Title: Elucidating multipollutant exposure across a complex metropolitan area by systematic deployment of a mobile laboratory

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Abstract

This study evaluates a deployment strategy of a heavily instrumented mobile lab for characterizing multipollutant spatial patterns based upon a limited number of measurement days spread over different seasons. The measurements obtained through this deployment strategy are used to gain insight into average pollutant levels between routine monitoring sites and in relation to emission sources in the region, as well as to assess correlations between pollutant patterns to better understand the nature of urban air pollutant mixtures. A wide range of locations were part of the deployment in order to characterize the distribution of chronic exposures potentially allowing development of exposure models. Comparison of the mobile lab averages to the available adjacent air quality monitoring network stations to evaluate their representativeness showed that they were in reasonable agreement with the annual averages at the monitoring sites thus providing some evidence that through the deployment approach the mobile lab is able to capture the main features of the average spatial patterns. The differences between mobile lab and network averages varied by pollutant with the best agreement for NO_2 with a percent difference of 20%. Sharp differences in the average spatial distribution were found to exist between different pollutants on multiple scales, particularly on the sub-urban scale, i.e., the neighbourhood to street scales. For example, NO_2 was observed to be 210-265% higher by the main highway in the study region compared to the nearby urban background monitoring site, while black carbon was higher by 180-200% and particle number concentration was 300% higher. The repeated measurements of near-roadway gradients showed that the rate of change differed by pollutant with elevated concentrations detected up to 600-700 meters away for some pollutants. These results

demonstrate that through systematic deployment mobile laboratory measurements can be used to characterize average or typical concentration patterns thus providing data to assess monitoring site representativeness, spatial relationships between pollutants, and chronic multi-pollutant exposure patterns useful for evaluating and developing exposure models for outdoor concentrations in an urban environment.

1 Introduction

Long-term or chronic exposure to air pollution has been shown in many epidemiological studies of different types, such as cohort studies, case-control studies and cross sectional studies, to be associated with adverse human health outcomes (Ren and Tong, 2008). While most studies have focused on correlations of outcomes (e.g., mortality) with a small number or even a single pollutant (e.g., $PM_{2.5}$, NO_2) (Adar et al., 2013; Jerrett et al., 2009; Pope and Dockery, 2006), it is generally believed that no single pollutant is solely responsible. Rather features of the mix of pollutants in the air, particularly when the myriad of possible adverse health effects is considered, are more likely to be exerting the effects, possibly synergistically (Mauderly and Samet, 2009). Furthermore different pollutants in the mix likely have different effects exhibited by different endpoints, acting through different mechanisms.

In studies where exposure to a single pollutant is used for assessing associations with health outcomes, it is often acknowledged that this pollutant is a proxy for a specific source or more-complex mixture of concern. For example, NO_2, ultrafine particles (particles with a dynamic range of >0- 100 nm) and black carbon (BC) are often considered as proxy pollutants for traffic related air pollution (TRAP) or fossil fuel combustion in general (e.g., Brook et al., 2007; Bukowiecki et al., 2003; Crouse et al., 2010; Janssen et al., 2011). SO_2 is thought of as a proxy for heavy industry such as emissions from petroleum refineries (e.g., Smargiassi et al., 2009) and benzene could be related to both these sources in some cities (Wheeler et al., 2008) or mainly to one in others (Levy et al., 2014). $PM_{2.5}$ is perhaps the pollutant of greatest interest (Brook et al., 2010) and is itself a mixture of compounds related to multiple sources and/or secondary

formation processes. While it is generally suspected that certain types of $PM_{2.5}$, related to certain sources or chemical composition, are more-likely responsible for health effects and that this could vary by outcome, this has yet to be demonstrated with enough confidence to explicitly support more-focused policies (Kelly and Fussell, 2012; Lippmann et al., 2013; Rohr and Wyzga, 2012).

The air pollutant mix we breathe as urban dwellers is the result of multiple emission sources (traffic, industry, residential, commercial and biogenic activities), each of which is emitting a different distribution of constituents. Many of these directly emitted primary pollutants are subsequently involved in reactions in the atmosphere to produce secondary pollutants (e.g., $NO+O_3$ to form NO_2; nitrogen oxides (NO_x) and volatile organic compounds (VOC) interacting to form ozone in the presence of sunlight; secondary organic aerosols derived from oxidation of VOCs). Given the spatial variability in pollution sources and sinks over short distances, the heterogeneity in the pollutant mix emitted and the non-linearity and varying time scales of secondary formation processes, the urban environment is challenging with respect to accurate characterization of air pollutant patterns and subsequent estimates of population and individual-level long term or chronic exposures (e.g., Costabile et al., 2009; Freiman et al., 2006). It should be noted that other factors also take part in determining exposure such as indoor-outdoor infiltration and individual time activity patterns (Monn, 2001). These factors can obscure how well outdoor spatial contrasts represent actual exposure leading to apparent modification of the effect of ambient concentrations (e.g., Janssen et al., 2002).

Although different methods for estimating chronic human exposure patterns across urban areas are being used, ultimately they all depend upon measurements of concentrations at multiple locations that ideally reflect long term average conditions and the range of levels that occur across space. Such measurements are used to develop empirical models and/or to validate physically-based models predicting concentrations based upon meteorological and emissions data. Point measurements obtained at routine air quality (AQ) monitoring sites have been used for both of these purposes (e.g., Beckerman et al., 2013; Cyrys et al., 2005; Hystad et al., 2011; Yuval et al., 2013) and they have the advantage of capturing long term conditions. However, because routine monitoring tends to focus on urban background locations, particularly in North America, the data are rarely able to sufficiently capture the spatial variability existing in the urban environment (Wheeler et al., 2008) failing to adequately quantify the peak concentrations and hence the actual concentration and chronic exposure gradients. Clearly, spatial interpolation of the same AQ site data (e.g., ordinary kriging and inverse distance weighting) will also fail to show the true spatial pattern in concentration since this approach does not take into consideration urban features like road networks, point sources and green spaces that influence local emissions and concentrations (Jerrett et al., 2007).

Intensive campaigns that deploy on the order of 10-100 monitors across a city over limited time periods have successfully been used in many cities to obtain information that more realistically captures the concentration gradients. These ‘saturation monitoring’ datasets can then serve as the source of the dependent variable in empirical exposure model development, such as Land Use Regression (LUR) (e.g., Brauer et al., 2003;

Henderson et al., 2007; Johnson et al., 2010; Kanaroglou et al., 2005). These approaches may also include datasets collected sequentially to increase the overall number of locations with active sampling for measurement of particles, although this requires temporal adjustments (e.g., Cyrys et al., 2005; Eeftens et al., 2012; Hochadel et al., 2006; Wang et al., 2013) or more complex spatial-temporal modeling methodologies (Gryparis et al., 2007; Szpiro et al., 2010) in the model development. Since the magnitude of the temporal adjustment can also vary spatially and is not well-characterized this can result in additional uncertainty in estimates of the long-term spatial patterns.

Dispersion models that predict the spatial pattern across the entire urban area (Cyrys et al., 2005; Hirtl and Baumann-Stanzer, 2007) or within the grid squares of more-advanced numerical models (Beevers et al., 2012; Isakov et al., 2007a; Nordling et al., 2008), have also been developed for estimating chronic human exposure patterns. These more physically-based dispersion modeling approaches are limited by the accuracy and level of detail of the meteorological and emissions input data and are generally used for predicting the spatial pattern of a small number of tracer pollutants (i.e., non-reactive). The more-advanced models, such as CMAQ, have also been considered directly for chronic exposure assignment (Marshall et al., 2008) and have the potential advantage of predicting concentrations of multiple primary and secondary pollutants in a consistent and comprehensive manner, although with varying degrees of confidence. However, they are limited by the grid cell size of a few square kilometers at best and therefore cannot resolve sub-grid (i.e., neighbourhood scale) variability. Hybrid approaches combining physical and empirical models in order to represent exposure across grid and sub-grid scales have also recently been proposed (Crooks and Isakov, 2013; Yuval et al., 2013).

