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The Development of a Land-use Regression Model for Ultrafine Particles in Toronto, Canada

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Abstract

This study applies land-use regression (LUR) to characterize the spatial distribution of ultrafine particles (UFP) in a large city. Particle number (PN) concentrations were measured in residential areas around Toronto, Canada, between June – August 2008. A combination of fixed and mobile monitoring was used to assess spatial gradients between and within communities. The fixed monitoring locations included a central site, two downtown sites, and four residential sites located 6 to 15km from the downtown core. The mobile data included average PN concentrations collected on 112 road segments from 10 study routes that were repeated on three separate days. The mobile data was used to create the land-use regression model while the fixed sites were used for validation purposes. The predictor variables that best described the spatial variation of PN concentration ($R^2 = 0.72$, validated $R^2 = 0.68$) included population density within 300m, total resource and industrial area within 1000m, total residential area within 3000m, and major roadway and highway length within 3000m. The LUR model successfully predicted the

afternoon peak PN concentration (slope = 0.96, $R^2 = 0.86$) but over-predicted the 24-hour average PN concentration (slope = 1.28, $R^2 = 0.72$) measured at seven fixed monitoring sites.

1.0 Introduction

Particulate matter (PM) exposure has been identified as a significant risk factor for the development of lung cancer and adverse health outcomes from cardiovascular and respiratory causes (Pope et al., 2002). These associations have been observed worldwide despite different sources of PM and different pollutant mixtures. More recently, ultrafine particles (UFP: particles with diameters less than 100nm) have been scrutinised due to their preferential deposition in the deepest regions of the human respiratory tract (Daigle et al., 2003), ability to promote the production of inflammatory biomarkers (Araujo et al., 2008; Bräuner et al., 2007; Delfino et al., 2010), and possible translocation to secondary organs within the body (Elder et al., 2006; Oberdörster, 2004). The health consequences resulting from UFP exposure remain uncertain due to the lack of long-term temporal and spatially resolved data (HEI, 2014).

UFP concentrations are influenced by numerous combustion related sources, secondary formation pathways and transformation processes that alter their number, shape, size and chemical composition. In urban areas, vehicles, residential heating, cooking and industrial activities are common sources of UFP (Aalto et al., 2005; Buzorius, et al., 1999; Wang et al., 2011a; Sabaliauskas et al., 2012). Previous investigations have found both the particle number (PN) concentration and size of UFP exhibit significant seasonal, diurnal and weekday-weekend variation (Cyrus et al., 2008; Puustinen et al., 2007; Wang et al., 2011b). Although the number of urban areas with long-term temporal UFP datasets is growing, the majority of these data are from single monitoring sites. Assessing UFP concentration gradients across an urban area remains challenging. Efforts to characterize UFP concentration gradients in urban areas have measured UFP simultaneously at multiple sites (Buzorius et al., 1999; Cyrus et al., 2008; Johansson et al., 2007; Kim et al., 2002; Puustinen et al., 2007; Ragettli et al., 2014; Sabaliauskas et al., 2014), while walking along roadways with different traffic intensities (Kaur et al., 2006), and while driving (Kittelson et al., 2006; Pijola et al., 2006).

Land-use regression (LUR) is a commonly used modelling technique that establishes empirical relationships between air pollutant concentrations and geographical predictor variables. This technique has been applied to NO/NO₂, PM_{2.5} and VOCs (reviewed by Hoek et al., 2008) and recently applied to UFP (Abernethy et al., 2013; Hoek et al., 2011; Rivera et al., 2012). Although the application of LUR methods presents an opportunity to characterize the range of UFP concentrations that may exist within a city, numerous challenges relating to instrumentation, logistics and statistics remain. To develop a reliable LUR model for any pollutant, the number of sites is an extremely important parameter. Unlike the passive NO/NO₂ badges that enable reliable simultaneous sample collection over long time horizons (weeks) and at many sites (at least 20, but ~ 100 is common), existing UFP instrumentation is expensive and often requires substantial user intervention to collect accurate results. Consequently, existing UFP LUR models developed for large urban areas rely on measurements collected at many sites (>20) over short time horizons (hours) (Abernethy et al., 2013; Rivera et al., 2012) or at many sites (50) over longer time scales (weeks) but non-concurrently (across multiple seasons) (Hoek et al., 2011).

