Assessing the Spatial Representativeness of $PM_{2.5}$ and O_3 Measurements from the National Air Pollutant Surveillance System

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Preface

This report has been prepared by its authors based on a scope of work prepared by Environment Canada. While the report has been reviewed by representatives of Environment Canada and the National Air Pollution Surveillance (NAPS) network as sponsors of the study, the findings and conclusions expressed in this report are the opinions on the consultant authors, and do not necessarily represent the opinion of Environment Canada or NAPS.

Provided along with this report are:

- One Refworks database containing all published materials found in the literature reviews conducted.
- One Excel file summarizing the individual studies used in the $PM_{2.5}$ and O_3 literature reviews.
- One Excel file containing all $PM_{2.5}$ and O_3 NAPS monitoring stations with derived siting characteristics.

Table of Contents

List of Figures

List of Tables

Executive Summary

Our literature review demonstrated that within-city spatial variation of fine particulate matter ($PM_{2.5}$) mass is case specific, and that in some cities significant spatial variation exists while in others $PM_{2.5}$ mass is spatially homogenous (15 studies reviewed identified within-city $PM_{2.5}$ mass concentration spatial variation greater than 20%, while 21 studies found $PM_{2.5}$ mass concentration spatial variation less than 20%). Primary $PM_{2.5}$, including Black Carbon and ultrafine particle mass and number concentration, demonstrated significant within-city spatial variation (20 of 22 studies reviewed identified within-city spatial variation in primary $PM_{2.5}$ greater than 20%). The average within-city spatial variation of primary $PM_{2.5}$ was greater than 200%.

In most circumstances, within-city ozone (O_3) variation is larger than within-city $PM_{2.5}$ mass variation; however, much smaller than that seen for measures of primary $PM_{2.5}$. Significant spatial variation in O_3 concentrations was seen at all spatial scales (local, neighborhood, city, urban/suburban and urban/rural) and due to scavenging by NO, large O_3 gradients are present at fine-scale spatial resolution (e.g. around major roads and other high traffic areas). Of the 26 studies reviews, 21 found within-city O_3 spatial variation greater than 20%.

Recommendations:

- **Statistical methods could be applied to data from the NAPS monitoring network to examine within-city variability when multiple monitors exist. Currently, 18 metropolitan areas have greater than two O_3 monitors and 23 metropolitan areas have greater than two $PM_{2.5}$ monitors. Correlation and coefficient of divergence measures could be calculated for monitor pairs using hourly, daily, monthly and yearly averages to identify areas where additional monitoring to characterize spatial variability could be prioritized.**
- **The use of $PM_{2.5}$ mass is not adequate for capturing primary $PM_{2.5}$ and masks significant within-city spatial variation that exists in measures of primary $PM_{2.5}$. Specific $PM_{2.5}$ components of concern should be considered when examining the influence of within-city spatial variation.**

We demonstrated that existing Canadian city-specific land use regression (LUR) models could be used to assess the spatial representativeness of NAPS O_3 monitors. Annual, summer,

and summer daylight models were able to explain 84%, 84%, and 79% of the spatial variation for 39 NAPS O_3 monitors. Small within-city O_3 spatial variation was present, however, likely due to monitor siting. From these models we illustrated how a spatial representative area could be defined for each monitor and show that these areas do not correspond to straight proximity (the most common extrapolation method used for NAPS data).

Recommendations:

- **Existing city-specific NO_2 LUR models can be used to examine O_3 spatial variability and define representative areas around each NAPS monitor; however, further work is needed to incorporate more predictor variables and to examine the spatiotemporal stability of the O_3 and NO_2 relationship.**

With regard to population coverage of the current $PM_{2.5}$ and O_3 NAPS monitoring network, 60% of the Canadian population is within 10km of a monitor. However, our preliminary analysis of NAPS monitor representative areas indicates that monitor proximity alone does not fully characterize population exposure.

We derived objective monitor siting characteristics (e.g. proximity to roads, road density, landuse, industrial emissions, population density) for various buffer distances around each monitor using geographical information systems (GIS) and nation data. There was generally poor agreement between the existing NAPS monitor classifications (e.g. agricultural, residential, industrial) and landuse characteristics derived in a GIS.

Recommendations:

- **NAPS monitor classifications could be refined to better provide information on source influences and representativeness. At minimum, a traffic influenced monitor code should be added to the current NAPS monitor classification. In addition, the full description of monitor siting characteristics should be made available with NAPS data as the siting characteristics of most importance will change based on different users. For this reason, simple monitor classifications can be misleading.**
- **Additional siting variables could be derived for NAPS monitors using other readily available geographic data.**
- **Land use may change around monitors over time and it is recommended that NAPS periodically evaluate and update monitor siting characteristics.**

Ideally, the spatial representativeness of NAPS monitors would be assessed using saturation measurements at various locations in space and time around each monitor. We reviewed measurement methods that could be used to assess representative areas and since complete monitoring coverage is not feasible, we also reviewed modeling methods. We briefly introduced each method and summarized their strengths and weaknesses for determining the spatial representativeness of NAPS monitors.