Despite the need for intensive monitoring campaigns, spatial or spatial-temporal empirical models are thus far the most common approach for chronic exposure prediction within urban areas. As a result, they have been applied successfully in a large number of epidemiological studies in North America and Europe (e.g., Brauer et al., 2007, 2008; Brook et al., 2008; Crouse et al., 2010; Gehring et al., 2010; Jerrett et al., 2009; Rivera et al., 2013; Thiering et al., 2013; Villeneuve et al., 2013). One limitation with these approaches is the small number of air pollutants they consider because of difficulty in simultaneously measuring multiple pollutants during the multi-site intensive campaigns. As a result, it remains difficult to fully explore mutlipollutant exposures to study their combined effect or to assess the differential effects of different pollutants or to gain insight regarding the characteristics of the pollutant mixtures typically being represented by single indicator pollutants (Levy et al., 2014).

Mobile laboratories have also frequently been used for examining intra-urban variability of a range of air pollutants and may provide an alternate approach for obtaining data for spatial or spatial-temporal empirical model development (e.g., Larson et al., 2009; Patton et al., 2014). If sufficiently large to house and power multiple instruments and if deployed in a systematic manner, mobile labs could potentially generate comparable data to the traditional fixed site intensive monitoring campaigns for multiple air pollutants simultaneously.

Mobile labs or mobile measurement strategies have often been used to obtain highly resolved measurements both in time and space, but typically during relatively short time periods and for a limited number of pollutants (Bukowiecki et al., 2003; Durant et al., 2010; Fujita et al., 2011; Hagler et al., 2010; Isakov et al., 2007b; Weimer et al., 2009;

Westerdahl et al., 2005; Zwack et al., 2011). Short term measurement campaigns are common in atmospheric chemistry and air quality studies (e.g., Daum et al., 2003; Brook et al., 2013; Pennington et al., 2012) and for exploring chemical and physical processes the short time frame may not be a limitation if relevant cases are captured. However, for describing spatial and temporal behaviour, such as intra-urban variability for informing and developing empirical population exposure models (e.g., Dijkema et al., 2011) or the nature and frequency of certain events of interest (e.g., exceedence of guidelines) it is important to consider the representativeness of the short term study period. Yet this is rarely examined rigorously, though it can be relevant for framing the conclusions in the proper context.

Similarly, while saturation monitoring has proven to be useful for exposure model development, it is also important consider whether the time period(s) when the intensive campaign is conducted characterizes actual long-term spatial gradients. Capturing a typical period is challenging because of temporal variability in meteorology. Jerrett et al. (2007) compared their intensive campaign measurements and the NO_2 predictions from the model they developed to longer term values reported at monitoring sites. Differences of up to 26% were observed, although this could only be assessed for a few sites. Xu et al. (2007) examined representativeness versus number of measurements for multiple pollutants and several Canadian cities. For VOCs Miller et al. (2012) showed that measurements in the transition seasons tend to be more representative. Consistent with this, Henderson et al. (2007) carried out their two week Vancouver, BC, monitoring campaigns in the late winter and late summer based upon the greatest likelihood of representativeness. In terms of chronic exposure rankings, Wheeler et al. (2008)

compared the variation observed across 54 sites in Windsor, Ontario, among seasons. They found Spearman correlations between an individual season and the average pattern among seasons of 0.84 (summer) to 0.97 (spring) for NO_2. This range decreased to 0.75-0.93 for SO_2, but overall the ranking of locations captured by a discrete two week periods appeared to be stable. Wheeler et al. (2008) obtained similar results for benzene and toluene. Cyrys et al. (2006) compared the annual, seasonal and monthly means calculated from daily $PM_{2.5}$ measurements taken over 7 days per month for one year to daily measurements taken on all days of the same year in Erfurt, Germany. They found that while the annual and seasonal means showed small bias (1.7 μg m^3 and 2.7 μg m^3, respectively) the monthly means "can only be considered to be a crude estimate that may substantially under- or overestimate the true monthly mean value".

Mobile measurements have two main challenges associated with their use for examining long term average air pollutant patterns. The first is the fact that a mobile lab can only measure one location at a time and the second is that a relatively limited number of measurement days can be done because of the costs involved, particularly when large, heavily instrumented mobile labs are involved. A systematic approach is therefore needed to efficiently characterize representative long term patterns of the multipollutant mix in a complex urban environment from mobile measurements. This approach should involve multiple measurements at the same locations over different days and different seasons with high spatiotemporal resolution and with a large suite of measuring devices examining both gaseous and particulate pollutants simultaneously.

In this study, high resolution mobile measurements of multiple air pollutants were taken in the city of Montreal, Quebec, Canada, over multiple days with a focus on

multipollutant spatial contrasts. The underlying hypothesis explored is that a systematic deployment strategy of a mobile laboratory can produce measurements that are representative for long term exposure levels and gradients within urban areas. Therefore, the objectives of this paper are to evaluate a deployment strategy of a mobile lab in a large city undertaken to characterize longer term average concentrations relevant to chronic exposures and then to apply the location-average data collected by this strategy to assess multi-scale, multipollutant spatial contrasts to reveal more about sources, hotspots and how exposure potentially varies within the population. This is hoped to move us closer to understanding urban mixtures and to provide insight to help in the interpretation and development of epidemiological studies and statistical methods that seek to address the multipollutant exposure and health effect questions, particularly as it pertains to TRAP.

2 Methods

Air quality and meteorological measurements were taken by Environment Canada's mobile lab: Canadian Regional and Urban Investigation System for Environmental Research (CRUISER). CRUISER is a diesel engine vehicle (GMC C7500 medium duty truck) equipped with a power generator and climate control systems that maintain stable conditions inside the customized vehicle body, housing research-grade measurement instruments. To maximize data collection during deployments CRUISER measurements are typically taken on a 24 hour basis for the entire time period it is present in a study location. Thus, much of the data are obtained when the vehicle is parked and plugged in at its "home base" location. Depending upon the study objectives there are typically

multiple periods of mobile measurements when CRUISER drives and measures, occasionally stopping to obtain fixed point data, meteorological measurements and to cross-reference with existing AQ monitoring sites.

2.1 Study Area

The study was conducted on the Island of Montreal, which has 1.8 million inhabitants (Statistics Canada, 2011), but a larger population in the city of Montreal and the surrounding area of 3.8 million, being the second largest city in Canada (Statistics Canada, 2011). As with most large cities, air pollution in Montreal is spatially variable (Crouse et al., 2009; Gilbert et al., 2005). Pollution sources on the island besides traffic include a variety of industrial activities, oil and gas refining, storage and distribution facilities, petrochemicals, metal refining, light manufacturing, multiple port areas, as well as heating (in the winter) (Environment Canada, 2006). Figure 1 shows a map of the city providing information on the different land uses, main roads and major point sources.

2.2 Measurements

The Montreal measurement campaign was conducted during 2009 in three seasons (winter, summer and autumn), with the combined analysis of the entire period referred to as the campaign average. There was an approximately three week deployment in each season (13/01 – 11/02; 8/07 – 3/09; 19/11 –3/12) and this paper focuses on the mobile portion of these measurements (i.e., excluding times CRUISER was parked overnight and other non-driving days) in order to characterize spatial patterns in detail.

Measurements of 19 different parameters were taken simultaneously from the CRUISER platform throughout the campaign at time resolutions ranging from 0.5 second

to 2 minutes. Four additional species were derived from the measurements. Geo-location was recorded with a Garmin 176C GPS system every second and CRUISER's speed was determined from the GPS data. A list of the parameters measured, instruments used, their temporal resolution and detection limit is given in Table 1. The inlets for the air quality instruments were located at the roof of the vehicle, 3.6 m a.g.l. oriented near the front left side. The GRIMM Dust monitor, used for particle mass measurements (i.e., PM_{10}, $PM_{2.5}$ and $PM_{1.0}$), had a separate inlet to enable the capture of coarse particles, but due it is slow flow rate and the impact of horizontal speed on the capture efficiency of larger particles a correction based upon CRUISER's speed was applied. A description of the inlets and data management is given in the Supplemental Material C. Speed correction to the GRIMM particle measurements is discussed in Supplemental Material D.