The abovementioned model development approaches result in a few unavoidable analysis challenges. Firstly, UFP exhibits a strong diurnal pattern from a combination of vehicle emissions and secondary particle formation (Jeong et al., 2010; Kulmala et al., 2004). Studies that rely on a short-duration measurement to characterize the UFP concentration at each site need to either collect measurements at all sites at roughly the same time of day or apply a correction factor to make the mornings and afternoons comparable. However, the shape of the diurnal trend can vary substantially between days and even between sites due to localized differences in meteorology and emissions (Jeong et al., 2010; Ketzel et al., 2004a; Sabaliauskas et al., 2012). Therefore, simply applying the same diurnal correction factor across all sites may not be appropriate. Secondly, UFP exhibit an inverse relationship with temperature, with the highest PN concentrations typically observed during the cooler winter months (Sabaliauskas et al., 2012; Wang et al., 2011b). As a consequence, measurements need to be collected during periods with similar meteorological conditions, repeated at the same location over multiple seasons, or corrected (Hoek et al., 2011). Thirdly, the PN concentration can be impacted by the presence of larger particles (Kulmala et al., 2001). The particle mass concentration can vary on a seasonal

basis with higher temperatures favouring the formation of secondary aerosol. As a result, despite measuring the UFP concentration under similar meteorological conditions, the PN concentration may be suppressed due to the presence of large particles on one day and enhanced by their absence on another. Finally, nucleation and growth events can dramatically impact the observed PN concentration during the afternoon periods (Jun et al., 2014) and may result in more homogeneous PN concentrations and size distributions across a region (Jeong et al., 2010).

This study describes the development of an LUR model for UFP in Toronto, Canada. UFP has been measured continuously in downtown Toronto since 2006. Long-term trends suggest a gradual reduction in the total number concentration between 2006 and 2011, likely due to changes in the vehicle fleet, reductions in coal-fired power plant usage and the economic downturn that impacted the United States and Southern Ontario (Sabaliauskas et al., 2012). A recent analysis of UFP measurements collected at six fixed residential field sites in Toronto suggests that vehicle counts may be an important predictor of only a portion of the UFP concentration (Sabaliauskas et al., 2014). This study applies a hybrid methodology and relies on the measurements collected at the same six fixed field sites supplemented with mobile monitoring to better characterize and establish spatial relationships across the city. Specifically, an LUR model was created based upon mobile measurements and tested against measurements from the fixed sites.

2.0 Methodology

2.1 Study Design

UFP measurements were collected using a combination of fixed site and mobile monitoring (Figure 1) in Toronto, Canada. The fixed monitoring portion of the study was executed at a central site, two downtown sites (A, B) and four residential sites (C, D, E, F) for 1-3 weeks throughout the summer of 2008 (Figure 2). The residential sites were located between 6 and 15km away from the downtown core (Sabaliauskas et al., 2014). The mobile phase of the study assessed spatial gradients within neighbourhoods by collecting UFP measurements while walking along roadways with different traffic densities near and between the fixed field sites. Two routes were designed within each neighbourhood to ensure that locations both upwind and

downwind from major sources such as expressways, major arterial roads and railroads were visited. The maximum distance between the paired mobile measurements varied by route but ranged between 0 and 6 km. Each route was followed on three separate days.

2.2 Instrumentation

Multiple particle sizing and counting instruments were deployed at the sites for the study (Figure 2). The Central Site was located at the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR) near downtown Toronto. The air sampling inlet was 6m above the ground and 20m from a major arterial roadway with a weekday traffic volume of 20,000 vehicles per day. UFP was monitored continuously at the Central site using a Fast Mobility Particle Sizer (FMPS, Model 3091, TSI Incorporated, Shoreview, MN, USA). The FMPS provided particle number based size distributions over the range of 5.6 to 560nm with 1-second time resolution. One-minute averaged data were extracted and hourly averages were calculated for hours with more than 70% of the data availability. The FMPS used in this study was compared against two Scanning Mobility Particle Sizers (SMPS) in Jeong and Evans (2009). While the FMPS and SMPS instruments reported similar total PN concentrations, the shape of the particle size distributions differed due to limitations in the FMPS inversion algorithm. In this study, the empirical correction factors developed by Jeong and Evans (2009) were applied to the FMPS size bins (8.06 to 93.1nm) to ensure equivalency between the FMPS and SMPS instruments. The counting efficiency of the FMPS for size bins less than 8nm and greater than 300nm was low. To ensure consistent results, only particles between 8 and 300nm were included in this study.

