorological variables such as temperature and relative humidity. In addition, all road segments with <4 visits during the sampling campaign were excluded from the LUR development in an effort to improve the robustness of the generated model. The choice of using a cutoff value of 4 visits in both cities was based on a sensitivity analysis that looked at maximizing the performance of the models (largest R^2) without loss of significant spatial coverage. Minet et al. (2017) also found that excluding sampling sites with <4 measurements led to better models while also preserving spatial heterogeneity. This decreased the number of road segments from 3791 to 2415 in Toronto and from 2279 to 1529 in Montreal. Natural log-transformation of UFP concentrations was conducted in order to meet the assumptions of parametric regression. Adopting log-transformation is typical for environmental variables given that most variables are log-normally distributed (Ott, 1994). A summary of the two sampling campaigns is presented in Table 2. Detail on both campaigns can be found in Farrell et al. (2016) and Minet et al. (2018) for Montreal and Toronto respectively.

2.2. Statistical analysis

2.2.1. Potential predictors of UFP

Potential predictors for the development of UFP LUR models were determined from the literature. Previous studies have reported a strong correlation between land use predictors and measured UFP levels (Table 3). In addition, both road density and distance to pollution sources (industrial and airports) were found to be strong predictors of ambient UFP concentrations (Table 3). Based on previous studies, two types of predictors were generated for Toronto and Montreal, namely

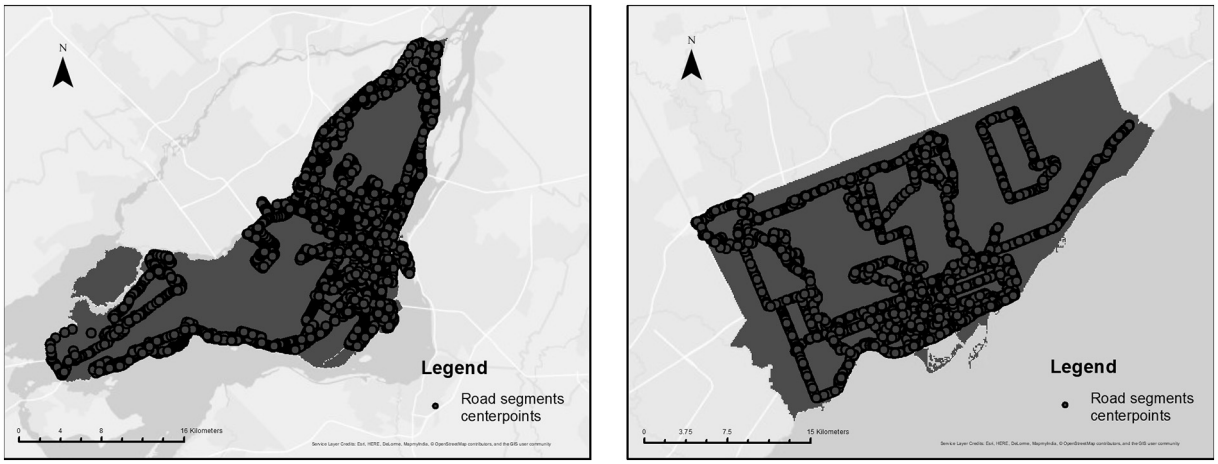

Fig. 1. Montreal road segments midpoints on the left and the Toronto road segments midpoints on the right.

Table 2
Comparison of monitoring campaigns.

City	Date of campaign	# of weekdays	Equipment used	# of roads segments	Average length of segment	Average # of visits/segment (range)	Average time spent in segment
Montreal	June–July 2012	23	TSI, CPC Model 3007	4058	117 m	4.2 (1–53)	45.3 s
Toronto	May–August 2016	39	Testo, DiSCmini	3791	150 m	4.3 (1–36)	24.6 s

1) time variable predictors that included the meteorological data (wind speed (m/s), temperature (°C), relative humidity (%), and on-road NOx emissions (g), and 2) the temporally invariant predictors. The latter included both distance to pollution sources (airports, highways, industrial point sources) as well as land-use variables such as length of roads (m), length of major roads (m), length of highways (m), building footprint (m^2), commercial area (m^2), governmental area (m^2), industrial areas (m^2), length of bus routes (m), number of bus stops, open area (m^2), park area (m^2), residential area (m^2), waterbody area (m^2), average building height (m), and length of rail (m). All land-use variables were reported in terms of their occurrence within a predefined buffer. Buffers were generated from the center-point of each road segment using ArcGIS 10.5.1 (ESRI, 2017). The radii of the generated buffers ranged from 25 m up to 1000 m. In an attempt to remove potential problems caused by autocorrelation, the correlations between all buffers of the same predictor were assessed using Pearson correlation. Given the high correlation found between the different buffer distances, only the buffered predictor that had the highest correlation with the measured UFP readings were kept.

2.3. Model development

2.3.1. City-specific models

Multiple linear regression models were developed to predict UFP concentrations for each city separately. The city-specific multiple linear regression models were developed using a supervised forward selection procedure. The final models were selected based on their Akaike's Information Criterion (AIC) (Akaike, 1974). At each step, the predictor resulting in the model with the lowest AIC value (i.e. the most parsimonious model) was added. Each selected predictor was plotted alone against the UFP concentrations to ensure that the sign of the model variable coefficient matched the univariate relationship. Change of sign is an indication of multi-collinearity issues. Moreover, any predictors that resulted in a change in the sign of a previously added predictor once included in the model was discarded to avoid overfitting or the introduction of strong interactions between predictors. Finally, predictors that resulted in other predictors becoming insignificant (with p-value < 0.1) were also excluded. Problems associated with collinearity were continuously assessed through tracking the variance inflation factors (VIF). Predictors with VIF values >5 were removed from the final model. Final residual diagnostic tests were applied to check for influential observations (Cook's D), heteroskedasticity, and non-normality. Additionally, the spatial autocorrelation of the residuals was assessed using Moran's I. Moran's I values vary between −1 and 1, with a value of 0 signifying no spatial autocorrelation, values close to −1 signifying dispersion in the residuals, and values close to 1 signifying clusters in the residuals. The percent bias (PBIAS) was also calculated as a measure of the tendency of the models to overpredict or underpredict. All statistical analyses were performed using the R software (R Core Team, 2015).

Model performance was evaluated using the R^2, the adjusted R^2 and the root mean squared errors (RMSE). A 4-fold (k-fold) cross-validation was also performed on the developed models, using the DAAG package (Maindonald and Braun, 2014) in the software R (R Core Team, 2015). Cross validation assesses the predictive power of the generated models.

2.3.2. Transferred models

Initially, city specific models were developed and validated. Then each of the two developed city-specific models was transferred without recalibrating their coefficients to the second city; and the models were

Table 3
Predictors used in previous UFP LUR models.