Data were flagged for invalid periods, below detection limit and missing values for each instrument. The data were then combined to one dataset with the time increment set to one second, and instruments with greater time intervals were given repeating values to reflect the more-integrated sampling. Although the spatial allocation of the measurements with longer time intervals is not as refined as for those with the one second time resolution, the road segments are sampled differently each time the route is driven and therefore the multiple repetitions of the route have good potential to spatially resolve the concentration patterns to finer scales than the distance traveled at these times would imply. An additional flag was used to indicate the likely impact of CRUISER's own plume when it was at stop sites (Fig. 1), so as to exclude such measurements in the analysis. Data from multiple studies indicate that this impact was most likely occurring

when the vehicle speed was <10 km h^{-1}, brief peak concentrations are observed with NO levels ≥ 20 ppbv and particle counts > 3000 #/cc. This resulted in 4% of the mobile measurements being flagged, of which those occurring at stop sites were excluded.

2.3 Mobile Measurement Strategy for Estimating Longer-Term Averages

The strategy for CRUISER's mobile measurements was to travel along pre-defined routes, passing along or near highways, main roads and local streets, as well as residential, commercial and industrial areas (Fig. 1c). Two routes were determined and systematically followed: 1) East Montreal; 2) Central and west Montreal. The east route was used most often due to greater impact of industrial emissions and because of an asthma study being conducted in that part of the city (Dobbin et al., 2011). For both routes, the starting point of travel along the route was changed randomly each day so as to avoid sampling the same location at the same time of day. On any given mobile measurement day, which was typically between 0900-2000 hr (i.e., nighttime conditions were not part of the mobile campaign), the entire route was completed, while on a few days the route was covered twice or 1.5 times with the reminder completed the next day. This allowed for multiple samplings of the same locations on different days, times and seasons, but with common days among all locations for key parts of the city (i.e., East Montreal). The goal was to have sufficient measurements spread over days and seasons so as to increase confidence that the longer term averages derived for any given part of the route and differences between locations were representative of typical conditions. This is assessed in the first part of the results.

The median travel speeds calculated from the GPS were 20, 28 and 30 km hr^{-1} for winter, summer and autumn, respectively. Due to these slow traveling speeds, CRUISER

was rarely on highways when in such areas and instead tended to drive on service roads parallel to the highways at some distance from the busiest lanes, sometimes with a dividing wall (2-5 m high) separating the express lanes from the service road. Figure 1c shows that a large majority of the sampling was conducted in residential areas, where local traffic was at a minimum, and other areas where the population spends time.

The mobile routine also involved measurements at 17 pre-defined parking locations (stop sites), such as government AQ monitoring sites. These stationary sites were visited for 10-30 minutes each time to create a selection of "point samples". Meteorological measurements were only possible during these stationary periods due to the 10 meter height of the telescoping tower. When stopping near a site was not possible (e.g., wind was too stagnant to allow orientation of CRUISER to avoid its own plume), the vehicle drove at slow speeds 'circling' the location by driving around the local block. These measurements are referred to as 'pseudo-stops' and all data were flagged to note times when CRUISER was stationary, pseudo-stationary or mobile.

There were 11, 17 and 6 mobile measurement days in the winter, summer and autumn, respectively, with 2-13 hours on each day (median of 9 hours). Figure 1c shows the number of measurements per km of road for the entire campaign; indicating more than 2,000 one second measuring points km^{-1} along the pre-defined route. The greater the number of visits to an area the more representative the available data will be of the long term. Xu et al. (2007) used Canadian monitoring data from multiple cities to show that the number of random visits needed to estimate the long term average with a desired percent error (e.g., 20%) depends upon pollutant. Fewer visits are required for NO_2,

while more are needed for NO or CO. About 20 one hour visits per season are needed to have a 95% chance of estimating the annual average NO_2 with 10% error.

The eastern route was completed 26 times, of which 11 were in the winter, 14 in the summer and 1 in the autumn. Based upon Xu et al. (2007) this translates, approximately, into 95% confidence that the estimate in annual average NO_2 is within 15% of the actual value for the summer and winter. Actual long-term representativeness is assessed below and as discussed above is important. However, it is useful to note that the spatial patterns and particularly the pollutant interrelationships observed are expected to be a reliable representation of the conditions in Montreal given that all locations were visited on the same days and all pollutant measurements were obtained simultaneously. In fact, the spatial patterns shown in this paper are believed to represent an improvement over most other mobile studies of spatial patterns and near roadway gradients (e.g., Beckerman et al., 2008; Hagler et al., 2010; Massoli et al., 2012) given the large number of measurements for multiple pollutants obtained in a systematic nature on multiple days and at different times in the year across a large, complex city.

2.4 Comparison to the Air Quality Monitoring Network

Stops and pseudo-stops next to seven different AQ sites (Fig. 1a) were used for comparison between the mobile and routine measurements (i.e., government monitoring network sites). In addition to providing some measure of quality control and assessment of comparability, this was done to provide a link between the mobile lab work and the traditional monitoring such as for obtaining insight into the spatial representativeness of the AQ network site locations. These concurrent, side-by-side measurements were also necessary to be able to quantify the long term representativeness of the average

CRUISER measurements derived from our deployment and data analysis strategy. One minute readings of CO, NO, NO_x, SO_2 and O_3 and one hour readings of $PM_{2.5}$ were provided by the local government agency (Ville de Montreal; 'VdM') for the periods corresponding to the CRUISER mobile campaign and hourly VdM data were obtained for all other times in 2009. Results of this direct comparison are given in Supplemental Material E.

Two different 2009 VdM annual metrics were calculated to represent the long term pollution levels: the daily average (i.e., all 24 hours) and the daytime average (i.e., between 0900-2000 hr), which better corresponds to the times of day CRUISER was driving. These two long term metrics are compared to two different short-term averages; (1) the VdM daily averaged values on the days CRUISER took mobile measurements during the nine separate weeks it spent in Montreal and (2) the average of all CRUISER stops and pseudo stops after eliminating minutes suspected of being impacted by the mobile lab plume. These CRUISER data were first averaged according to visit and then for all visits at an AQ site so that each visit received equal weight.

2.5 Spatial analysis

Since CRUISER`s measurements were taken along roads and because of the sharp gradient in concentrations of some pollutants with distance away from the road (e.g., Karner et al., 2010), analysis of the measurements was done by grouping them according to road segment, using ESRI's ArcGIS 9.3 (ESRI, Redland, CA). First, the road network of Montreal was used to extract only the roads CRUISER had visited, so as to avoid attributing measurements to the crossing roads. Then, each of CRUISER's measurement points were assigned to the nearest road segment within a distance of 20 meters. Samples

were averaged per road segment first by day and then averaged for all days, so as to give an equal weight for every day and avoid bias towards days with more measurements (e.g., greater measurements on a segment due to slower driving speed).

To assure proper representativeness of the measurements at each road segment, only segments that meet the following criteria were used in the analysis: more than 100 valid one second measurements of the examined pollutant, more than 100 valid measurements per km of the examined pollutant and that the measurements were spread over 3 or more different days. Most road segments included in the analysis were sampled on 17 different days or more. Statistical analysis was done with the open-source statistical language R (RDCT, 2009).

Since CRUISER is inherently limited to taking measurements while traveling on roads, an argument can be made that these measurements are limited in that they only represent road emissions and traffic related pollution and not exposure levels at a home address, which are often used in health studies, for example. However, for 38% of the time CRUISER took measurements on local roads in residential areas, where it met few other vehicles and therefore measured ambient residential pollution levels. Similarly, even on busier roads, there were many periods when cross-winds blew the air and pollutants from over the areas adjacent to the road to CRUISER's inlet. This implies that the dominant impact on the mean road segment concentrations was not solely the result of very local emissions associated with nearby traffic, but instead tended to be representative of a realistic mix of these local emissions and general "neighbourhood" conditions over that part of the driving route.

3 Results

In this section, we first compare the measurements obtained by CRUISER to routine measurements taken by the AQ monitoring network to compare campaign averages to the actual annual average (i.e., to assess representativeness). We then examine the spatial variability of several pollutants at the sub-urban scale with respect to their emission sources and quantitatively examine the representativeness of air quality monitoring sites to various microenvironments in their vicinity. We then compare the rates of decay of several pollutants with distance from a highway based upon campaign average observations.