The residential field sites (A-F) were equipped with one of three Water Condensation Particle Counter (WCPC, Model 3781, TSI, Shoreview, MN, USA). This portable WCPC measured particles between 6 and 1000nm. The instrument was operated at 1-minute time resolution. Any observations flagged as erroneous (due to out of range temperature or flow rate values) were removed and not included in the analysis. Hourly average concentrations were calculated for hours with more than 70% data availability. Side-by-side comparisons between the three WCPCs and FMPS (size bins 8-300nm) showed excellent agreement ($R^2 = 0.92-0.95$). All

WCPC measurements were scaled by a factor of 1.4 to ensure equivalency with the FMPS. The critical orifice was cleaned after every seven days of field operation.

UFP spatial gradients were measured using a handheld P-Trak Condensation Particle Counter (Model 8285, TSI Incorporated, Shoreview, Minnesota, USA). The P-Trak measured the total particle number concentrations (pt/cm³) for particles between 20 and 1000nm. The P-Trak had an upper limit of 500,000 pt/cm³ and logged data at 10-second time resolution. The P-Trak and FMPS exhibited good correlation for particles greater than 20nm when sampling 4m from a major arterial roadway (slope=1.24, R²= 0.83). A slope of 1.0 between the FMPS and P-Trak was observed for particles with diameters greater than 24nm suggesting that the size-cut off may be slightly higher than the factory reported value of 20nm. Given this slope of 1.0, the P-Trak data were used without any scaling. Matson et al. (2004) compared a P-Trak to a Condensation Particle Counter with a size cut-off of 10nm and observed good agreement to within 20% near traffic sources. Zhu et al. (2006) demonstrated that the P-Trak underestimates the total particle number concentration when sampling freshly emitted particles from traffic sources. The P-Trak was operated for no more than 3 hours on a single charge of alcohol under high relative humidity conditions (greater than 70%) to minimize any interference.

2.3 Data Analysis

2.3.1 Mobile Monitoring

A total of 112 road segments located along ten sampling routes were visited on three separate days between June 13th and August 20th 2008. To ensure sampling occurred under similar meteorological conditions, all measurements were collected on precipitation free days and on days with forecasted afternoon temperatures between 25 and 32°C. To minimize diurnal influences, measurements were collected on 15 weekday afternoons between 12:00 and 15:00. Two study routes were walked simultaneously. Each sampler carried a GPS unit and log sheets to record their position relative to street addresses and other local features at 1-minute intervals. This manual position logging ensured that the sample locations were well defined in areas with tall buildings.

The midpoint on each visited road segment was identified and used as an average position on the roadway (average road segment length = 430m; median = 330m). Routes that followed the same road past multiple major arterial roads were divided into multiple segments at the major arterial roads. The average UFP concentration was calculated for each road segment and matched to the average position on the road. The resulting UFP samples were between 5 and 10 minutes long per road segment. The UFP concentration that was used to produce the LUR model was a three visit average PN concentration for each road segment.

2.3.2 Central Site and Fixed Sites

A detailed description of the data treatment at the central site and fixed sites is available in Sabaliauskas et al., (2014). Briefly, the Central site was operated continuously while the fixed site monitors were operated for 1 – 3 week intervals throughout the summer of 2008. Only periods with valid data at both the Central site and fixed site were included in the analysis. In each neighbourhood, intermittent sources of UFP were likely present but could not be removed from the data set and may have included emissions from garbage trucks, street cleaning, landscaping services, cooking, individual vehicles and construction. Data were removed from the residential field sites for times during which it was known that the site was influenced by barbequing or lawn maintenance activities as reported by the study volunteers. However, these activities tended to occur during the evenings and on weekends and were for the most part outside of the mobile sampling period of 12:00 to 15:00.