Recommendations:

- **A representative area should be defined for each NAPS monitor, either using measurements or modeling approaches, such as those demonstrated using existing NO_2 LUR models. Representative areas would inform use of NAPS measurements and their spatial extrapolation, and future monitor siting.**
- **Along with the representative area for each NAPS monitor, the population representativeness should also be determined (e.g. the percentage of the population in a CMA represented by the monitor). This would be useful for prioritizing NAPS stations and for future monitor siting.**

Background

Recent health research has demonstrated the importance of intraurban (within-city) spatial differences in air pollution levels and their relationship with measures of health impact (e.g. Brauer et al. 2008, Jerrett et al. 2005). This research has led to efforts aimed at improving within-city exposure assessment, particularly for traffic-related pollutants, such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) (Hoek et al. 2008). In Canada, within-city monitoring campaigns examining the spatial variation of NO_x and VOCs have been conducted in nine cities, all of which indicated substantial within-city spatial variation. In comparison, relatively little work has been conducted in Canada to examine the within-city spatial variation in fine particulate matter ($PM_{2.5}$) mass or composition and ozone (O_3).

National Air Pollutant Surveillances (NAPS) monitors are used in Canada to represent $PM_{2.5}$ and O_3 concentrations for areas of varying geographic size. This assumes that one monitor, or the average of a few monitors, is representative of the complex spatial and temporal patterns of air quality in that area. The assessment of the representative area of a monitoring station allows extending information observed at one point – the monitoring site – to a geographical area of representativeness. How well monitors represent the pollution concentration in a given area depends largely on local sources, topography and meteorology, monitor siting, and the spatial variability of the pollutant of interest. The assessment of spatial representativeness can be based on various sources of information including: (i) additional air pollutant measurements, (ii) modeled air pollutant concentrations, and (iii) spatial surrogate data (e.g. landuse characteristics and emissions sources).

Here we examine the spatial representativeness of NAPS $PM_{2.5}$ and O_3 measurements in Canada. First, we conducted a comprehensive literature review of Canadian studies documenting within-city $PM_{2.5}$ and O_3 measurements, as well as studies conducted elsewhere that specifically examine within-city $PM_{2.5}$ and O_3 spatial variability. Next we conducted a literature review examining the associations between the spatial variation of $PM_{2.5}$ and O_3 and NO_x. We then explored how models that capitalize on existing high-resolution spatial pollution models in Canadian cities could be developed and used to assess NAPS representativeness. Specifically we examine the feasibility of developing O_3 models from existing information on NO_2 spatial variability. Since spatial representativeness of a monitoring network is also influenced by monitor siting, we use a Geographic Information System (GIS) to examine surrogate emissions information (e.g. road proximity, landuse information) for each $PM_{2.5}$ and O_3 monitor in the NAPS network. Finally, we summarize measurement and modeling methods that could be used to more fully assess the representativeness of NAPS measurements.

Literature Reviews

We conducted literature reviews during January 2011 using Pubmed, Web of Science and Google Scholar. An Excel file is provided that summarizes individual studies, including 12 Canadian $PM_{2.5}$ studies, 57 international $PM_{2.5}$ studies, 23 Canadian O_3 studies and 38 international O_3 studies. A RefWorks database is also provided. Grey literature was included if identified through published or known studies; however, the primary focus of the literature review was published articles.

Fine Particulate Matter

$PM_{2.5}$ consists of particles ≤2.5μm in aerodynamic diameter originating from anthropogenic and natural sources, and may be further classified as primary or secondary pollutants (Wilson et al. 2005). Primary $PM_{2.5}$ is formed directly and is most commonly associated with combustion sources including traffic, industry and domestic heating. Several studies reviewed here examine Black Carbon (BC) as an indicator of primary $PM_{2.5}$ mass. Secondary $PM_{2.5}$ forms in the atmosphere through chemical and physical conversions of gaseous precursors such as NO_x, sulphur oxides, and VOCs. The major chemical constituents of $PM_{2.5}$ are sulfate, nitrate, ammonium, organic carbon, and BC, as well as a variety of trace metals formed in combustion processes and inorganic material of crustal origin (Bell et al. 2007).

Primary particles can demonstrate fine spatial variability, due to localized sources, while secondary particles affect regional and wider-ranging areas (Blanchard et al. 1999, Ito et al. 2004, Pinto et al. 2004). A large portion of $PM_{2.5}$ is secondary in origin, resulting in some degree of spatial uniformity across urban areas (Pinto et al. 2004); however, the level of uniformity is affected by several factors, including local sources of primary $PM_{2.5}$, transient emissions events, topographic barriers, meteorological phenomena, and differences in behaviors of semi-volatile components (Pinto et al. 2004).

Spatial heterogeneity/homogeneity of $PM_{2.5}$ is not well defined in the published or regulatory literature and as a consequence it has been interpreted in a number of different ways (Wilson et al. 2005). Here we defined $PM_{2.5}$ heterogeneity as >20% differences between intraurban sites (this cutoff was used by Blanchard et al. (1999) and is based primarily on a threshold that might be important from a health analysis perspective. We examined heterogeneity of $PM_{2.5}$ at different spatial scales, based on the classification scheme of Watson and Chow (2001), including: local (<1km), neighborhood (<1-5km), and urban (<1-50km). We

conducted a review of Canadian and international studies that monitored $PM_{2.5}$ concentrations within cities to determine how $PM_{2.5}$ concentrations vary at these different spatial scales.