3.1 Representativeness of the Mobile Measurements

Although important for mobile surveys to undertake, comparison to AQ network sites is complicated by the fact that there can be differences in the measurement methods used. Results of direct comparisons of concurrent CRUISER and VdM measurements are described in Supplemental Material E and here the focus is on assessing long-term representativeness. Figure 2 shows a comparison of CRUISER's mean NO_x during stops near VdM monitoring sites to VdM's concurrent measurements, to VdM's 2009 daytime averages and to VdM's 2009 daily averages. Ratios of these different metrics are given in Table 2. This one pollutant (NO_x) is selected because, due to the methodological differences discussed in Supplemental Material E for each pollutant, the most confident comparison of CRUISER's estimate of the annual average in each area to the actual AQ network values is expected to be based upon NO_x. Comparisons of CRUISER vs. the VdM 2009 averages for the other pollutants are shown in Fig. SM-A1 in the supplemental material.

The VdM average among all the driving days was calculated to determine if, collectively, the driving days in each season were atypical of the annual averages (ratios C/D and B/D in Table 2). Table 2 shows that on the selected driving days NO_x tended to be higher on average by 18%, compared to the 2009 daily averages. However, the overall ranking among the sites during these days was similar to the annual pattern (Fig. 2). For the other pollutants (CO, $PM_{2.5}$, SO_2, O_3, NO and NO_2) the average difference between the study period VdM observations and the annual average among the sites with measurements were 9%, 16%, -1%, -23%, 28% and 12%, respectively (Table 2). Thus average concentrations on the days that the mobile measurements were conducted tended to be biased high compared to the 2009 average, except O_3, which was biased low. However, in terms of combustion pollutant levels (NO_x), the average high bias was 18% for the period, while for NO_2, which is often of most interest as an exposure indicator, the bias was smaller, at 12%.

CRUISER's deployment strategy of limited, but random collection of daytime measurements at and around each VdM site also tended to yield reasonably representative values for NO_x. Table 2 shows the ratio of CRUISER's estimates to the 2009 daily average (ratio type A/D) was between 23% and -24%. Furthermore, despite even fewer measurements at AQ5, CRUISER's campaign average observations correctly identified this site as having the highest NO_x. Consistent with the 2009 average pattern CRUISER also showed that the variability among all sites except AQ5 was relatively small. However, CRUISER observed a greater variability than tended to exist. Further focusing on the two VdM sites measuring NO_x where there was >18 visits, which corresponds to the survey in east Montreal, the observed range was 16.1-16.6 ppbv (2009 daily average)

and CRUISER reported a 17.4-19.8 ppbv range. Information on the differences between CRUISER short-term estimates of the average and the actual 2009 average for the other pollutants is shown in Fig. SM-A1. These comparisons should be interpreted with caution, however, given the measurement method differences discussed in Supplemental Material E and the overarching issue of limited data at most sites except 2-3 in the east. Figure SM-A1 shows, for example, that for NO_2 CRUISER was within 20%, on average. This is reasonably close, but 5% less accurate than the expected error upon Xu et al., (2007). As expected, for the sites only visited 8 times the CRUISER longer term average NO_2 estimate was biased low by up to 35% compared to the true annual average.

3.2 Intra-urban variability observed by CRUISER

The multiple pollutant measurements on CRUISER allowed for focus on differences in campaign average spatial patterns among pollutants. This is exemplified for four pollutants (NO_2, particle number concentration (PNC), SO_2 and Benzene) in the east Montreal maps shown in Fig. 3. Traffic related pollutants, such as NO_2 and PNC, show highest concentrations near the highways (e.g., NO_2 mixing ratios of up to 80 ppbv and PNC counts of up to 215,000 #/cc near Highways 40 and 25 shown in Figs. 4a and 4b). Lower concentrations were measured on major roads (12-28 ppbv NO_2 and 30,000-55,000 #/cc PNC) and the lowest in residential areas (<12 ppbv NO_2 and <30,000 #/cc PNC). These differences are consistent with the reported emissions for Quebec in 2007, that the transportation sector accounted for 74% of NO_x emissions, compared to 15% from industrial sources, including 2% from oil refineries (Busque et al., 2009).

Other pollutants that have a significant contribution from industrial emission sources, such as SO_2 and Benzene, show a different intra-urban spatial pattern, with high

concentrations around the industrialized eastern part of the city between highways 40 and 138 (Figs. 4c and 4d, respectively; roads identified in Fig. 1b). For SO_2, Busque et al. (2009) report that, provincially, transportation accounted for only 11%, compared to 7% from the oil refineries, with the aluminum smelters contributing 30% and other industrial sources contributing 24%. With no aluminum smelters on the Island of Montreal and transportation being spread over the entire road network, oil refineries and other industrial point sources have a dominant contribution to SO_2 as is seen in Fig. 3c. Benzene emissions are not reported independently in Busque et al. (2009), however for volatile organic compounds (VOC's) transportation accounted for 37%, gasoline and diesel marketing for 5% and other industries for 27%.

Sulphur dioxide and VOCs such as Benzene are of interest in East Montreal given their link to the refining and petrochemical industries. The mobile measurements indicate that even though broad similarities for these two pollutants are evident over the urban scale shown in Fig. 3, there are differences in their behaviour on smaller scales. For example, the peak area with SO_2 concentrations (marked A in Fig. 3c) is located further north-east than the Benzene peak area (marked B in Fig. 3d). Also, the area of elevated SO_2 identified as 'C' is not accompanied by higher Benzene. Instead, this area appears to be accompanied by higher NO_2. These elevated campaign average concentrations are hypothesized to be indicative of the influence of ship emissions and/or diesel trucks servicing one of the port areas, given the proximity of this portion of the driving route to the St. Lawrence River and the large port area for container ships. Apart from the four pollutants described above, spatial patterns of all the other pollutants measured on this study were evaluated (not shown) and have been used in subsequent

analyses (Brook et al., 2013; Levy et al., 2014). Spatial differences are examined further based upon urban micro-environment and distance from the highway in the next section below.

Levy et al (2014) reported on the spatial correlations among all the parameters measured across Montreal highlighting which pollutants tend to co-vary and which are associated with different sources. From Fig. 3 and Levy et al. (2014) it can be seen that some mixes of pollutants show affinity to roads (BC, CO, HOA, NO, NO_x, OM, PM_1, $PM_{2.5}$, PM_{10}, PNC) and others are related to industry (SO_2, Toluene, Xylene). Clearly, a range of interesting features related to sources and potentially more-relevant to chronic exposures are revealed through CRUISER's deployment approach. Another example is the port area mentioned above (high SO_2 and NO_2). However, describing in detail how the multipollutant mix associated with the large number of potential sources varies is beyond the scope of this paper. Furthermore, the behavior of the mixtures varies on multiple spatial scales, each requiring follow-up analysis. For example, differences in the variation in SO_2 and Benzene concentrations along a single road crossing Highway 40 and passing next to the Petro-Canada oil refinery to the north and the Ultramar oil distribution terminal to the south (Avenue Marien, location shown by arrow in Fig. 3a) shows that some peaks in concentration occur together while others do not (Fig. SM-A2). Furthermore, ratios of Toluene to Benzene in the peaks and between peaks also vary. These small scale differences are indicative of how different processes within a single large industrial facility (refinery) may differentially impact surrounding locations with systematically different pollutant mixtures.

3.3 Microenvironments and sub-neighbourhood scale variability

In order to characterize the extent that chronic exposure levels can vary among a range of typical settings in an urban environment, ten small East Montreal areas that were visited by CRUISER were selected and compared (Fig. 4). The distributions of the campaign averages among these different settings are also compared to measurements taken by CRUISER when at the nearest AQ site during stops and pseudo-stops. These sites are included in Fig. 4 to provide insight into the representativeness of the monitoring site locations and hence the possible error or bias resulting from assigning or modeling chronic exposure estimates based only upon available monitoring network data.