2.4 Study Method Validation

A Monte Carlo algorithm was developed to determine the probability that the short duration mobile data is representative of data collected at the fixed sites over longer time horizons. The algorithm was implemented as follows:

1. An overall mean PN concentration was calculated for each of the fixed sites based on all the five minute averaged data collected on weekdays between 12:00 and 15:00.
2. Three five minute averages were randomly selected for a given site from three separate days

3. The mean value of the three random samples was calculated
4. This process was repeated 100,000 times to ensure that a representative distribution of all possible combinations of the data was generated.
5. A probability distribution of simulated averages was obtained from the 100,000 averages resulting from step 4
6. Based on the probability distribution from step 5, the likelihood that a simulated average would be within $\pm 5\%$, 10% , ... 95% , 100% of the overall average at each fixed site was calculated

2.5 Geographical Variables

The UFP LUR was developed from the mobile measurement data using geographical and socioeconomic data from multiple sources (Table 1) including variables for roadway length, roadway distance, traffic density, land use classification, population characteristics, restaurant density and address density. Circular buffer sizes of 50, 100, 200, 300, 500, 750, 1000, 1500, 2000, 2500 and 3000 m were examined. All variables were extracted using ArcGIS software (Version 10.1, ESRI, Redlands, CA, 2011). Most predictor variables were derived for different buffer sizes by summing the length of lines (e.g. roads and highways), area (e.g. park area) or the number of points (e.g. number of restaurants) within the circle.

2.6 Model Development and Validation

A total of 343 variables were evaluated for inclusion into the land-use regression model. This work applied the model building algorithm described by Henderson et al., (2007) with a slight modification. 1) all variables were ranked by absolute strength of their correlation with the measured pollutant; 2) the highest ranking variable and its corresponding buffer size was identified in each subcategory; 3) within each subcategory, the correlation coefficient was compared to all other variables with smaller buffer sizes; 4) if a variable had a slightly lower correlation with the measured pollutant than the highest ranked variable ($< 2\%$ difference), the variable with the smallest buffer size was selected; 5) variables exhibiting correlation with the highest ranked variable in each subcategory (Pearson's $r > 0.6$) were removed; 6) all remaining

variables were entered into a forward stepwise linear regression; 7) variables with insignificant t-statistics ($p > 0.05$) were removed; 8) Steps 6 and 7 were repeated until convergence and variables that contributed less than 1% to the R^2 were removed; 9) models were rejected if the signs (+/-) of the regression coefficients were inconsistent with a priori assumptions (e.g. sources of UFP should have positive regression coefficients). The outcome was a multiple linear regression model of the form Equation 1:

Equation 1:

$$\text{UFP Concentration} = \text{intercept} + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \dots + \beta_n X_n + \varepsilon$$

where β is the coefficient for each predictor variable X and ε is the random error term.

The LUR model was evaluated using leave-one-out (LOO) cross validation recreating the LUR model with $n-1$ sites. The predictive power of the LUR model was tested by predicting the UFP concentration measured at the central site and the six residential field sites.

2.7 UFP Surface Generation

To create the UFP surface, 4000 random points separated by minimum distances of 200m were generated within the city limits of Toronto. At each of the 4000 locations, the LUR model's predictor variables were extracted and UFP concentrations were calculated using the LUR model. Universal kriging was applied to the UFP concentration estimates resulting in a representation of the UFP concentrations across Toronto. Maps were truncated to the maximum measured PN concentration during the mobile phase of the study.

3.0 Results

3.1 Mobile Measurements

The average $\pm$ 95% confidence interval for major arterial ($>20,000$ vehicles per day) and local roads (<2000 vehicles per day) for each neighbourhood are reported in Table 2. In all neighbourhoods, the mean PN concentration on major arterial roadways was higher than on local roads (except Midtown 1, Route 1). The UFP concentration measured on downtown major

arterial roadways were often 3 to 5 times higher than those in residential areas. Further, major arterial and local roadways located in downtown and mid-town neighbourhoods were significantly higher than those located in areas away from the downtown core.