Source	Location	Meteorological predictors	Land-use predictors	Traffic predictors	Other predictors
Hoek et al. (2011)	Amsterdam, Netherlands	–	Port area, building density	Traffic intensity	–
Rivera et al. (2012)	Girona, Spain	–	Building area	Distance to highway, length of bus route, number of vehicles, length of major roads	–
Abernethy et al. (2013)	Vancouver, Canada	–	Governmental area, residential area, restaurant density	Road length, vehicle density, truck route length, truck count, distance to intersection, distance to major road, distance to highway	Latitude, distance to port
Sabaliauskas et al. (2015)	Toronto, Canada	–	Residential area, industrial area	Length of major roads, length of expressways	Population density
Weichenthal et al. (2016a)	Montreal, Canada	Temperature, Wind speed	Park area, Open area	Length of roads, Length of rail	Annual NO_x, Population density
Weichenthal et al. (2016b)	Toronto, Canada	Temperature	Park area, Open area, Commercial area, number of trees	Distance to highway, distance to major road, distance to bus route, length of bus route, number of intersections	Distance to airport, distance to central business district
Cattani et al. (2017)	Rome, Italy		Green area	Distance to major road, traffic intensity,	Population
Wolf et al. (2017)	Augsburg, Germany	–	Building number, green area, semi natural area, industrial area	Traffic load	–
van Nunen et al. (2017)	6 European cities	–	Green area, restaurants, industrial area	Traffic load	Population, distance to port, distance to airport
Kerckhoffs et al. (2017)	3 Dutch cities	–	Industrial area, port area, urban green area, residential area	Traffic intensity, heavy traffic intensity, length of major roads	Population density

Table 4
UFP concentrations in Toronto and Montreal.

City	UFP concentrations in (parts/cm^3)				
	Mean	Standard deviation	Min	Max	Interquartile range
Toronto	23,059	12,493	5831	241,547	13,522
Montreal	20,578	8188	4518	53,105	10,310

assessed for their predictive skill. This was achieved by comparing the model predictions against the actual observations in the concerned city. Additionally, we assessed the performance of the models in the event that the transferability was restricted to fixing the model structure, while allowing the model coefficients to change between cities. This allows the models to recalibrate based on the city-specific data, while allowing the model structure to transfer. It is expected that the latter method would generate better predictions as compared to the transferable models without calibration, as was the case for NO_2 developed LUR models (Poplawski et al., 2009). It should be noted that changes in the coefficients of the models between the two cities while fixing the model structure may reflect differences in the sources, fate and transport of UFP between Toronto and Montreal. Model assessment was determined in terms of the R^2, PBIAS, and RMSE calculated from the predicted versus observed concentrations for each city.

2.3.3. Combined models

Combined models were generated by pooling the data of Montreal and Toronto together and fitting a single model for both cities. The first combined model assumed that the relationship between the predictors and the measured UFP concentrations would remain spatially invariant between the two cities; yet the model allowed for city-level differences in the background UFP levels (model fit with two intercepts). The second combined model relaxed the assumption of spatial invariance for some of the model coefficients and thus allowed the relationships of some predictors to vary between the two cities. For example using distance to airport as a predictor, one would expect that the effect of the variable on UFP concentrations may show spatial differences between the two cities given that the cities have different air traffic and the location of their airports are different with regards to the city center. Prediction surfaces were then generated for each city using the two pooled models, and the results were compared with surfaces generated from the city-specific models. Note that the generated exposure UFP predicted surfaces represent typical summer averaged UFP concentrations that cyclists and pedestrians are expected to be exposed to in the two cities during peak hours. Surfaces more conducive towards epidemiological studies would require a longer sampling period and possibly a greater spatial extent. Yet, the proposed modeling framework and the conclusion made in this work should be readily adaptable for such studies.

3. Results

3.1. Descriptive statistics

UFP concentrations in the city of Toronto (mean = 23,059 particles/cm^3) were found to be slightly higher than those in Montreal (mean = 20,578 particles/cm^3). UFP concentrations in Toronto ranged between 5831 and 241,547 particles/cm^3, while those in Montreal ranged between 4518 and 53,105 particles/cm^3 (Table 4). The distribution of UFP concentrations in both cities was found to follow the lognormal distribution (Fig.2).

3.2. The Montreal city-specific model (MM)

The city-specific model for Montreal (MM) explained 41.5% of the variability in the observed UFP concentrations. Significant predictors included the length of major roads within a 100 m buffer, commercial area within a 300 m buffer, waterbody area within a 300 m buffer, NOx daily emissions within a 300 m buffer, wind speed, industrial area within a 300 m buffer, and building footprint within a 300 m buffer (Table 5). The length of major roads within a 100 m buffer was found to have a positive effect on the concentrations of UFP, whereby a 100 m increase in roads would result in 3.1% increase in UFP concentrations. Increases in the waterbody area and wind speed tended to decrease UFP concentrations. Building footprint, NOx concentrations and industrial area had

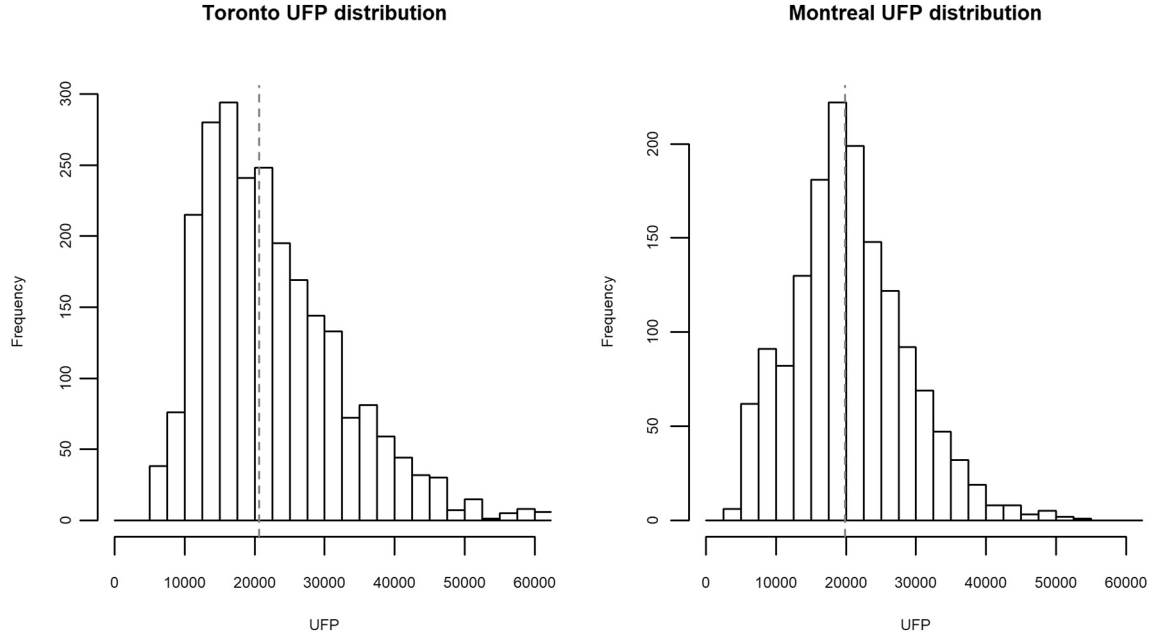

Fig. 2. Distribution of UFP concentrations in (a) Toronto and (b) Montreal, with the gray dashed line signifying the median of the UFP concentrations (20,608.36 parts/cm^3 for Toronto and 19,844.18 parts/cm^3 for Montreal.