The ten areas were selected so that they represent a range of urban microenvironments while also being relatively close in proximity to one another, thus reflecting the activity space over which individuals might typically travel, especially children and the elderly. Their locations within the city are shown in Fig. 1b, labeled as A-E, and in greater detail in Fig. 4. The areas selected include: the intersection of two main roads - Rue Sherbrooke East and Blv. St-Jean-Baptiste (A1); an active commercial area on Rue Sherbrooke East (A2) near A1; a residential street (Rue Forsyth) ~80 m east and parallel to A2 (A3). These are located on the east side of the island in proximity to the oil refineries and are compared to the nearest monitoring site (AQ1, within 1.2 km of all these areas).

In the middle of Fig. 4 another grouping of sites is shown in comparison to their local monitoring site (AQ2), at distances of 0.7-1.0 km. The settings for these five areas are: a moderately busy road in a residential area (Anjou) next to houses (B1); a section of the same road next to an active commercial area (B2); a local road in a residential area next

to a service road and a busy highway (HW-15), but with a noise blocking wall separating the highway and the service road from the local road (C1); near an exit ramp of the local major highway (HW-15) bordering the residential area without a noise blocking wall (C2); a street corner in a residential area with a small car repair shop (D) in Anjou. The pair B1 and B2 is located east of the busy interchange of Highways 40 and 25.

The last grouping of microenvironments, shown on the right side of Fig. 4, corresponds to monitoring site AQ3 at a distance of 2 km or less. These two areas are located west of the other areas and are generally upwind from the major stationary emission sources and traffic corridors and include a residential street (E1) and an intersection of two local roads (E2) some 500 m from E1.

Each area in Fig. 4, as well as the AQ sites, were visited an equal number of times, mostly on the same days and in all seasons. The distributions in Fig. 4 show all of CRUISER's measurements for these periods, first averaged by day and then presented in box plots for all days for different pollutants. Visually, differences in the levels among the areas are evident. Significant differences between the areas were assessed for each pollutant separately by the non-parametric Kruskal–Wallis test followed by the Wilcoxon Rank Sum test for pairwise group comparisons. Areas with a similar letter are significantly different (p-value<0.05). Based only upon the distributions of the values averaged by day for each location the differences were only significant for NO_2 and BC. However, due to the small sample size for these tests (<20 days) the analysis was repeated with the actual 1 second data. As shown in Fig. SM-A3 many more significant differences are detectable for all pollutants examined using the higher resolution data.

The mean NO_2 mixing ratios (red squares in Fig. 4) at the traffic affected areas A1 and A2 are double the 10 ppbv measured next to the nearest monitoring site, AQ1, while the residential area A3 has mixing ratios similar to AQ1. This indicates that the monitoring site under-estimates the exposure levels for A1 and A2 while providing a representative estimate for A3. AQ1 also under-represents $PM_{2.5}$, BC and PNC for A1 when examining both median (A1 is higher by 49%, 21% and 60%, respectively) and mean (A1 is higher by 78%, 68% and 128%, respectively). Comparing the busy intersection A1 with the active commercial area A2 and the residential street A3, concentrations of NO_2 are higher for A1 than A2 and for A2 than A3 (Fig. 4a), as can be expected given the characteristics of those areas. The same differences in the distributions are also seen for PNC for these three areas (Fig. 4e).

NO_2 measurements next to the main road microenvironment at C1 and C2 are considerably higher than the monitoring site AQ2 with the median (mean) higher by 10 and 14 ppbv, respectively (20 and 16 ppbv, respectively), an increase of 210-265%. Ozone at C1 and C2 is showing a corresponding decrease of 32% from AQ2 levels. BC and PNC also have higher concentrations at C1 and C2 compared to AQ2 of more than 180% for the median and 200% for the mean of BC and 300% for both mean and median of PNC. The mean $PM_{2.5}$ next to C2 is also higher than AQ2 by about 20%, providing evidence of its spatial variability. The resemblance between C1 and C2 for these pollutants, however, implies that on average the noise blocking wall in C1 has little effect on air pollution. Comparing areas E1 and E2, higher values of NO_2 (13.6 vs. 9.4 ppbv), BC (3.1 vs. 2.0 $\mu g\ m^{-3}$), PNC (27,950 vs. 21,575 #/cc) and Toluene (0.63 vs. 0.48 ppbv) and lower O_3 (18.1 vs. 19.3 ppbv) are measured at the intersection area of E2 compared

to the residential area E1 near it. Clearly, any time spent at such an urban microenvironment enhances exposure and leads to greater misclassification if exposure is assigned according to the nearest AQ site.

Last, area D shows similar values to what is measured next to the monitoring site AQ2 for most pollutants, with the exception of Toluene. The presence of a small paint and body shop in that area caused the high values of this organic compound with a mean of 1.14 ppbv. Toluene levels in this localized microenvironment were 2-3 times higher than at the nearby monitoring site AQ2 (0.37 ppbv), and versus the nearby areas B1, B2, C1, C2 (0.40, 0.45, 0.53 and 0.45 ppbv, respectively). The levels were even somewhat higher than the mean measured at areas A1-A3 (0.76, 0.78 and 0.69 ppbv, respectively) and the monitoring site AQ1 (0.98 ppbv), which are in closer proximity to the petroleum industry. The difference between area D and the others for Toluene is much higher in the summer season (not shown), probably due to greater evaporation of this compound in higher temperatures and the fact that the shop more often operated with open doors in the summer.

3.4 The impact of a busy highway: Near road to deep into residential areas

The comparisons shown in Fig. 4 provide some information on the increase in concentrations associated with proximity to traffic. To more-directly assess how the presence of a major highway impacts multipollutant exposure within an adjacent neighbourhood the measurements along a 1200 m cross-section perpendicular to Highway 40 in Anjou were isolated for further study. The observations start at a service road running parallel to Highway 40 and continue along Avenue Azilda (a one way street in a residential area with little traffic), as shown by an arrow in Fig. 1e. This cross-

section was sampled at different times of the day on 15 separate passes in the summer, 9 in the winter and twice in the autumn and includes varying wind conditions (i.e., measurement periods were not selected to always represent downwind conditions, although prevailing wind does place Avenue Azilda downwind more often than upwind). Measurements along this cross section for the whole campaign were aggregated according to 20 m bins and the median and inter-quartile range (IQR) per bin are plotted according to distance from the center of Highway 40 in Fig. 5. Along this cross-section there were occasional streets crossing perpendicular to Ave. Azilda and these are indicated by arrows across the top of the figure. We also added the mean of all measurements taken on the highway (east and west bound combined) as a point marker for indicating the on-road values, though these are not co-measured with the cross-section measurements. Also shown for comparison (blue lines in Fig. 5) is the synthesis of 41 studies of near-road gradients presented by Karner et al. (2010) and (in black lines) spline smooth curve fits to the CRUISER median concentrations in order to remove small scale variability and help reveal the shape of the observed decreases in concentration.

With the exception of ozone, all pollutants in Fig. 5 show a decrease in concentration away from the highway for the campaign average. Some pollutants show a rapid decrease of more than 60% within a few tens of meters (i.e., decreases of 85%, 83%, 63% and 60% for NO, PNC, BC and CO, respectively, after 70 m), while others show a more moderate trend (i.e., decreases of 50%, 43%, 55%, 30% and 30% for NO_2, $PM_{2.5}$, PM_{10}, HOA (combustion-related organic $PM_{1.0}$) and Benzene, respectively, after 70 m). There appears to be some contribution from emissions in the vicinity of the first cross street although the amount of traffic on this street was much less than the highway and service

road. After adjustment to the edge of road values measured by CRUISER, these decreases and the curve fits are in general agreement with Karner et al. (2010) for most pollutants (NO, PNC, BC and CO). However, CRUISER's measurements did tend to show a more rapid change.

For particulate matter ($PM_{2.5}$ and PM_{10}) the gradients measured by CRUISER are greater than what is shown by Karner et al. (2010). Even though a sharp near road gradient in PM was observed in the summer (not shown), the main contribution to the sharp drop in the campaign average data is the elevated particles measured near Highway 40 in the winter (not shown) in Montreal. On average, $PM_{2.5}$ doubled from the neighbourhood background to the near highway environment while HOA, a component of $PM_{2.5}$ which is expected to be a more-specific measure related to traffic fine particles (Sun et al., 2012), more than tripled in magnitude (see Fig. 6 described later).