3.2 Method Validation: Selected Measurement Period

The average diurnal trend observed at the central site and six residential sites is presented for weekdays in Figure 3. The diurnal trend presented for the central site encompasses the entire measurement period; however, the shape and magnitude were consistent even when truncated to match the sampling periods of the individual residential sites. The downtown sites (A, B) had similar diurnal behaviour as the central site, while the residential sites (C, D, E, F) exhibited distinctly different behavior, presumably due to differing activities at the sites related to their land use. Across the city, the minimum PN concentration occurred between 4:00 and 6:00 in the morning. The initial increase in PN concentration between 6:00 and 9:00 corresponded to the morning rush hour period. At most sites, the daily maximum PN concentration was observed between 12:00 and 15:00. Although afternoon PN concentration peaks are often associated with particle formation and growth events, simultaneous events of the same magnitude were not observed at the residential sites. On some days, slight increases in PN concentration were observed during the afternoon periods at the residential sites; however, the contribution of these formation events to the overall PN concentration appears to be small.

The mobile measurements were collected between 12:00 and 15:00 (Figure 4). At the residential sites (C, D, E, and F), the PN concentrations were significantly lower than those observed at the central site and sites A and B. These concentration differences were likely due to different emissions profiles surrounding the monitoring sites. Further, the PN concentration exhibited relatively stable behaviour between 12:00 and 15:00 (within 20%) suggesting that the daily maximums have been reached and this period offers an opportunity to study the spatial variation of UFP without the need to correct for diurnal variation. However, the resulting predictive model likely overestimates the 24-hour average.

3.3 Method Validation: Sample Time

Each mobile route was followed on three separate days to capture a range of possible conditions in each neighbourhood. The combination of mobile and fixed site monitoring enabled better quantification of the uncertainties surrounding the mobile measurements. Using the Monte Carlo simulation method described in the methods section, a probability distribution of averages was obtained based on random subsamples from the fixed monitoring sites. The percent of simulated averages that were within $\pm 5\%$, 10% ... 100% of the measured mean are presented in Figure 4 for each field site. Notably, at the downtown sites (A, B), there exists high probability ($>90\%$) that the simulated means are within 30% of the measured PN concentration average. However, at the residential sites, the probability that the simulated means were within 30% of the measured PN concentration average was between 54 and 74% .

3.4 The UFP LUR Model

Table 3 summarizes the LUR model obtained from the mobile PN concentration data. The model has an $R^2 = 0.72$ and a leave one out (LOO) cross validation $R^2 = 0.68$. Variance inflation factors (VIF) were < 5 (ranged from 1.37 to 4.07) suggesting limited collinearity among model variables. There was spatial autocorrelation of the PN concentrations (Moran's $I = 0.73$) and no spatial autocorrelation of the residuals (Moran's $I = -0.09$).

The parameters included in the model were related to population density, dwelling density, roadway length and resource and industrial activities. Two traffic related parameters were included in the model: length of major arterial roadways and highways within 300m and within $300\text{-}3000\text{m}$. In the initial development of stages of the LUR model, a single road length parameter was included (3000m buffer); however, the PN concentrations on major arterial roadways were consistently underestimated. Upon separating the two traffic-related terms, the total model R^2 increased by 2% and the predictive power of the model for PN concentration data collected on major roadways improved. Although resource and industrial area within 1000m was a good predictor variable, Toronto does not have many large emitting facilities. The roadways in and near areas with resource and industrial land use are known to have more heavy duty diesel

truck traffic than other areas in the city (Roorda et al., 2010). Thus the resource and industrial land area parameter may be a partial proxy variable for freight traffic.

3.5 Mapping UFP

Figure 5 shows PN concentration distribution across Toronto during summer between 12:00 and 15:00. The map suggests that the PN concentration in Toronto may vary by as much as a factor of 10 during the afternoons. The highest PN concentrations were observed in the downtown core (near the central site, sites A and B), near the highway junction points and around industrialized areas in the city. The PN concentrations were slightly higher on the west side of the city. The lowest PN concentrations were found along the shore of Lake Ontario to the east of the city.