Table 5
Montreal developed UFP LUR model structure calibrated for Montreal (MM model) and the Toronto calibrated and transferred Montreal model (TCMM).

	Montreal UFP LUR Model (MM)	Toronto model based on transferring and recalibrating the Montreal model (TCMM)
	$R^2 = 0.410$; Adj $R^2 = 0.410$; 4-fold CV $R^2 = 0.401$; RMSE = 0.340	$R^2 = 0.360$; Adj $R^2 = 0.360$; 4-fold CV $R^2 = 0.353$; RMSE = 0.370
Predictors	Coefficients (95% confidence interval)	Coefficients (95% confidence interval)
Intercept	10.70 (10.52, 10.86)	10.65 (10.56, 10.73)
Commercial area (Km^2 in buffer = 300 m)	2.85 (1.52, 4.18)	−0.25 (−1.61, 1.1)
Water area (Km^2 in buffer = 300 m)	−2.19 (−3.53, −0.85)	−3.01 (−4.17, −1.83)
NOx daily average emissions (average within buffer = 300 m)	2.58×10^{-7} (1.89×10^{-7}, 3.25×10^{-7})	1.76×10^{-8} (7.49×10^{-9}, 2.77×10^{-8})
Average windspeed (Km/h)	-0.089 (-9.83×10^{-2}, -8.02×10^{-2})	-0.052 (-5.63×10^{-2}, -4.71×10^{-2})
Major roads (Km in buffer = 100 m)	0.31 (0.22, 0.40)	0.46 (0.32, 0.60)
Industrial area (Km^2 in buffer = 300 m)	0.846 (0.52, 1.17)	−1.75 (−2.32, −1.18)
Building area (Km^2 in buffer = 300 m)	3.75 (2.82, 4.68)	11.50 (10.40, 12.60)

a positive effect on UFP concentrations. The model didn't show any significant biases with a PBIAS value of 0 (Fig. 3a). In addition, the model didn't show any spatial autocorrelation in the residuals with a Moran's I value of 0.09 (close to 0).

Transferring the MM to Toronto without recalibration resulted in a poor model performance. The transferred model (referred to as the Non-Calibrated Montreal Model = NCMM) tended to consistently under-predict the observed UFP concentrations (PBIAs = −6.1%) (Fig. 3c). When the model predictions were compared to the observed values, it was found that the model only explained 11.1% of the UFP variability in Toronto ($R^2 = 0.11$) (Fig. 3). Additionally, the model showed large spatial clustering with a Moran's I value of 0.9. Upon recalibrating the structure of the Montreal model with the Toronto data (referred to as the Toronto Calibrated Montreal Model = TCMM), the performance of the model significantly improved and the R^2 increased to 0.36. Recalibrating the model also removed the bias found in the previous model and the model no longer showed a tendency to under-predict UFP levels (PBIAS = 0) (Fig. 3b). Moreover, the transferred recalibrated model did show that the effects of the commercial areas were largely insignificant (p-value > 0.1) for the case of Toronto. The sign of the remaining coefficients didn't change except for the percent industrial areas within a 300 m buffer; unlike Montreal the TCMM indicated that UFP levels in Toronto tended to decrease as the percentage of industrial areas within the 300 m buffer increased. The TCMM is summarized in Table 5. The residuals of the TCMM showed some evidence of spatial clustering, with a Moran's I value of 0.477.

3.3. The Toronto city-specific model (TM)

The city specific UFP LUR model for Toronto (TM) performed better than that developed for Montreal (MM) and explained 48.9% of the variability in the observed UFP concentrations in the city (Table 6). Significant predictors included the length of highways within a 500 m buffer, building footprint within a 50 m buffer, distance to airport, wind speed, length of major roads within a 100 m buffer, waterbody area within a 300 m buffer and park area within a 300 m buffer. The length of highways within a 500 m buffer, the building footprint within a 50 m buffer and the length of major roads within a 100 m buffer were found to have a positive effect on UFP concentrations. Increases in the distance to airports, wind speed, waterbody area within a 300 m buffer and park area within a 300 m buffer tended to decrease UFP concentrations. The model didn't show any biases with a PBIAS value of 0 (Fig. 4a). Yet, the model residuals showed evidence of clustering, with a Moran's I value of 0.42. (See Fig. 5.)

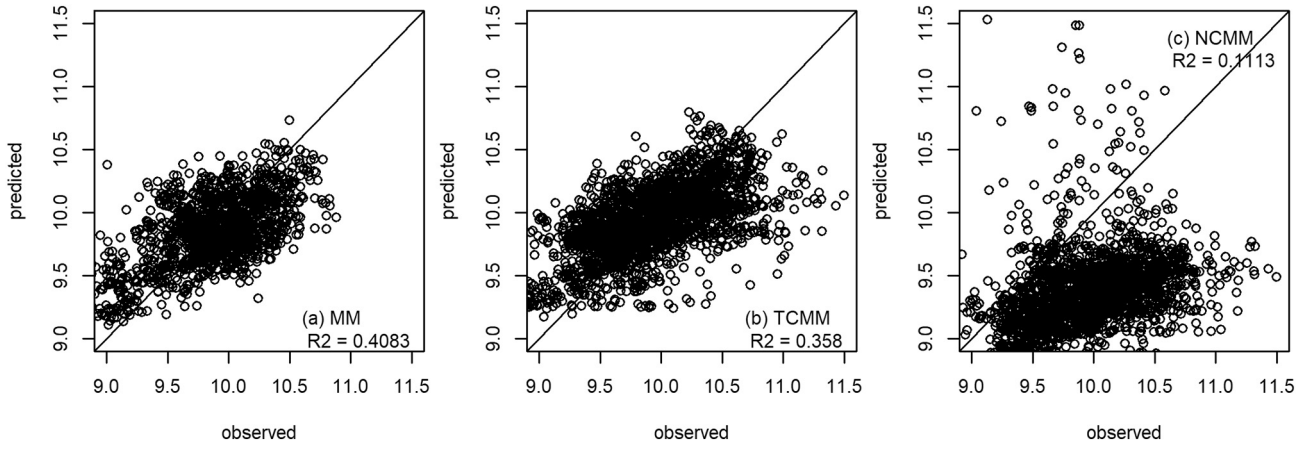

Fig. 3. Predicted vs observed plots (a) Montreal UFP LUR Model (MM) (b) Montreal model transferred and recalibrated with Toronto data (TCMM) (c) Montreal model transferred to Toronto without recalibration (NCMM). Diagonal line represents the 1:1 line.