There is evidence in the cross section plots that some pollutants (i.e., NO_2, CO and HOA) remain slightly elevated above the background up to ~700 m from the highway. This was much more evident among the winter measurements although they are not shown here given this paper's focus on longer-term, average pollutant patterns. The maximum distance of the potential highway impact was further examined by calculating the normalized cross sections relative to the neighbourhood background levels, assumed to be represented by the concentrations at 1000-1500 m away, which is where the levels reached a minimum and showed no evidence of increasing or decreasing (Fig. 6). The bins used in Fig. 6 include the highway from 20 m west of the highway center to 20 m east, accounting for the measurements on the highway from separate visits traveling east and west bound up to a dividing acoustic wall (3 m high) separating it from the parallel

service road. The bins continue with separate 50 m bins (i.e., 20-70 m, 70-120 m,…) from the service road and along the transect on Avenue Azilda.

CO was observed to decrease consistently to the background concentration at about 600 m from the highway, while NO_2 and HOA both appear to reach the background further away at about 700 m. There is an increase in some pollutants around 360 m from the highway which is most noticeable for NO, $PM_{2.5}$ and BC (Figs. 5b, 5e and 5g respectively) and is associated with a decrease in O_3 (Fig. 5c). This is likely due to the contribution of the crossing road at that location, thus providing some insight into the potential impact smaller roads can have on exposure. Although the other crossing roads were not observed to have such an effect, it is likely that the road at 360 m contributed somewhat to the pollutant levels further downwind on some days. For NO_2 the elevated levels observed by CRUISER up to 700 m are supported in the synthesis presented by Karner et al. (2010) in Fig. 5a with NO_2 gradually reducing up to at least 500 m where the data ends, but appear to still be decreasing. The similarity between the decrease in NO_2 and HOA suggests a link between NO_2 and the organic fraction of fresh traffic-related particles.

Scale-up factors (i.e., percentages of increase relative to the neighborhood background levels) for multiple pollutants were calculated as a function of distance away from the highway center as described above for Fig. 6. These scale-up factors, which may enable estimates of typical near-road levels relative to urban background monitoring sites, are tabulated in the Supplemental Material Table SM-A1. The factors include in them the apparent contribution of the crossing roads which (mainly the first) appeared to have a stronger effect on NO, O_3, BC and Benzene.

4 Discussion

In this study, strategic short term measurements taken with a mobile lab were compared both to the concurrent measurements at AQ monitoring sites in Montreal and to the annual averages at these sites to assess representativeness of the average concentrations derived from the mobile lab deployment. Determining how well a limited set of mobile lab measurements spread among three seasons captures the true long term pattern is complicated due to the fact that it is necessary to rely on the existing monitoring sites to determine the long term value, yet there are only a limited number of monitoring sites, which are often in urban background locations, that measurement methods are not the same between the network and the mobile lab due to different technical requirements and that the mobile lab is not always able to measure right next to the sample inlets at the sites. Despite these limitations we compared the averages of the short term (from CRUISER and from the corresponding day's 24 hr data from VdM) and 2009 daily and daytime average values at a number of sites. Overall, except for O_3, the study measurement days were found to have experienced higher than average pollutant levels. For example, NO_x, which is a good indicator of urban combustion pollution, was higher on average by 18%, compared to the 2009 daily average. Averages derived from CRUISER's systematic visits of generally one per day spread through three seasons were within ±24% of the 2009 daily average, thus only increasing the discrepancy by 6% despite much fewer data points.

Identification of the AQ sites with notably higher vs. lower annual average concentrations was generally possible with the limited number of mobile lab visits. For

example, both CRUISER's observations and the VdM data during the corresponding period correctly identified the location (AQ site) with the highest NO_x levels, thus providing evidence that through a systematic deployment approach major concentration gradients across the city can be quantified. However, there were discrepancies, particularly when the differences between the long term values at the sites were small. For example, the 2009 mean NO_x at four of the five sites visited only differed by a range of 1.3 ppb. Not surprisingly, the short term visits are not able to achieve the precision in their estimate of the annual average to similarly rank these sites within such a small overall concentration range. However, for characterizing spatial gradients and chronic exposure levels and for exposure model development detecting such small differences is less important than capturing the larger exposure differences that exist among various areas. While it is not surprising that AQ sites generally do not document the full exposure gradients, this also means that they are of less use for assessing whether the exposure gradients captured by a short term mobile lab survey are indicative of the true long term pattern.

Campaign average concentrations derived from CRUISER have provided a better picture of multipollutant spatial patterns and correlations (Levy et al., 2014) across multiple scales. They reveal systematic differences between neighbourhoods in the pollutant levels and in the characteristics of the multipollutant mixture that are related to variations in sources. The relatively large spatial coverage that mobile monitoring can provide also helps uncover changes in concentrations on very small scales and can even identify and characterize concentration hotspots resulting from small businesses operating in residential areas (e.g., dry cleaners, car repair shop or a restaurant), from

commercial areas and from intersections in a residential neighbourhood. Such small scale variations may result in large differences in exposures to air pollutants among the population, yet they are often not included in air quality models or other exposure models because of lack of detailed knowledge about the location and type of activity of small emitters or due to their small emission volumes compared to other major sources or inaccurate traffic data. Furthermore, our results show that local emitters may have an impact on a local scale that can be greater than that of major sources further away, even in comparison to levels closer to such sources.

The identification of unknown local air pollution hot spots is impossible with traditional measuring techniques (i.e., stationary AQ monitors), given that measuring sites are selected to either measure ambient levels or examine previously known emission sources. The ability to identify small scale emission sources and exposure hotspots with mobile measurements has been previously demonstrated (e.g., Fujita et al., 2013; Larson et al., 2007; Spengler et al., 2011). For example, Dionisio et al (2010) showed that measurements of $PM_{2.5}$ and PM_{10} at stop sites with multiple woodstoves were 30 μg m^{-3} and 85 μg m^{-3}, respectively, higher than the neighbourhood average at the same times in a study in Accra, Ghana. Larson et al., (2007) also obtained more detailed understanding of areas with higher exposures to woodsmoke emissions through mobile measurements in the Vancouver area. Levy et al. (2001) showed some evidence of elevated $PM_{2.5}$ near diesel buses in a walking-mobile measurements study in Roxbury, Massachusetts, United States. Apart from the mobility aspect of these campaigns, which allows them to cover larger spatial domains, another advantage of mobile measurements is that they typically use shorter averaging times for the measurements of few seconds to few minutes (e.g.,

one second in this study and one minute by Levy et al. (2001)). This enables them to map fine spatial structures needed to identify hot spots.

Mobile labs are also a particularly useful platform for measuring spatial gradients in concentrations relative to sources and to study small scale processes. In this study, average CRUISER measurements from 26 independent days taken perpendicular to Highway 40 through a residential area have led to new insight into chronic exposure gradients. While all pollutants (with the exception of ozone) decrease in concentrations away from the highway, the rate of decrease differs by pollutant as is the distance that they spread into the adjacent neighbourhood. For several pollutants (i.e., NO_2, CO and HOA) there is evidence in our measurements that on average they remain elevated above the background for up to 700 m. It should be noted that in Anjou some additional emissions (i.e., cross roads) may have played a role in the CRUISER observations, but these were relatively small compared to the highway. With HOA being associated with traffic fine particles (Canagaratna et al., 2010), this may hint to one reason for why NO_2 has often been used successfully as a traffic exposure indicator in assessing traffic-related health effects (e.g., Crouse et al., 2010; Jerrett et al., 2009). Firstly, the elevated NO_2 associated with a traffic source affects a larger percentage of the neighbourhood compared to some pollutants (e.g., PNC and NO). Secondly, if this greater extent of higher NO_2 is also accompanied by higher CO and HOA (and possibly other unmeasured pollutants such as trace metals or other more toxic organic particles) up to 700 m from a highway then the effects attributed to NO_2 are possibly due to these and other similarly-behaving pollutants and/or some or all of the mixture of particles and gases.