Canadian $PM_{2.5}$ Studies

A limited number of studies have been conducted in Canada examining within-city $PM_{2.5}$ spatial variability with non-NAPS data. Henderson et al. (2007) created a land use regression (LUR) model of $PM_{2.5}$ and BC (absorbance) for Vancouver from 25 monitor sites in 2003[1] Automobile density within 100 meters significantly predicted $PM_{2.5}$, and major roads within 100m and truck density within 1km predicted light absorbance. The final model for $PM_{2.5}$ predicted 52% of the within-city variation and 41% for BC. Miller et al. (2010) measured PM_1 (range: 2.7-6.1µg/m^3), $PM_{1-2.5}$ (range: 0.6-7.6µg/m^3) and $PM_{2.5-10}$ (range: 0.02-7.8µg/m^3) at 15 sites in Windsor during 2008 and found low to high correlations between sites (PM_1=14%, $PM_{1-2.5}$=40% and $PM_{2.5-10}$=95%). Using semi-variograms they determined the spatial scale of PM_1 was approximately 6km's and $PM_{1-2.5}$ 10km's. Recently, Health Canada has conducted within-city monitoring campaigns in Ottawa, Hamilton, and Windsor (Data unpublished). In Hamilton and Ottawa PM_1, $PM_{1-2.5}$ and $PM_{2.5\ -10}$ were collected during 2009 and 2008/2009 and $PM_{2.5}$ in Windsor during 2006.

Other Canadian studies have used mobile monitors to examine specific sources of $PM_{2.5}$. Larson et al. (2007) monitored woodsmoke in the Greater Vancouver Regional District (GVRD) using mobile monitoring and found significant spatial variation that could be modeled with landuse characteristics and catchment areas (R^2=0.58). Lightowlers et al. (2008) examined the spatial scale of woodsmoke in the capital regional district (CRD) around Victoria and found spatial dependency up to 2.6km – indicating neighborhood level spatial variation. Thai et al. (2008) used mobile monitors to examine particulate matter exposure along bicycle routes in Vancouver and found relatively homogeneous $PM_{2.5}$ concentrations along routes, while PM_{10} and UFP displayed large spatial differences. Larson et al. (2009) also used mobile monitoring to develop LUR models for BC (absorbance) concentrations in Vancouver, and were able to predict a large portion of the spatial variation in the mean of the 25th and 50th percentile absorption coefficient measurements (R^2=0.72 and 0.68. respectively).

There have also been a small number of monitoring studies conducted in Canada that examine $PM_{2.5}$ gradients around major roads. In Montreal, substantial spatial variability in BC (absorbance) was observed with the difference between urban background and traffic

[1] $PM_{2.5}$ ranged from 0.92 to 8.91 µg/m^3 and PM light absorbance 0.24-2.36x10^{-5}m^{-1} – note that for Vancouver PM light absorbance can be roughly converted into EC using 10^{-5}m^{-1} = 0.8µg/m^3 EC (Rich, 2002).

influenced sites ranged from 12% to 120% (Smargiassi et al. 2005). Beckerman et al. (2008) also examined gradients in $PM_{2.5}$ and ultra-fine particle mass (UFPM) around an expressways in Toronto and found large spatial variability, with $PM_{2.5}$ and UFPM highly correlated with NO_2 measurements (0.70 and 0.64 respectively). A mobile monitoring unit, the Environment Canada Canadian Regional and Urban Investigation System for Environmental Research (CRUISER), was also piloted to examine gradients and pollutant relationships and found low correlations between NO_2 and $PM_{2.5}$ (r=0.54) but higher correlations for UFPM (r=0.87) and BC (r=0.91) and NO_2.

International $PM_{2.5}$ Studies

There are a number of studies documenting $PM_{2.5}$ intraurban measurements in non-Canadian cities that are useful for determining the different scales of $PM_{2.5}$ spatial variability. Table 1 summarizes the number of monitoring studies demonstrating $PM_{2.5}$ variation above/below 20% for each spatial domain (i.e. local, neighborhood, urban) and the approximate mean and range of $PM_{2.5}$ spatial variation. In each study, monitoring at different locations occurred simultaneously or were standardized to examine spatial differences. The spatial domain refers to the lowest area investigated or reported in each study. Indicators of primary $PM_{2.5}$ (i.e. BC, UFPM/N [ultrafine particle number]) are also included. In general, most studies contain data predominantly at the urban scale, with studies at the local scale examining specific sources and gradient – typically vehicle emissions. At all spatial domains, $PM_{2.5}$ mass exhibits mixed results in terms of spatial variation, while primary $PM_{2.5}$ showed large spatial variability at all spatial domains.

Table 1. Summary of $PM_{2.5}$ spatial variability by geographic scale.