Table 6
Toronto developed UFP LUR model structure calibrated for Toronto (TM model) and the Montreal calibrated and transferred Toronto model (MCTM).

	Toronto UFP LUR Model (TM)	Montreal model based on transferring and recalibrating the Toronto model (MCTM)
	$R^2 = 0.489$; Adj $R^2 = 0.488$; 4-fold CV $R^2 = 0.485$; RMSE = 0.330	$R^2 = 0.383$; Adj $R^2 = 0.380$; 4-fold CV $R^2 = 0.370$; RMSE = 0.347
Predictors	Coefficients (95% confidence interval)	Coefficients (95% confidence interval)
Intercept	10.95 (10.87, 11.03)	10.77 (10.6, 10.95)
Highways (Km in buffer = 500 m)	1.62×10^{-2} (3.23×10^{-3}, 2.92×10^{-2})	4.59×10^{-2} (3.27×10^{-2}, 5.91×10^{-2})
Building area (Km^2 in buffer = 50 m)	82.0 (57.0, 107.0)	52.3 (36.7, 67.9)
Distance to airport (Km)	-4.0×10^{-2} (-4.29×10^{-2}, -3.72×10^{-2})	2.83×10^{-2} (2.09×10^{-2}, 3.57×10^{-2})
Average wind speed (Km/h)	-0.042 (-0.046, -0.038×10^{-2})	-0.10 (-0.11, -0.092)
Major roads (Km in buffer = 100 m)	0.861 (0.73, 0.99)	0.38 (0.30, 0.47)
Waterbody area (Km^2 in buffer = 300 m)	-4.62 (-5.71, -3.53)	-3.73 (-5.05, -2.41)
Park area (Km^2 in buffer = 300 m)	-9.15 (-12.1, -6.18)	-7.43 (-11.3, -3.56)

Transferring the Toronto model (TM) to Montreal without recalibration resulted in a poor model that could only explain 18.1% of the variability observed in the UFP concentrations ($R^2 = 0.18$) (Fig. 4c). The model (referred to as the Non-Calibrated Toronto Model = NCTM) tended to over-predict the concentrations of UFP with a PBIAS of 1.6%. The model residuals showed little spatial autocorrelation, with a Moran's I value of 0.14. Upon recalibrating the model coefficients of the Toronto model with the Montreal data (referred to as the Montreal Calibrated Toronto Model = MCTM), the performance of the model significantly improved and the R^2 increased to 0.38. Recalibrating the model also removed the positive bias found in the previous model (PBIAS = 0) (Fig. 4b). Nevertheless in the recalibrated model, the effect of the distance to airport on UFP levels changed sign and became positive. The signs of the remaining coefficients didn't change, however their values were found to be different than those for Toronto. In addition, the residuals of the calibrated model didn't show any spatial autocorrelation with a Moran's I value of 0.01. The model structure of the MCTM is summarized in Table 6.

3.4. Combined models

Two different model structures were adopted and fitted to the combined data from the two cities. The first allowed for the use of city as a categorical predictor (CM1). This model structure allows for background UFP levels to be different by city; but it assumes that the effects of the predictors on the UFP levels remain invariant between the two cities (Table 7). The performance of the model was acceptable with an R^2 of 0.40. Overall, PBIAS was found to be 0, with both Montreal and Toronto predictions showing 0 bias. The model indicates that overall the increases in distance to airport, wind speed, the waterbody area with a 300 m buffer, the park area in 300 m buffer, and distance to major road all had a negative effect on UFP concentrations, whereas the increases in the number of bus stops within a 200 m buffer, the open space areas within a 100 m buffer, as well as the building area within a 300 m buffer had a positive effect on the observed UFP levels. The model intercepts for the two cities were found to be statistically different, with the concentrations in Toronto being slightly higher (13% higher) than those expected in Montreal under the same conditions. The model shows that the relative impact of distance to major roads played a much more important role in modulating the UFP levels in both cities. Overall, the median UFP levels decreased by 3% for every increase of 1 km distance between a site and the airport, while an increase of 1 km distance between a site and the nearest major road resulted in a 16% drop in the UFP levels. With respect to the effects of the land use on the UFP concentrations, it was found that the largest statistically significant impact was attributed to the presence of buildings within a 300 m buffer of a site. Model predictions showed that on average a site that had 100% of its 300 m buffer area occupied by buildings had on average

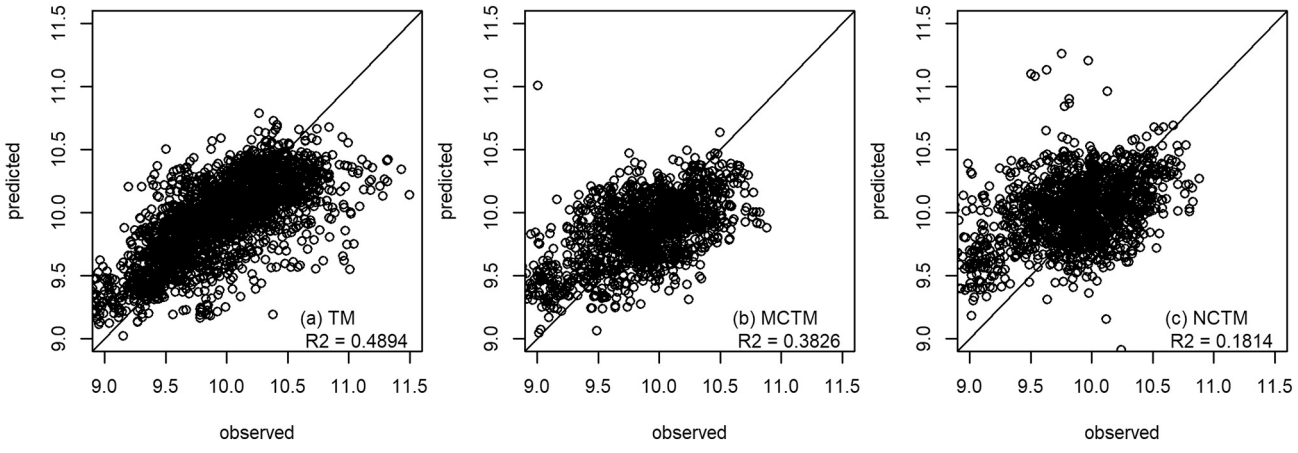

Fig. 4. Predicted vs observed plots (a) Toronto UFP LUR Model (TM) (b) Toronto model transferred and recalibrated with Montreal data (MCTM) (c) Toronto model transferred to Montreal without calibration (NCTM). Diagonal line represents the 1:1 line.