Greater spread of traffic particles has been observed elsewhere under certain conditions (e.g., Hu et al., 2009). Similarly, in Montreal it was wintertime conditions that favoured a greater spread of air pollutants (not shown), which was likely due to lower mixing heights and longer lifetimes of some pollutants due to less evaporation (i.e., colder temperatures) and/or less photochemistry. Even though the measurements were obtained from multiple days and seasons, which has rarely been done, the prevalence of the behaviour observed by CRUISER in the Anjou neighbourhood of Montreal and its relevance to other areas is unknown and thus some follow-up study should be considered to strengthen their value to exposure model development and possibly to guide urban planning. Nonetheless, here we have reported average concentration scale-up factors relative to urban background in Montreal as a function of distance from the highway as a potential first step in generalization to support future use.

Due to its relatively long lifetime in the atmosphere PM is generally considered to be spatially homogeneous over distances of a few kilometers (e.g., Martuzevicius et al., 2004), although Beckerman et al. (2008) has shown significant increases close to a major highway in Toronto. Here we also show that $PM_{2.5}$ and PM_{10} are affected by proximity to roads. This is also reflected by two important $PM_{2.5}$ constituents; BC and HOA. The impact of the highway was particularly evident in the winter, when higher concentrations were measured up to 370 m away due to the combined effect of a more stable atmosphere near the ground (Bergeron and Strachan, 2012) and the use of road salt and sand for tire traction (Gertler et al., 2006). However, compared to the other pollutants explored in this paper and in Levy et al. (2014) average $PM_{2.5}$ concentrations were, overall, more homogeneous and, with the exception of near the highways, the observed peak

concentrations appeared to be localized, short-lived and difficult to explain in terms of their source(s).

The capabilities of physically-based models to determine urban spatial patterns have improved in recent years. For example, the ability of comprehensive, multipollutant air quality models to run for longer periods at grid resolutions of 1-5 km is increasing (e.g., Makar et al., 2010; Shrestha et al., 2009). Output from these higher resolution models may eventually lead to multipollutant information relevant to chronic exposure estimation. However, as shown in this paper and most other mobile lab studies, there remains a large and complex variability even within the finest model grids (at best ~1 km^2 area) in use today. For example, concentrations reduce from 20-80 ppbv for NO_2 and 55,000-215,000 #/cc for PNC to less than 12 ppbv and 30,000 #/cc, respectively, between a highway and a residential area over a distance of less than 1 kilometer in the residential area of Anjou. While such gradients are being predicted for a limited number of pollutants by dispersion or LUR models or other hybrid-models, as discussed in the introduction, stationary monitoring networks clearly cannot account for them. Although multipollutant mobile studies such as described in this paper can help provide the information needed, they are not feasible in many locations (mainly due to the high costs involved). The relatively recent emergence of portable technologies for measurements of environmental parameters (i.e., coupling portable digital devices and GPS with microsensors for air pollution (Mead et al., 2013) atmospheric parameters and biomonitoring in distributed stationary or mobile sensors networks) may help fill the gap in our ability to estimate the spatio-temporal variability at the intra-urban scale, though

likely with some cost to measurement accuracy or precision and only for certain pollutants.

5 Summary and Conclusions

Although mobile measurement campaigns have been used often to characterize intra-urban variability in air pollution, most have been based upon short periods and it is not known if the patterns observed reflect typical or long term conditions. When applied for evaluating average or long term spatial patterns a mobile lab has two main limitations: its inherent inability to take simultaneous measurements at multiple locations and the limited number of measurement days that can be done at multiple locations due to cost. This study evaluated and applied a mobile monitoring approach that was designed to reduce the effects of these limitations in order to infer intra-urban variability of multiple pollutants representative of long term exposure patterns.

The approach we evaluated was based on repeated measurements along the same route covering a wide variety of urban micro-environments on multiple days, different seasons and different times of the day. This approach has the advantage of covering a relatively large spatial domain that surpasses what an AQ monitoring network can achieve, even monitoring enhanced by additional satellite sites. Comparison of the average concentrations derived from the mobile lab at permanent monitoring sites showed that when a site was visited 18 or more times the actual annual averages could be estimated to within 25% for NO_2, NO_x, CO and O_3 and within 30% for $PM_{2.5}$. Maximum representativeness errors were almost 60% for SO_2 and over 100% for NO. As expected, errors tended to be greater when fewer visits were completed. The mobile survey also correctly identified the larger spatial differences seen between some of the AQ

monitoring network sites. Furthermore, comparison of mobile lab average concentrations across the full driving route in East Montreal showed that concentration variations observed among locations were considerably greater than the representativeness errors. This suggests that the deployment approach undertaken with CRUISER was sufficient to detect true spatial differences in average concentrations. Therefore, we conclude that it is feasible for a mobile lab survey to provide a large amount of spatial data to inform chronic exposure assessment potentially for better understanding of the distribution of population risk and for developing exposure models. Ideally, more long-term sites measuring a greater number of pollutants and across contrasting locations are needed in future studies to be able to carry out a more detailed evaluation and optimization of mobile deployment strategies designed to characterize long-term patterns.

Mobile labs, such as CRUISER, have a greater capability to characterize spatial patterns for multiple pollutants compared to intensive fixed site monitoring. The results shown in this paper, based upon best estimates of longer term averages, have uniquely quantified the magnitude of exposure differences for more pollutants simultaneously than has previously been studied. In Levy et al. (2014) these differences were assessed in terms of multipollutant spatial correlations. In this paper, these differences were assessed in the context of evaluating the representativeness of routine monitoring locations in Montreal and in the context of impacts from a complex mixture of emission sources. This included mobile sources, marine sources, heavy and light industry, small businesses and residential heating, including wood smoke. Long term average concentrations were also examined in terms of the impact of a busy highway deep into an adjacent residential area, also comparing gradients among pollutants. The CRUISER deployment also

involved a large coverage of East Montreal to provide observations representative of longer term conditions over a diverse range of urban microenvironments.

Through examination of both maps depicting the overall variability for several different pollutants and of small scale co-variations in pollutants we identified how the impact of sources varies spatially. For example, while SO_2 was well-known to be emitted from the petrochemical industries, with concurrent measurement of benzene and NO_2, we were able to show that in some parts of East Montreal SO_2 was associated with a different source, more likely marine vessels. Closer to the petrochemical facilities we showed how the heterogeneous nature of such facilities leads to small scale (i.e., ~100 m) differences in the mix of pollutants affecting the surrounding area. Thus, systematic mobile measurement of multiple pollutants can be a valuable approach for gaining more insight into which sources are having a greater long-term impact on local air quality and how these impacts vary on relatively small spatial scales.

Through examples comparing a range of locations to each other and to the nearest monitoring site we demonstrated, quantitatively, how much exposure can be influenced by spending time in different common microenvironments such as commercial areas and intersections of busier roadways. The mobile survey demonstrated that the routine monitoring sites are providing representative information on neighbourhood background conditions throughout the portion(s) of East Montreal they are expected to represent. However, relatively short distances away (i.e., 100 m) from locations experiencing background conditions, in areas where residents can be expected to spend time, concentrations for some pollutants (e.g., NO_2, PNC) can be a factor of two higher. The extent of this small scale variability depends upon source. The distribution of traffic

leads to the greatest local scale variability, while the impact of industrial facilities tends to vary on larger scales (i.e., 500 m). Although, as described above, the nature of this impact can vary on smaller scales in the case of large facilities involving a range of processes.

While a large amount of data are available on traffic air pollution gradients downwind of roadways, the CRUISER measurements are unique due to the relatively large number of different days and times the measurements represent, the simultaneous measurement of 23 separate parameters and the greater distance away that was explored. From these data we showed that the rate of decay in concentration with distance differs by pollutant. This is related to differing atmospheric processes occurring on short time scales. Moreover, we reported some evidence suggesting that while concentrations of some of the pollutants decay to background levels within a relatively short distance (<50m) from a highway, some (NO_2, CO and traffic-related organic particles (HOA)) remain elevated up to 700 m. Clearly, the mixture of traffic-related air pollution is complex and variable on multiple scales and not all the details were explored in this paper. However, of importance here is that some aspects of this mixture, and likely unmeasured components, may be felt above urban background levels on a long term, chronic basis significantly beyond 500 meters.