Geographic Scale	Pollutant	Variability	Sources
Local (<1km)	$PM_{2.5}$	>20% (n=1) <20% (n=3) Mean=~19% (Range:5-30%)	(Cyrys et al. 2003, Fischer et al. 2000, Janssen et al. 1997, Roorda-Knape et al. 1998, Ross et al. 2007, Sun et al. 2004, Reponen et al. 2003)
	Primary $PM_{2.5}$	>20% (n=5) <20% (n=0) Mean=~310% (Range:50-1000%)	(Janssen et al. 1997, Roorda-Knape et al. 1998, Sun et al. 2004, Reponen et al. 2003)
Neighbourhood (0-5km)	$PM_{2.5}$	>20% (n=2) <20% (n=3) Mean=~19% (Range:6-60%)	(Allen et al. 2010, Goswami et al. 2002, Noble et al. 2003, Snyder et al. 2010, Lena et al. 2002, Ye et al. 2003)
	Primary $PM_{2.5}$	>20% (n=2) <20% (n=1) Mean=~390% (Range:10-300%)	(Goswami et al. 2002, Noble et al. 2003, Lena et al. 2002, Ye et al. 2003, Gryparis et al. 2007, Gryparis et al. 2007)
City (5-50km)	$PM_{2.5}$	>20% (n=12) <20% (n=15) Mean=~36% (Range:5-200%)	see [a]
	Primary $PM_{2.5}$	>20% (n=13) <20% (n=1) Mean=~218% (Range:27-1300%)	see [b]

[a] Sun et al. 2004, Sun et al. 2004, Goswami et al. 2002, Snyder et al. 2010, Gryparis et al. 2007, Suh et al. 1997, Houthuijs et al. 2001, Biswas et al. 2004, Bari et al. 2003, Brauer et al. 2003, Chow et al. 1994, Fraser et al. 2003, Russell et al. 2004, Yttri et al. 2009, Nerriere et al. 2005, Maciejczyk et al. 2004, Dongarrą et al. 2010, Chen, Mao 1999, Tang et al. 2004, Wade et al. 2006, DeGaetano, Doherty 2004

[b] Snyder et al. 2010, Yttri et al. 2009, Schwab et al. 2004, Hochadel et al. 2006, Venkatachari et al. 2006, Kinney et al. 2000, Butler et al. 2003, Sardar et al. 2005, Krudysz et al. 2008

At the local spatial domain (<1km) there is low evidence for significant $PM_{2.5}$ mass concentration variation but high evidence for variation in components of $PM_{2.5}$ representative of primary $PM_{2.5}$ (BC and UFPM/N). Zhou and Levy (2007) conducted a systematic review of measured and modeled pollutant gradients from roads and found that the spatial extent of BC and $PM_{2.5}$ (excluding background concentration) was 100–400m (note that BC and $PM_{2.5}$ mass were not separated) and UFPN 100–300m. Karner et al. (2010) also synthesized findings from 41 monitoring studies to examine traffic-related pollutant gradients, and found little evidence for

a gradient with $PM_{2.5}$ mass but gradients for UFPN (up to 570m) and BC (elemental carbon) (up to 130m). Gradients determined by Karner et al. (2010) are illustrated in Figure 1.

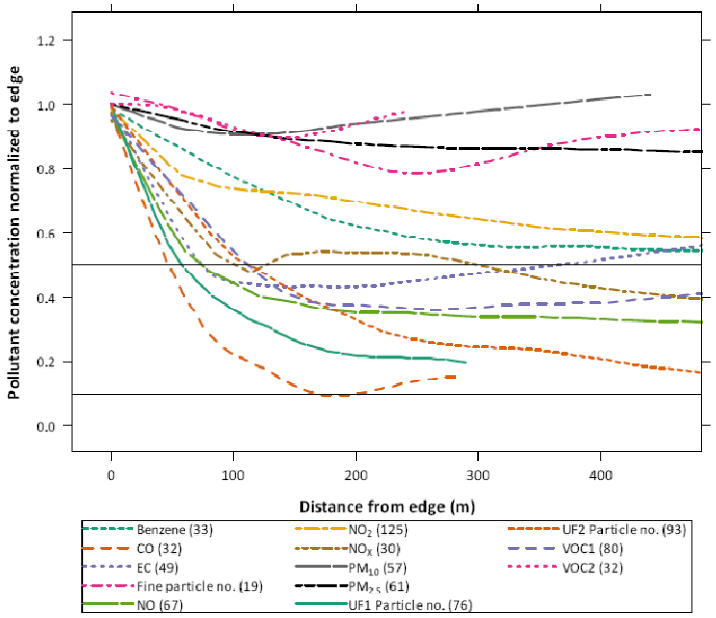

Figure 1. From (Karner et al. 2010): "Local regression of edge normalized concentrations on distance. The horizontal black lines show a reduction from the edge of road concentration of 90% (at 0.1) and 50% (at 0.5). A loess smoother (alpha-0.70, degree=1) was fitted to pollutant data. The regression sample size, n, is given in parentheses after each pollutant. The n includes as estimates (not in the literature) edge-of-road value to facilitate normalization."

At the neighborhood spatial domain (<1-5km) there were relatively few studies documenting $PM_{2.5}$ spatial variability, due primarily to studies not explicitly stating the study spatial domain. Nevertheless, studies at the neighborhood spatial scale agreed with the spatial variability seen in local settings. $PM_{2.5}$ mass showed mixed results while components of primary $PM_{2.5}$ showed high spatial variability. LUR studies of $PM_{2.5}$ mass (e.g. Ross et al. 2007, Hochadel et al. 2006, Henderson et al. 2007, Clougherty et al. 2009, Moore et al. 2007) indicate that geographic variables at this spatial domain are significant predictors of $PM_{2.5}$, with traffic indicators and landuse characteristics predicting $PM_{2.5}$ mass spatial variability. Much lower spatial variability is documented, however, in $PM_{2.5}$ mass compared to BC or NO_2.