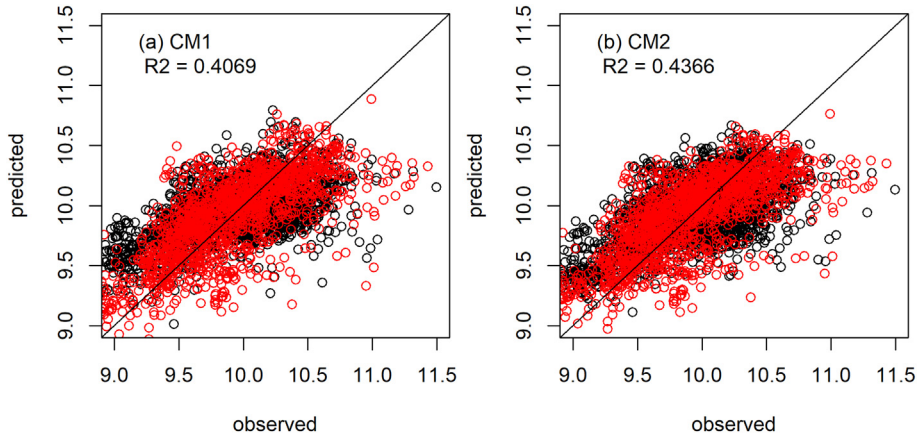

Fig. 5. Performance of the two combined models. a) The left panel shows the results from the combined model that only allowed for different UFP background levels between the two cities (CM1). b) The right panel shows the results from the pooled model that allowed for city differences in both UFP background levels as well as in the relationships between the predictors and the UFP levels (CM2). Performance is assessed in terms of agreement between predicted and observed results for both cities (open black circle = Toronto; open red circle = Montreal).

77.5% higher UFP levels as compared to a site that had no buildings within the same buffer distance. Conversely, the presence of water bodies resulted in the biggest drop in UFP concentrations with regards to the prevailing land uses around a site. On average the UFP levels at a site that had 100% water within its 300 m buffer area were 40% lower than those for a site that had no water bodies within its 300 m buffer. Note that with respect to the presence of open areas within a 100 m buffer of a site, model results showed that on average a site that has 100% open areas within its immediate 100 m buffer tended to have 16% higher UFP levels as compared to a site with no open areas within the same buffer distance.

The second model structure allowed for the inclusion of interaction terms, whereby the rate at which each predictor affects the UFP concentrations can vary by city (CM2). The second model performed slightly better than the first, explaining 43.7% of UFP variability across both cities, while the first model only explained 40.7% of the variability (Table 7). This shows that the magnitude of some predictors on UFP levels does vary from city to city; yet the overall impact of using interaction terms in CM2 was relatively small with regards to model fit (check supplementary material). Overall, the model's PBIAS was found to be 0, with both Montreal and Toronto predictions showing 0 bias. Interestingly, all the predictors that were identified in the first model structure (CM1) were also found to be statistically significant in the second model (CM2) and had the same signs. The statistically significant differences in the model coefficients between the two cities included the effects of building area coverage within a 300 m buffer, the percentage of open areas within a 100 m buffer, the distance to airport, and the wind speed. Overall, UFP levels were found to be more sensitive to building footprint within a 300 m buffer in Montreal as compared to Toronto. On average, sites in Montreal that had buildings covering 100% of their 300 m buffer area were expected to have around 51% higher UFP levels as compared to similar sites in Toronto. Similarly, UFP levels in Montreal were found to be more sensitive to the presence of open space as compared to Toronto. On average, sites in Montreal that had open space covering 100% of their 100 m buffer area were expected to have 41% higher UFP levels as compared to similar sites in Toronto. With regards to distance to airport, the UFP levels in Toronto dropped at a rate of 4% for each 1 km increase in distance, while levels dropped on average only by 1% for each 1 km increase in distance in the case of Montreal. This indicates that the effect of proximity to the main airport appears to have a

Table 7
UFP LUR models developed by combining the data from the city models.

	Combined model with city as categorical predictor (CM1)	Combined model with interactions (CM2)
	$R^2 = 0.407$; Adj $R^2 = 0.406$; 4-fold CV $R^2 = 0.404$; RMSE = 0.338	$R^2 = 0.437$; Adj $R^2 = 0.435$; 4-fold CV $R^2 = 0.432$; RMSE = 0.329
Predictors	Coefficients (95% confidence interval)	Coefficients (95% confidence interval)
Intercept Toronto	11.2 (11.13, 11.29)	11.1 (11.02, 11.18)
Intercept Montreal	11.08 (10.99, 11.17)	11.297 (11.10, 11.48)
Waterbody area (Km^2 in buffer = 300 m)	−1.78 (−2.18, −1.38)	−2.02 (−2.41, −1.62)
Park area (Km^2 in buffer = 300 m)	−0.48 (−0.63, −0.34)	−0.56 (−0.70, −0.42)
Number of bus stops (# in buffer = 200 m)	7.08×10^{-3} (3.23×10^{-3}, 1.09×10^{-3})	7.543×10^{-3} (3.79×10^{-3}, 1.13×10^{-2})
Distance to major road (Km)	−0.17 (-2.35×10^{-1}, -9.85×10^{-2})	−0.19 (−0.25, −0.12)
Building area (Km^2 in buffer = 300 m)	2.03 (1.65, 2.4)	T: 1.25 (0.83, 1.68) M: 2.71 (1.98, 3.44)
Open area (Km^2 in buffer = 100 m)	4.85 (3.57, 6.12)	T: 3.96 (2.66, 5.27) M: 14.85 (10.27, 19.44)
Distance to airport (Km)	-3.09×10^{-2} (-3.35×10^{-2}, -2.82×10^{-2})	T: -3.64×10^{-2} (-3.92×10^{-2}, -3.35×10^{-2}) M: -1.01×10^{-2} (-1.70×10^{-2}, -0.31×10^{-2})
Windspeed (Km/h)	-5.85×10^{-2} (-6.23×10^{-2}, -5.46×10^{-2})	T: -4.70×10^{-2} (-5.11×10^{-2}, -4.28×10^{-2}) M: -9.71×10^{-2}(-10.59×10^{-2}, -8.82×10^{-2})

longer lasting effect on UFP levels in Montreal as compared to Toronto. Nevertheless, the relative impact of wind in depressing the UFP levels in both cities was found to be more pronounced in Montreal as compared to Toronto. In Montreal, UFP levels dropped on average by 9% for each 1 Km/h increase in windspeed; levels in Toronto on average dropped by <5% for a similar increase in windspeed. The residuals of both CM1 and CM2 showed slight clustering in both cities, with the Toronto residuals showing more clustering in both models. Moran's I values were 0.127 and 0.432 for CM1 (for Montreal and Toronto respectively) and 0.163 and 0.437 for CM2 (for Montreal and Toronto respectively). Overall, the performance of the CM1 was found to be slightly worse than the performance of the two city specific UFP LUR models (MM and TM) with regards to R^2. The performance of the CM2 exceeded the performance of the Montreal city-specific model, the MCTM, as well as the TCMM and it was only slightly worse than that of the Toronto city-specific model.