In order to improve understanding of the overall impact of chronic air pollutant exposures in urban areas, future studies examining intra-urban variability of air pollutants will need better exposure predictions than currently available. One possible path to follow is to combine several modeling approaches (e.g., dispersion, air quality and LUR models) as well as different measurement techniques (e.g., remote sensing and mobile

measurements). Furthermore, with improvement in publicly available databases on small scale emitters, emerging technologies (e.g., distributed sensors networks) and statistical modeling approaches, some progress on these challenges can be expected, although understanding combined and differential multipollutant effects will remain a significant challenge.

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8 Tables

Table 1: List of pollutants measured by CRUISER or derived from CRUISER measurements and the methods employed.

Parameter	Avl[a]. (%)	Instrument model	Time resolution	Detection limit
NO	80	Thermo Scientific / TECO 42CTL	1 sec	0.4 ppbv
NO_2	64	Thermo Scientific / TECO 42CTL with Photolytic converter	1 sec	0.8 ppbv
NO_y	62	Thermo Scientific / TECO 42CTL with Photolytic converter & Mo converter	1 sec	0.4 ppbv
NO_x	64	Calculated ($NO + NO_2$)	NA	NA
NO_z	46	Calculated ($NO_y - NO_x$)	NA	NA
SO_2	79	Thermo Scientific / TECO 43 TLE with a 5 µm pore size Teflon filter	10 sec	1 ppbv
CO	78	Thermo Scientific / TECO 48 with a 5 µm pore size Teflon filter	10 sec	100 ppbv
O_3	79	Thermo Scientific / TECO 49	20 sec	1 ppbv
O_x	53	Calculated ($NO_2 + O_3$)	NA	NA
PM_{10}	88	GRIMM Dust Monitor 1.100	6 sec	0.1 µg m^{-3}
$PM_{2.5}$	88	GRIMM Dust Monitor 1.100	6 sec	0.1 µg m^{-3}
$PM_{1.0}$	88	GRIMM Dust Monitor 1.100	6 sec	0.1 µg m^{-3}
particle number (PNC)[b]	87	GRIMM CPC 5.403	1 sec	0.6 # /cc
BC (Black carbon)	49	Droplet Measurement Technologies / Photo Acoustic	1 sec[c]	<3.3 µg m^{-3}

OM (Organic matter)	73	Aerodyne Aerosol Mass Spectrometer	2 min	0.15 µg m^{-3}
Sulfate	73	Aerodyne Aerosol Mass Spectrometer	2 min	0.04 µg m^{-3}
Nitrate	73	Aerodyne Aerosol Mass Spectrometer	2 min	0.02 µg m^{-3}
HOA (Hydrocarbon-like organic aerosols)	36	Aerodyne Aerosol Mass Spectrometer (PMF application[d])	2 min	0.15 µg m^{-3}
MZ57 (mass to charge ratio of 57)	36	Aerodyne Aerosol Mass Spectrometer	2 min	0.01 µg m^{-3}
Benzene	73	IONICON High Sensitivity PTR-MS	10 sec	20 pptv
C3 Benzene	73	IONICON High Sensitivity PTR-MS	10 sec	20 pptv
Toluene	73	IONICON High Sensitivity PTR-MS	10 sec	20 pptv
Xylenes	73	IONICON High Sensitivity PTR-MS	10 sec	20 pptv

[a] Availability: percent of valid measurements from all 1 second measurements.

[b] PNC measurements here refers to the commonly named UFP in other studies.

[c] Vibrations during mobile measurement result in a poor signal to noise ratio for 1 second resolution data of the Photo Acoustic's BC data. However, the analysis presented here aggregates multiple measurements and thus improves the detection limit. With a 30 min averaging time the detection limit improves to 0.71 µg/m^3 and road segment average values used in this paper generally represent averaging over at least 1700 seconds.

[d] For more information see *Description of the instrumentation inlets to CRUISER*

Table 2: Ratios between different average pollution levels shown in Figs. 2 and SM-A1.

	Ratio type[a]	AQ3	AQ1	AQ2	AQ4	AQ7	AQ5	AQ6	Average
	N (visits)	21	18	20	8	5	8	8	
NO_x	C/D		0.93	0.87	0.92		1.02	0.83	0.91
	B/D		1.17	1.20	1.20		1.09	1.22	1.18
	A/D		1.23	1.05	0.97		0.77	0.76	0.96
NO	C/D		1.01	0.89	0.94		1.06	0.81	0.94
	B/D		1.25	1.35	1.32		1.12	1.37	1.28
	A/D		2.04	1.80	1.76		1.64	2.69	1.99
NO_2	C/D		0.89	0.86	0.91		0.98	0.84	0.89
	B/D		1.14	1.13	1.15		1.05	1.15	1.12
	A/D		0.95	0.78	0.99		0.68	0.65	0.81
O_3	C/D	1.19	1.20		1.20		1.19	1.22	1.20
	B/D	0.79	0.81		0.76		0.73	0.78	0.77
	A/D	0.88	0.99		0.63		0.60	0.72	0.77
CO	C/D		0.94				1.08	0.90	0.98
	B/D		1.02				1.12	1.12	1.09
	A/D		0.99				1.61	1.47	1.36
$PM_{2.5}$	C/D	0.95	0.97	0.98	0.96	0.98	1.01	0.96	0.97
	B/D	1.16	1.12	1.36	1.13	1.10	1.14	1.08	1.16
	A/D	0.95	0.70	1.06	1.36	1.64	1.27	1.14	1.16
SO_2	C/D		1.02	0.98	0.98				1.00
	B/D		1.10	0.77	1.10				0.99
	A/D		1.37	1.56	0.99				1.31

[a] A: CRUISER's average during stops; B: VdM's average during CRUISER's measurement days; C: VdM 2009 daytime annual averages; D: VdM 2009 daily annual averages.

Figure legends:

Fig. 1: Map of the study area showing major roads, land use types, major emission sources and CRUISER's stop sites (a), with a higher resolution on the east part (b), as well as measurement density per kilometer of road segment (measurements / km) for the annual period (c) and at higher resolution on the east part (d) and the Anjou neighborhood (e). Letters A-E in b) refer to areas discussed in Section 3.3 and Fig. 4. Black arrow in (e) refers to the cross section discussed in Section 3.4 and Figs. 6 and 7.

Fig. 2: Comparison of CRUISER's average pollution levels during time it was parked next to VdM's AQ monitoring sites (gray squares), VdM's average levels during CRUISER's measurement days (red circles), VdM 2009 daytime annual averages (blue triangles) and VdM 2009 daily annual averages (green diamonds) for seven relevant pollutants. Whiskers denote one standard deviation. Number above whiskers denotes number of visits to site.

Fig. 3: Mean pollution levels for the entire study at road segments for NO_2 (a), PNC (b), SO_2 (c) and Benzene (d), along with National Pollutant Release Inventory (NPRI) reported point emissions for the relevant pollutants. Roads were filtered as described in section 2.4. NPRI emissions for NO_2 are nitrogen oxides expressed as NO_2.

Fig. 4: Box plots showing pollutant levels (NO_2, O_3, $PM_{2.5}$, BC, PNC and Toluene; a-f, respectively) at ten different areas (green) compared to measurements taken next to three air quality monitoring sites (blue). Red squares are the mean and numbers above each box mark the number of days included in the statistics. Right panel shows local settings of each area marked in blue (A-E). See Fig. 1b for locations within the city.

Fig. 5: Median pollution levels (red line) and IQR (grey) at 20 m bins for the entire study (a), summer (b) and winter (c) seasons, showing NO_2, NO, O_3, PNC, $PM_{2.5}$, BC, CO, traffic-related PM_1 (HOA) and Benzene along the cross section marked in Fig. 2d. Arrows mark the locations of perpendicular roads meeting the cross section. Blue lines show the change in pollution from edge of road based on Fig. 3 from Karner (2010) and adjusted to CRUISER's measurements at the edge of the road to facilitate comparison. Black lines are a spline smoothing of the measurements. Red points mark measurements taken passing through the highway.

Fig. 6: Normalized cross sections of NO_2, CO, HOA and $PM_{2.5}$ along the cross section described in the text and Fig. 5. Calculated as the median at each distance divided by the average of the 1000-1500 m medians. Full points mark measurements taken passing through the highway.