Most studies explicitly examining the spatial variability of $PM_{2.5}$ have been conducted at the city-level and show mixed results in terms of $PM_{2.5}$ spatial variability. Twelve studies indicated heterogeneity in $PM_{2.5}$ measurements at the city scale and 15 studies indicated homogeneity (<20% differences in spatial $PM_{2.5}$ measurements). The approximate mean and

range of $PM_{2.5}$ mass variability was 36% (Range:5-200%), indicating large differences in spatial uniformity. This corresponds to the findings of Wilson et al. (2005) who reviewed intraurban $PM_{2.5}$ mass variation using 16 published studies, of which 8 classified $PM_{2.5}$ as heterogeneous at an intraurban scale and 8 as homogeneous, and concluded that the degree of intraurban variation in $PM_{2.5}$ was case-specific. Another review of the spatial variability of $PM_{2.5}$ in 27 urban areas in the United States (Pinto et al. 2004) also found $PM_{2.5}$ mass concentrations to vary widely within urban areas, using the coefficient of divergence (COD) as a measure of spatial variability. Correlation coefficients between monitor pairs examine both spatial and temporal variation, while the COD can be used to specifically examine the spatial variability of concentrations among monitor measurements, with 0 representing identical measurements and 1 representing low similarity (a cut-point of 0.20 is typically used to represent heterogeneity). The COD for paired monitors is calculated as:

$$\mathrm{COD}_j^{i,k} = \sqrt{\frac{1}{p_j^{i,k}} \sum_{t(j \in i,k)}^{p_j^{i,k}} \left(\frac{x_{t,j}^i - x_{t,j}^k}{x_{t,j}^i + x_{t,j}^k} \right)^2}$$

where $x^i_{t,j}$ and $x^k_{t,j}$ are the concentration of constituent j on day t at monitors I and k, respectively, and $p_j^{i,k}$ is the number of days with data for constituent j for both monitors I and k.

Figure 2 illustrates a map of the COD estimated by Pinto et al. (2004) for metropolitan areas in the United States. No specific distances were found in which $PM_{2.5}$ concentration differences stabilized and considerable spatial variability was found in the west (Pinto et al. 2004).

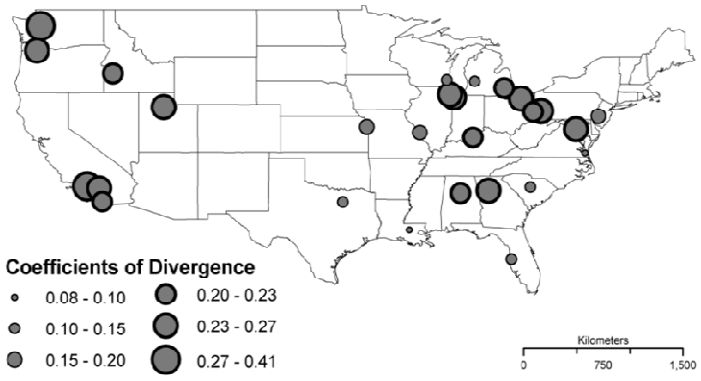

Figure 2. From (Pinto et al. 2004). Maximum COD measures for $PM_{2.5}$ monitors in US metropolitan areas (bold outline indicates metropolitan areas with COD > 0.20).

Particle composition may also affect the spatial representativeness of NAPS $PM_{2.5}$ mass measurements – particularly at the city-level. In a national US study, 7of 52 $PM_{2.5}$ components examined contributed ≥ 1% to the $PM_{2.5}$ total mass for yearly or seasonal averages. These included [ammonium (NH4+), BC (elemental carbon), organic carbon matter (OCM), nitrate (NO_3–), silicon, sodium (Na+), and sulfate (SO42–)] (Bell et al. 2007). In Canada, Evans et al. 2009 found that secondary sulfate and nitrate were the most significant contributors at urban sites in Canada, accounting for ~61% in Windsor, ~60% in Toronto, ~48% in Montreal, ~47% in Halifax, and ~41% in Edmonton of $PM_{2.5}$ mass concentrations. Using source apportionment modeling, the combined contribution of traffic and road dust factors as a motor vehicle-related source ranged from 14% to 21% at the urban sites, indicating a relatively small range of vehicle-related $PM_{2.5}$ mass between monitoring locations. Allen and Turner (2008) found that the nature of the regional contributions to $PM_{2.5}$ levels in urban areas differed from region to region in the United States, with sulfate dominating aerosols in Eastern and Midwestern locations and nitrate being a prominent component in the Central Valley of California. Bell et al. (2010) also examined whether $PM_{2.5}$ chemical constituents varied within US metropolitan areas and found spatial heterogeneity for BC (elemental carbon), organic carbon(OC), ammonium, sulfate, nitrate, silicon and sodium – however, spatial differences were lower for ammonium, sulfate, and nitrate compared to the other measured components as illustrated in Figure 3. Figure 4 also illustrates correlations between monitors matched by distance and chemical constituents.

