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Development of a land-use regression model for ultrafine particles in Toronto, Canada

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HIGHLIGHTS

- Ultrafine particle (UFP) land-use regression (LUR) model was developed for Toronto.
- UFP was measured using a combination of mobile and fixed site monitoring.
- Predictor variables included road length, population density and industrial areas.
- The LUR model predicted the UFP concentration at six fixed monitoring sites.

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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 and 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 6–15 km 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 300 m, total resource and industrial area within 1000 m, total residential area within 3000 m, and major roadway and highway length within 3000 m. The LUR model successfully predicted the afternoon peak PN concentration (slope = 0.96, $R^2 = 0.86$) but over-predicted the 24-h average PN concentration (slope = 1.28, $R^2 = 0.72$) measured at seven fixed monitoring sites.

1. 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 100 nm) 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), and possible translocation to secondary organs within the body (Oberdörster et al., 2004). The health consequences resulting from UFP exposure remain uncertain due to the lack of long-term temporal and spatially resolved data (HEI, 2013).

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 (Buzorius et al., 1999; Wang et al., 2011a; Sabaliauskas et al., 2012). Previous investigations have found both the particle number (PN)

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concentration and size of UFP exhibit significant seasonal, diurnal and weekday-weekend variation (Cyrys et al., 2008; 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; Cyrys et al., 2008; 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).

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, PM2.5 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 above mentioned 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., 2004; 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. Methodology

2.1. Study design

UFP measurements were collected using a combination of fixed site and mobile monitoring (Fig. 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 (Fig. 2). The residential sites were located between 6 and 15 km 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 (Fig. 2). The central site was located at the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR) near downtown Toronto. The air sampling inlet was 6 m above the ground and 20 m 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 5.6–560 nm with 1-s 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 (8.06–93.1 nm) to ensure equivalency between the FMPS and SMPS instruments. The counting efficiency of the FMPS for size bins less than 8 nm and greater than 300 nm was low. To ensure consistent results, only particles between 8 and 300 nm were included in this study.

The residential field sites (A-F) were equipped with one of three Water Condensation Particle Counters (WCPC, Model 3781, TSI, Shoreview, MN, USA). This portable WCPC measured particles between 6 and 1000 nm. The instrument was operated at 1-min time