3.6 Model Evaluation

The LUR model developed for Toronto had a total model $R^2 = 0.72$. The individual models developed for the LOO cross-validation had R^2 values ranging from 0.72 to 0.76 and RMSE values ranging from 3,150 to 3,380 cm^{-3} or between 23 and 25% of the average measured PN concentration. This study benefited by collecting measurements at approximately the same time every day to avoid needing to apply temporal correction factors and by visiting each road segment on multiple days to avoid the need to correct for day-to-day variability.

Figure 6 presents the predictive power of the UFP LUR model (upper: measurement period 12:00 – 15:00, lower: 24-hour average). Overall excellent agreement was observed between the measured and predicted PN concentrations. The LUR model surface had stronger predictive power for measurements collected between 12:00 – 15:00 ($R^2 = 0.86$) than the 24-hour average regression ($R^2 = 0.72$). The LUR model surface had higher R^2 values than the manual calculation of UFP concentrations at the fixed sites (afternoon manual calculation $R^2 = 0.76$ versus surface $R^2 = 0.86$ and 24 hour average manual calculation $R^2 = 0.65$ versus surface $R^2 = 0.72$) but similar slopes. The LUR model overestimated the 24-hour average PN concentration at the fixed site by a factor of 1.28. This is unsurprising since the mobile measurements were collected during the afternoon when the PN concentration is likely to be highest.

4.0 Discussion

Initial efforts to develop a UFP LUR model focused on normalizing the mobile PN concentration data with simultaneous measurements collected at the central site. This effort was ultimately abandoned because the central site was located 20m from a major arterial roadway and the impacts of individual vehicles disproportionately skewed the results. In future studies, an urban background monitoring site might prove to be appropriate to minimize the impacts of day-to-day variability in PN concentration. This study demonstrates that LUR modelling can be successfully applied to raw mobile UFP concentration data ($R^2 = 0.72$, validated $R^2 = 0.68$) provided that the measurements are collected at the same time of day and under similar meteorological conditions. The Toronto LUR model has a similar R^2 value as the Hoek et al., (2011) model for Amsterdam ($R^2 = 0.65$) and considerably higher R^2 values than the LUR models developed by Abernethy et al., (2012) for Vancouver, Canada ($R^2 = 0.34 - 0.53$) and by Rivera et al., (2012) for multiple urban areas in Spain ($R^2 = 0.13 - 0.63$).

The need to optimize monitoring strategies to balance the competing needs of temporal and spatial coverage to produce an UFP LUR is an ongoing challenge. This work applied a mobile data collection strategy to produce an UFP LUR model. While each sampling route was followed at the same time each day and on three separate days, the optimal sampling time and number of repetitions was unknown *a priori*. Other studies have applied short duration data collection strategies to produce UFP LUR models. Rivera et al., (2012) visited 644 locations on one occasion to develop an UFP LUR model for multiple towns in Spain. For a small subset of the sites (25 locations), measurements were collected on two sampling days. However, the measured UFP concentration on day 1 and day 2 exhibited limited correlation. Abernethy et al., (2012) collected UFP concentrations at 20 locations over one hour intervals on two separate occasions. Van Poppel et al., (2013) collected mobile measurements on 10 days (twice daily) using a bicycle and found substantial variability within 6 study zones. Notably, each zone had different traffic influence and required a different number of repetitions and background correction before an average that was representative of the 20 sample mean was collected.

The results from the Monte Carlo simulation are intended to provide guidance for future mobile field study design. However, since the fixed site measurements were collected at the rear of the study participants' homes, the impacts of traffic at these locations is likely lower than those encountered while walking along roadways. The Monte Carlo simulation performed in this study indicated that the downtown neighbourhoods were more likely to be within 30% of the long-term average concentration (>90%) than the residential neighbourhoods (ranging from 54 to 74%). Although the variability at the residential sites (C, D, E, F) was considerably higher than observed at the downtown sites (A, B), this simulation may represent a worst case scenario for mobile studies. At the downtown sites, the traffic intensity is fairly constant. Consequently, when measuring the PN concentration near the downtown core, the likelihood that an individual measurement is skewed due to a local source is low because the source is continuous. In contrast, at the residential sites, significantly fewer vehicles pass the monitoring sites. Since the flow of vehicles is not continuous, these few vehicles have the ability to skew the PN concentration upwards over short time horizons. When walking in the residential areas, times when trucks passed the mobile samplers or times when lawn maintenance was occurring were recorded and subsequently removed from the data set. Since an equivalent record is unavailable for the fixed sites, intermittent sources may broaden the simulated probability distribution. As a result, the likelihood that the measured PN concentration data fall within the 30% of the longer term average is possibly higher than the simulated means suggest.