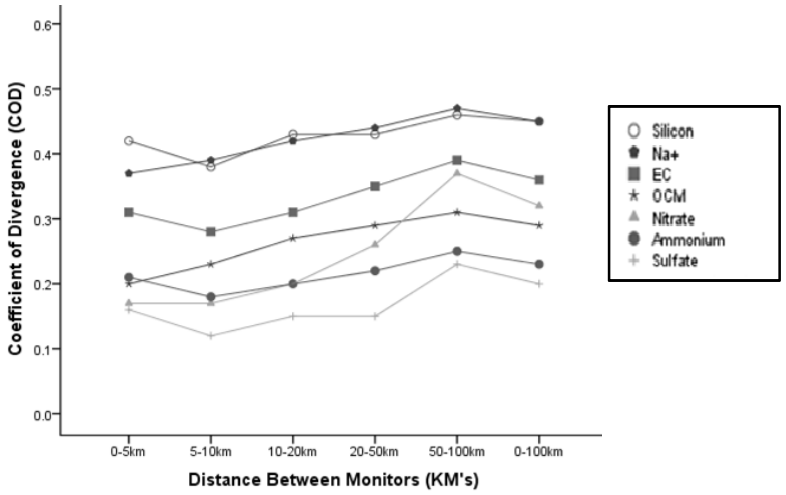

Figure 3. From (Bell et al. 2010): COD measures of $PM_{2.5}$ chemical constituent concentrations for monitor pairs by distance.

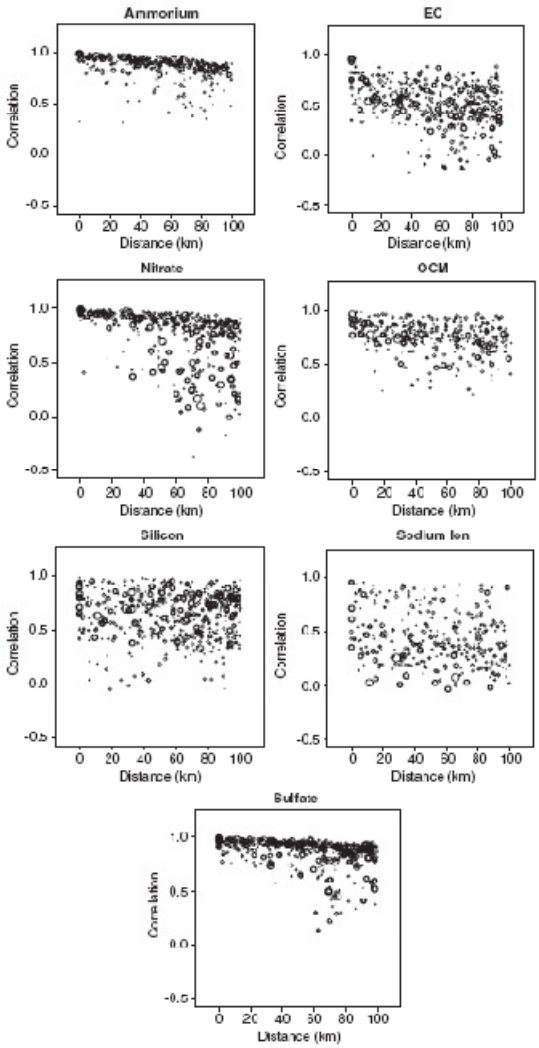

Figure 4. From (Bell et al. 2010): "Correlation of monitor pairs versus distance for $PM_{2.5}$ chemical constituents. Note: Each circle represents a monitor pair. The size of the circle represents the number of observation days in each monitor pair; large circles have larger sample sizes."

Relationship between $PM_{2.5}$ and NO_x Spatial Variation

Low spatial correlations have been found between $PM_{2.5}$ mass and NO_x; however, stronger spatial relationships have been found for primary $PM_{2.5}$. For example, in Toronto, Beckerman et al. (2008) found low spatial correlation between $PM_{2.5}$ and NO_2 (r=0.14), but high correlations between UFPM and NO_2 (r=0.85), NO (r=0.87) and NO_x (r=0.93) across monitoring transects. BC was also strongly correlated with NO_2 (r= 0.89), NO(r=0.50) and NO_x (r=0.64). Similarly, Smargiassi et al. (2005) found the spatial variability in BC (absorbance) was related to traffic intensity on a small scale in Montreal and Fischer et al. (2000) found that BC

(absorbance) could been used to represent exposure to the traffic air pollution mixture. Other studies, however, have found less spatial correlations between NO_x and $PM_{2.5}$. For instance, in Huddersfield England, modelled long-term concentrations of NO_2 provided only weak predictors of spatial variations in $PM_{2.5}$ mass and BC (absorbance) (Kingham et al. 2000).

Conclusions: $PM_{2.5}$ Intraurban Spatial Variability

Drawing conclusions about the degree of spatial variability of $PM_{2.5}$ is complicated by averaging times, spatial density of measurements, characterization of background levels, and statistical criteria employed (Levy and Neal 1999). The studies summarized in Table 1 have typically used relatively long time scales (e.g. monthly and yearly averages) and significant $PM_{2.5}$ spatial variation likely exists at shorter time-scales (Wilson et al. 2005).