3.5. Model comparisons

UFP prediction surfaces (100 m × 100 m) were generated for the city of Montreal from the four relevant models, namely the Montreal UFP LUR model (MM), the Montreal model based on transferring and recalibrating the Toronto model (MCTM), the Toronto model transferred to Montreal without calibration (NCTM), and the combined model with interactions (CM2), using ArcGIS 10.5.1 (Fig. 6). Note that the production of all prediction surfaces assumed a mean value for the windspeed that was observed at each site across the monitoring campaign for each city. The UFP predictions results show that the MM model generates the smoothest surfaces of all four models; yet it clearly predicts high levels only along the major highways, particularly highway 40 and to a lesser extent highway 20. Levels in downtown Montreal also tended to be high. On the other hand, the MCTM tends to predict higher UFP concentrations in the northern parts of the city as compared to the south. This is probably due to the positive coefficient associated with distance to airport. The effect of the major highways is also evident but not as accentuated as the MM. Predictions based on the Toronto model without calibration generated the highest concentration of all 4 models. This was to be expected as the model showed signs of a positive bias. Moreover, this model clearly emphasized the roadway effects on increasing UFP levels. Unlike the MCTM, the concentrations in the northern parts of the city were predicted to be the lowest of all four models. Finally, the CM2 predicted UFP levels in a manner similar to the MM model; yet there were no pollution hotspots along the major highways. In addition, its predictions pointed towards higher UFP levels in the downtown area as compared to the MM and the MCTM models. Interestingly, the MM model had the highest spatial correlation with

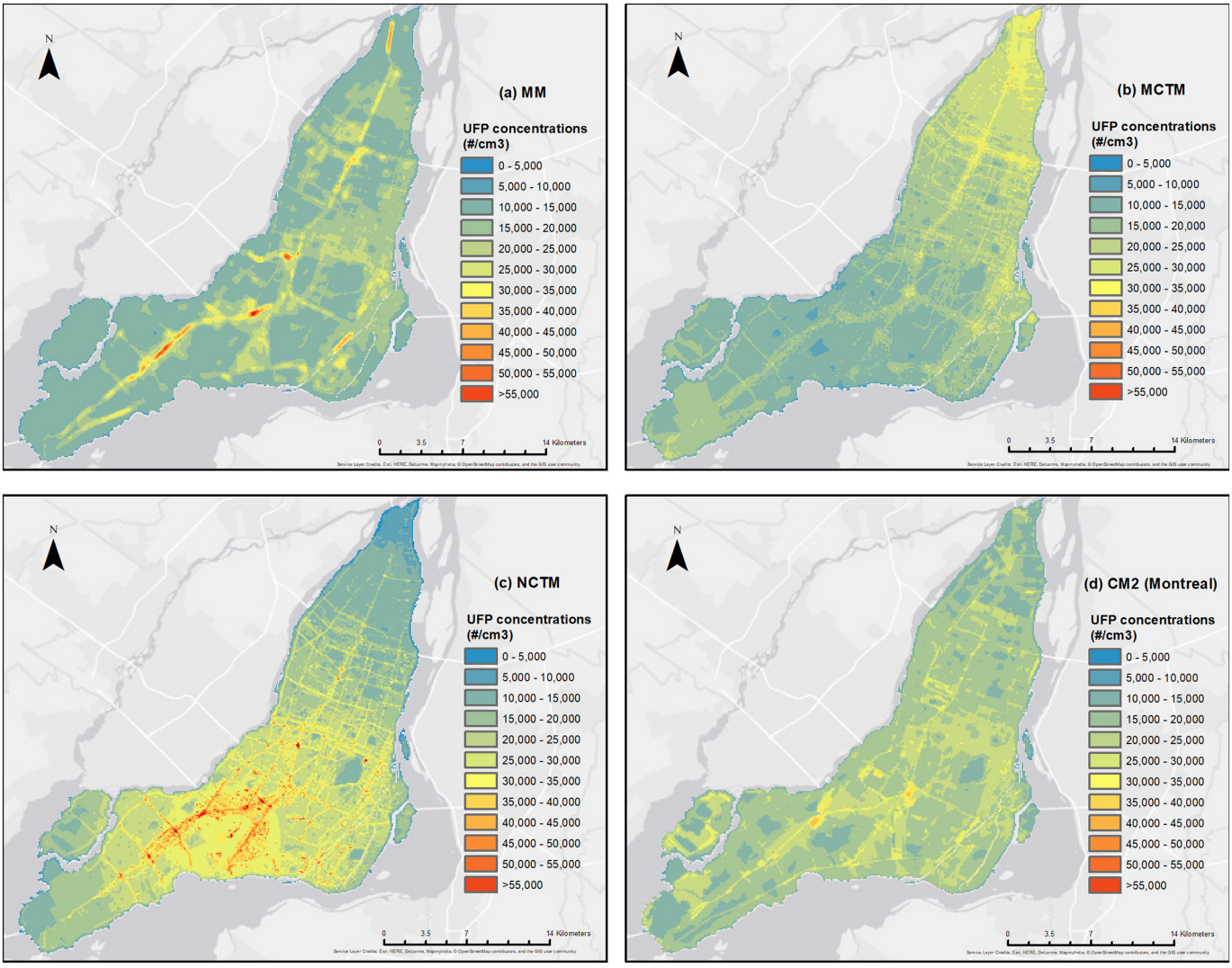

Fig. 6. a) Montreal UFP LUR model (MM); b) Toronto model based on transferring and recalibrating the Montreal model (MCTM) c) Toronto model transferred to Montreal without calibration (NCTM) d) Combined model with interactions (CM2).

Table 8
Spatial correlations between the 4 Montreal predicted surfaces. The correlation between two layers is a measure of dependency and agreement between the surfaces.