Abernethy et al., (2011) observed correlation between PN concentration and the density of fast food restaurants. This study also observed strong correlation between the number of restaurants and PN concentration; however, restaurant density was highly correlated with the length of major roadways and highways. Consequently, restaurant density was not included in this LUR model due to the likelihood that vehicles were the more dominant source of UFP. However, with greater spatial coverage in future studies, the relationship between UFP and restaurants may be further elucidated.

Photochemical nucleation events have been previously reported in Toronto and the surrounding areas in Southern Ontario (Jeong et al., 2010; Yun et al., 2014). These events are characterized by a sudden increase in PN concentrations with diameters less than 25nm followed

by a period of growth. Previously, these events have been observed simultaneously at multiple sites separated by hundreds of kilometers (Jeong et al., 2010). While the PN concentrations are heterogeneous across Toronto and the rest of Southern Ontario, the PN concentrations become more uniform during these events. In this study, UFP measurements were collected on days without photochemical nucleation events. As a result, the spatial relationships established in this LUR model are likely only valid on event free days.

This study relied on the P-Trak condensation counter with a lower size cut-off of 20nm. Previous investigation of UFP in Toronto and other urban areas suggests that the particle size distribution in areas away from traffic is dominated by larger particles (Ketzel et al., 2004a; Sabaliauskas et al., 2012). When walking along roadways far from major traffic sources, a large majority of all the particles are likely captured by the P-Trak. In areas close to major traffic sources, the UFP size distribution shifts towards smaller particles and the P-Trak likely underestimates the total PN concentration. Even though the size cut-off of the P-Trak ($>20\text{nm}$) was higher than the CPC 3786 ($>6\text{nm}$), only a small fraction of the UFP size distribution was less than 20nm at the residential sites (C, D, E, F) making the measurements from the two instruments directly comparable. However, in locations closer to major traffic sources, such as the central site and the downtown sites (A and B), particles $<20\text{nm}$ were likely present. At the central site (20m from a major arterial road), the average geometric mean diameter was smallest between the hours of 12:00 and 15:00 (mean = 28nm) and 40% of all particles were $<20\text{nm}$. Sites A and B were within 30 and 100m of a major arterial roadway, respectively. Given that the developed LUR model only captures particles $>20\text{nm}$, the predicted PN concentration at the central site, site A and possibly site B are likely underestimated.

This study presents a first approximation of the range of PN concentrations that are likely present in Toronto. The ability of the UFP LUR model to accurately predict the PN concentration in other seasons remains an open question. At the central site, the PN concentration is between 2 and 3 times higher during the winter than the summer due to changes in vehicle emissions, increased residential heating and the increased atmospheric lifetime of UFP (Sabaliauskas et al., 2012). Consequently, the spatial relationships established between PN concentrations and traffic-related and residential area variables will likely differ during the winter. Further, meteorological

patterns between summers can vary substantially in Toronto. Consequently, the ability to predict summer PN concentrations at different locations in different years based on this LUR alone may not be possible.

5.0 Conclusions

This study successfully developed an LUR model for UFP in Toronto, Canada using a combination of fixed site and mobile monitoring ($R^2 = 0.72$, validated $R^2 = 0.68$). The predictor variables included population density within 300m, resource and industrial area within 1000m, residential area within 3000m and major road and highway length within 300m and within 300 to 3000m. In addition, this study benefited from a hybrid sampling protocol that balanced the competing needs of maximizing sampling time and spatial coverage to develop a robust LUR model. The need to apply temporal correction factors to the mobile PN concentration data were minimized by collecting measurements at the same time and by selecting days with similar meteorological conditions. Assessing the ability of the LUR model to predict PN concentrations in other seasons and in other years remains an open question.

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