Spatial variability of intraurban $PM_{2.5}$ is case-specific, and is dependent on context at all geographic scales, with some circumstances leading to homogenous or heterogeneous spatial concentrations. $PM_{2.5}$ concentrations and the distance between sites in urban areas does not appear to be solely responsible for the spatial variability observed (Pinto et al. 2004) – indicating the importance of local sources and monitor siting. Due to local sources, such as motor vehicles, it has been found that the spatial variation of particulate air pollution is not sufficiently captured by $PM_{2.5}$ mass measurements alone (Hoek et al. 2002) and other measures, particularity BC, are needed to capture primary particles. It has also been shown that secondary particulate composition may introduce different spatial scales in $PM_{2.5}$ mass, for example, sulfate and nitrate exhibit spatial and temporal trends that are primarily regional in nature, while local emissions of primary OC and BC are expected to have a significant impact on the spatial distribution of $PM_{2.5}$ within cities (Bell et al. 2010).

The spatial variability in NAPS measurements should be evaluated using COD's by census metropolitan areas (CMA's) in Canada. This would provide a base-line for the spatial variability seen in NAPS monitors within each urban area. There are 18 CMA's in Canada with more than one O_3 monitor and 23 with more than one $PM_{2.5}$ monitor (see Table 2). 117 and 120 CMA's contain either one $PM_{2.5}$ or O_3 monitor, while 61 $PM_{2.5}$ and 65 O_3 monitors are not located in CMAs. It is important to note that the size of CMA's vary substantially and other metrics, such as distance between monitor pairs, could be used to examine spatial variation.

Table 2. CMA's with >1 $PM_{2.5}$ or O_3 NAPS monitor.

CMA	O_3 NAPS Monitors (n)	$PM_{2.5}$ NAPS Monitors (n)
Abbotsford	2	-
Calgary	4	4
Campbell River	-	2
Edmonton	6	7
Grande Prairie	-	3
Guelph	3	4
Hamilton	3	3
Halifax	3	-
Kitimat	-	2
Lethbridge	-	2
London	2	-
Montreal	15	17
Ottawa/Gatineau	4	4
Powel River	-	2
Prince George	-	2
Quesnel	-	3
Quebec	6	3
Saint John	4	2
St. Catharines	-	2
St. John's	2	-
Toronto	11	11
Trois-Rivieres	-	2
Vancouver	16	8
Victoria	3	4
Windsor	2	2
Winnipeg	2	2
Wood Buffalo	4	5
Total CMA's >1 NAPS Monitors	***18***	***23***

*Based on 2006 Statistics Canada CMA boundaries.
**Number of monitors was determined from the NAPS monitor meta-data and does not incorporate actual monitoring data.

Ozone

O_3 is a secondary atmospheric pollutant produced by photochemical reactions of natural and anthropogenic sources of NO_x and VOCs (García et al. 2010, Gibson et al. 2009, Lee et al. 2004). The main anthropogenic sources of ground level O_3 precursors (NO_x and VOCs) are emissions from motor vehicle exhaust, industrial facilities and electrical utilities (Yuval and Broday 2006, Beaney and Gough 2002). Factors influencing the formation of O_3 at a particular location include topography, local scale photochemistry affected by meteorology and O_3 precursors, regional and long range transport, and stratospheric-tropospheric exchange (Gibson et al. 2009, Krupaa et al. 2003).

Although the literature contains extensive studies examining ambient O_3 concentrations, those examining spatial variability have mainly focused on large spatial scales at coarse resolutions (Yuval and Broday 2006) and few studies specifically investigate local or within-city spatial variations of O_3 (Liu, Rossini 1996). We conducted a review of Canadian and international studies that monitored O_3 concentrations within-cities to determine how O_3 concentrations vary at different spatial scales, including local (<1km), neighborhood (<1-5km), urban (<1-50km), urban/suburban and urban/rural.

Canadian O_3 Studies

The majority of Canadian studies examining ground level O_3 use data from NAPS monitors. For example, Burnett et al. (1994) examine hourly concentrations from 22 air quality network monitors in Ontario over a five-year period (Jan. 1, 1983 – Dec. 31, 1988). Monitors located <50km from each other had a correlation coefficient of $r>0.71$, while monitors located <100km apart had a slightly lower correlation $r<0.68$. Ground level O_3 measurements from 9 network stations in Montreal (1984-1990) were analyzed by McKendry (1993) and concentrations above background were observed 40-70km downwind of Montreal, with intraurban variation of ~36% between monitors. This variation is slightly larger than the ~24% variation observed between 2 network sites in Winnipeg, Manitoba during 1998-1999 (Raddatz, Cummine 2001).