	MM	MCTM	NCTM	CM1	CM2
Montreal UFP LUR model (MM)	1	0.38	0.48	0.47	0.53
Montreal model based on transferring and recalibrating the Toronto model (MCTM)	–	1	−0.15	−0.36	0.15
Montreal model based on transferring the Toronto model without calibration (NCTM)	–	–	1	0.88	0.59
Combined model without interactions (CM1)	–	–	–	1	0.71
Combined model with interactions (CM2)	–	–	–	–	1

Table 9
Spatial correlations between the 4 Toronto predicted surfaces. The correlation between two layers is a measure of dependency and agreement between the surfaces.

	TM	TCMM	NCMM	CM1	CM2
Toronto UFP LUR model (TM)	1	0.59	0.56	0.86	0.86
Toronto model based on transferring and recalibrating the Montreal model (TCMM)	–	1	0.75	0.59	0.50
Toronto model based on transferring the Montreal model without calibration (NCMM)	–	–	1	0.53	0.44
Combined model without interactions (CM1)	–	–	–	1	0.99
Combined model with interactions (CM2)	–	–	–	–	1

the CM2 model and showed the lowest spatial agreement with the MCTM (Table 8). The CM2 model showed the strongest agreement with the NCTM and the lowest with the MCTM. This is probably due to the fact that the CM2 model was more influenced by the Toronto data as compared to the Montreal data. The lowest spatial autocorrelation was found between the MCTM and the NCTM; the autocorrelation was found to be even slightly negative (−0.15). (See Table 9.)

Similarly, UFP prediction surfaces (100 m × 100 m) were generated for the city of Toronto from the four relevant models, namely the Toronto UFP LUR model (TM), the Toronto model based on transferring and recalibrating the Montreal model (TCMM), the Montreal model transferred to Toronto without calibration (NCMM), and the combined model with interactions (CM2) (Fig. 7). Note that the production of all

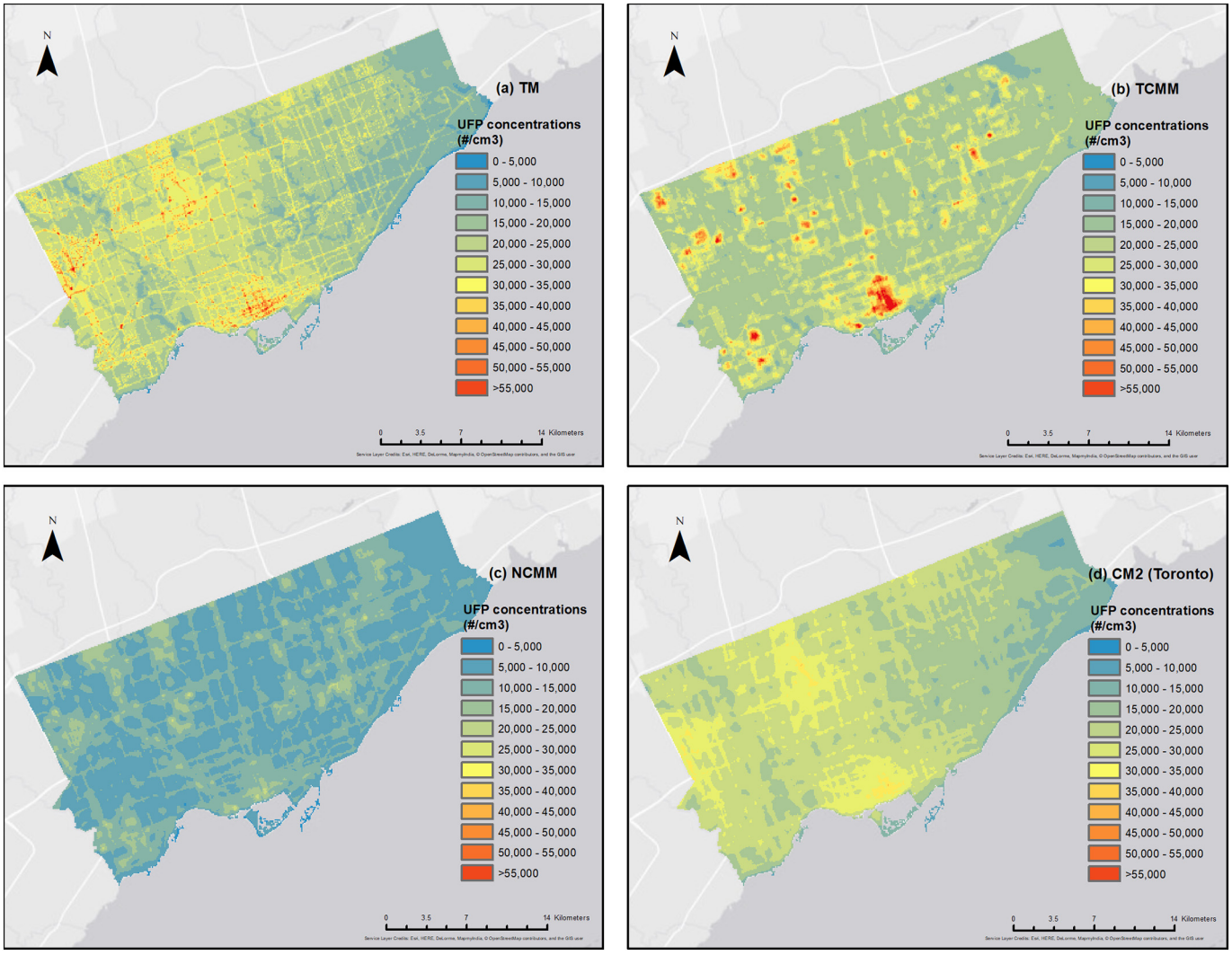

Fig. 7. a) Toronto UFP LUR model (TM); b) Toronto model based on transferring and recalibrating the Montreal model (TCMM) c) Transferred Montreal model transferred to Toronto without calibration (NCMM) d) Combined model with interactions (CM2).

prediction surfaces assumed a mean value for the windspeed that was observed at each sampling site during the monitoring campaign for each city. The prediction results show that the Toronto based model generated the most roadway dependent UFP concentrations of all other models. Moreover, the model also predicted high UFP levels at major intersections and in the downtown area. The Toronto model based on transferring and recalibrating the Montreal model (TCMM) showed very similar patterns; yet it predicted larger sections with very high UFP levels, particularly in the downtown area. The Toronto model based on transferring the Montreal model without calibration (NCMM) showed very similar patterns to the TCMM; yet its predictions were consistently lower. Lastly, the CM2 model predictions closely matched the TM model prediction; yet the levels were generally lower and the predicted concentrations were more smoothly distributed. Interestingly, the spatial autocorrelations between the 4 models were found to be consistently high; the highest correlation found was between the TM and CM2 predictions (0.85), while the lowest was between the CM2 predictions and those from the NCMM.

4. Discussion and conclusions

In this study, geographic transferability of the UFP LUR models was assessed using data collected through mobile monitoring campaigns in Toronto and Montreal. The model results showed that the city specific UFP LUR models developed for two distinct cities were reasonably similar in structure, with many common variables. Additionally, the signs on the coefficients of all predictor variables were alike, indicating that the developed models were largely able to capture the major promoters and suppressors of UFP pollution in the two urban cities. As a matter of fact, the structure of the recalibrated UFP models retained most of the predictors once transferred from one city to the other. Moreover, the effect of each of the predictors also remained relatively constant once transferred. This contrasts with the findings of Poplawski et al. (2009), who found that half of the predictors for their developed NO_2 LUR models became insignificant after recalibrating their initial Vancouver model with data from Victoria and Seattle, despite the 3 cities being fairly similar geographically. While this study points to the possibility that the relationship between UFP concentrations, land-use and the micro-traffic environment appears to be more stable across cities; such a conclusion will require support from additional studies conducted across multiple cities. However, one may conclude that some study-design factors tend to play a role in the potential transferability of LUR models between cities with similar geographic characteristics. These factors include 1) the use of the same (or similar but cross-calibrated) equipment and data collection protocols in the monitoring campaigns to reduce bias; 2) the collection of data under similar meteorological conditions (i.e. averaged over the same season, or averaged over a year) given that air pollutant concentrations are largely affected by meteorological variables; 3) spatially distributing the monitoring networks to adequately capture the variability in the land use types and the different road densities within each study area; and 4) the stability of the relationship between the pollutant on one hand and the LUR predictors on the other. However, given the limited work that has been done on LUR transferability, further studies are needed to validate the above-mentioned criteria and to agree on a generalizable approach for improving and assessing LUR model transferability.

The successful transferability of the model structure between the two cities encouraged us to develop two common combined models for both cities. Both of the combined models fared well and were comparable to the city-specific models with regards to their predictive powers. The fact that the performance of our UFP combined models was comparable to the city-specific models has also been reported by van Nunen et al. (2017) for UFP, Vienneau et al. (2010) for NO_2 and Wang et al. (2014) for $PM_{2.5}$. The CM2 model that allowed the magnitude of some variables to change across the two cities performed the best. These differences in model coefficients can largely be attributed to differences in city layout (e.g. location of airport), small-scale traffic characteristics (e.g. distance to major road), public transportation plans (e.g. density of bus stops), and urban landscapes (e.g. commercial and building areas).

Interestingly, the impact of changing the geographic location on the robustness and performance of the UFP LUR models was found be minimal and much less compared to the deterioration seen in the UFP model transferability for a given city using two different air quality sampling approaches (R^2 ranged between 0.018 and 0.088) (Minet et al., 2018). In this study, the lowest R^2 between the predicted and measured concentrations were found for the across city transferred models without recalibration. They ranged between 0.11 and 0.18. The R^2 improved significantly when the transferred model structures were allowed to be recalibrated with city-specific data, reaching R^2 as high as 0.38 for the Montreal-based model structure that was recalibrated with the Toronto data. Moreover, pooling the data together from both cities and fitting one combined model generated models with R^2 values that ranged between 0.41 for the CM1 model and 0.44 for the CM2. The performance of CM2 was found to be very close to the city-specific UFP LUR models (R^2 = 0.41 for Montreal and 0.49 for Toronto). Moreover, the predicted UFP model surfaces were all found to have a strong positive spatial autocorrelation, particularly in Toronto, where the combined CM2 model had a 0.85 spatial autocorrelation with the city specific model. Nevertheless, a closer look at the generated surfaces shows that while most models agreed on the areas of high and low pollution, their amplification of near-road and small-scale traffic variables differed.

In this study, the observed drop in the R^2 upon transferability was found to be lower as compared to other studies that reported a reduction ranging between 7% and 76% (e.g. Allen et al., 2011; Briggs et al., 2000; Marcon et al., 2015; Vienneau et al., 2010). This could be due to several factors that include: 1) the relatively low variability between the two cities in terms of geography and emissions as compared to other studies; 2) the spatial scale that the models were developed on was largely similar for the two cities; 3) the consistency in the adopted sampling protocols between the two study areas (timing, number of sites, equipment used, frequency, etc.); and 4) the decision to focus on developing robust models rather than potentially over-fitted models. Moreover, the larger drop in the R^2 in the case of the Poplawski et al. (2009) study in which the authors assessed the transferability of NO_2 models in cities with similar geography could be due either to the overfitting of city-specific models or to the properties of UFPs as compared to NO_2. However, more studies are needed to firmly conclude that UFP models tend to have better transferability characteristics as compared to other pollutants such as NO_2.

This study was the first attempt at studying the transferability of UFP LUR models. So far all previous transferability studies have focused on NO_2 (Allen et al., 2011; Briggs et al., 2000; Marcon et al., 2015; Poplawski et al., 2009; Vienneau et al., 2010; Wang et al., 2014), with a few that concurrently looked at the transferability of $PM_{2.5}$ (Wang et al., 2014), PM_{10} (Vienneau et al., 2010) and NO (Allen et al., 2011). The results of the current study agree with the conclusions reported in the previous transferability studies. Particularly, that attempting to transfer locally calibrated models to other areas without recalibration will result in poor performance. Moreover, transferring the model structure with recalibration resulted in a significant increase in the performance of the transferred models, similar to the conclusion made by Poplawski et al. (2009) when studying NO_2 LURs. Note that the model performance improvement with recalibration is not universal but appears to be pollutant dependent. In the case of PM_{10}, it was reported that the model performance remained poor even after recalibration, with the models' R^2 values ranging from 0.04 to 0.12 (Vienneau et al., 2010).

The results of this study appear to champion the need to develop national/regional models that pool data from multiple cities. Adopting a Bayesian hierarchical modeling (BHM) approach can provide a suitable framework to properly analyze cross-sectional data collected from

different cities. Such a modeling approach has the advantage of considering both the within- and between-city variances within a hierarchical modeling framework (Qian et al., 2010). The BHM framework is flexible to allow for the inclusion of inter-city differences, accounting for predictors at different spatiotemporal scales, the partial-pooling of data from different cities to predict in regions with sparse data, and to adaptively weigh the within- and between-group variances as a function of the relative magnitude of the between city variances and the size of the sampling program in each of the cities (Kashuba et al., 2010). The adoption of a BHM structure represents a data-based compromise between developing one common model for all cities (representing perfect transferability) versus developing separate models for each city (representing no transferability) (Gelman and Hill, 2014). The usefulness of the Bayesian hierarchical modeling approach in capturing the effects of factors that are operating at different scales has been demonstrated across various environmental systems (Cha et al., 2016; Cuffney et al., 2011; Dominici et al., 2002; Liu et al., 2008; Obenour et al., 2014; Qian et al., 2010).

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.01.123.

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