A general lack of Canadian studies with O_3 measurements independent from NAPS data was identified, with only three additional studies available in Canada documenting intraurban O_3 spatial variation. Liu, Rossini (1996) used passive O_3 samplers at 40 sites in Toronto during the summer of 1992 and found that daytime concentrations were more spatially variable, with spatial autocorrelation between O_3 measurements persisting to 15km's during the day, and 45km's at night. Gibson et al. (2009) examined the spatiotemporal variability of O_3 in the rural

Annapolis Valley of Nova Scotia from August 20, 2006 to Sept. 28, 2007 using passive samplers at 17 locations. They found significant spatial variation in O_3 levels due to topographic and meteorological influences. Beckerman et al. (2008) used passive samplers to examine the correlation of NO_2 with other traffic pollutants near a major expressway in Toronto. Passive samplers were deployed for 1-week in August 2004 at transects away from 2 expressways to examine spatial gradients in concentrations with distance from traffic sources (roadside to ~1000m). O_3 displayed large spatial variability due to scavenging by NO (Figure 5 illustrates gradients for O_3 and other pollutants up-wind and down-wind of the expressways). At both sites, on either side of the expressways, concentrations of O_3 first increase with distance from roadway, to a peak (between 200-400 m) after which they dropped off. At the first site, concentrations of O_3 varied between approximately 24% and 50% within 200m downwind and upwind of the expressway. At the other site, the upwind side varies by 25% within 200m, while on the downwind side concentrations vary between ~15%.

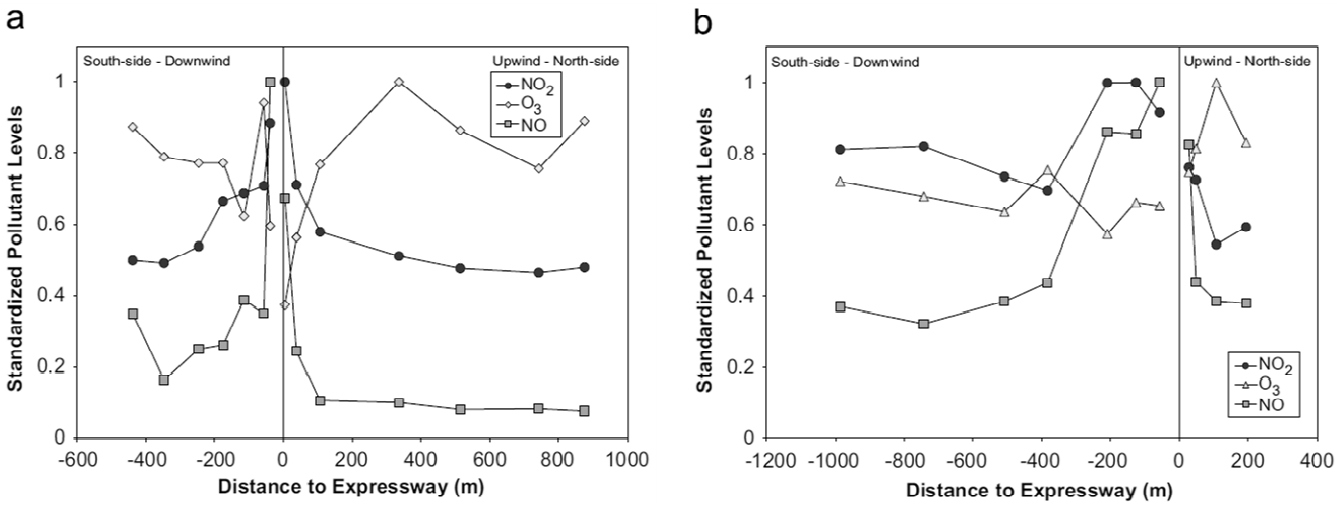

Figure 5. O_3, NO_2, and NO concentrations upwind and downwind of two expressways in Toronto (Beckerman et al. 2008).

International O_3 Studies

A number of international studies have measured the spatial variability of O_3 within cities (Table 3 summarizes the spatial variability of O_3 from identified studies). Spatial variability of O_3 was demonstrated at all spatial scales (local, neighborhood, city, urban/suburban and urban/rural). This agrees with a review of 50 O_3 mapping studies, that concluded that although the spatial scale of ground level O_3 is unknown, in metropolitan areas it is likely less than 10km, and possibly as small as 5km. At the local-scale the reactions between NO in fresh vehicle

exhaust and O_3 results in large spatial variation near major roads and heavy traffic areas. In a previous study, concentrations 8m downwind from a freeway were often less than 10% of background ambient levels, and reductions in O_3 concentrations were evident up to 500m from the freeway (Rodes, Holland 1981).

Table 3. Summary of O_3 spatial variability by geographic scale.

Geographic Scale	Variability	Sources
Local (0.1-1km)	27%; 500 m average spatial dependence	(García et al. 2010, Lin et al. 2001)
Neighbourhood (0-5km)	>20% (n=9) <20% (n=1) Mean=~39% (Range:4-64%)	(Lin et al. 2001, Geyh et al. 2000, Liu et al. 1993, Adame et al. 2008, McConnell et al. 2006b, BANJA et al. , Steurer et al. 2006, McNair et al. 1996, Bhugwant, Brémaud 2001, Laurinavičienė 2009)
City (5-50km)	>20% (n=11) <20% (n=4) Mean=~29 (Range:4-58%)	(García et al. 2010, Liu, Rossini 1996, Raddatz, Cummine 2001, Lin et al. 2001, Adame et al. 2008, McConnell et al. 2006b, BANJA et al. , Steurer et al. 2006, Laurinavičienė 2009, Diem, Comrie 2002, Xian, Crane 2006, Silva Júnior et al. 2009, NYC 2010)
Urban/Suburban	>20% (n=3) <20% (n=4) Mean=~21% (Range:3-40%)	(Liu, Rossini 1996, McKendry 1993, Steurer et al. 2006, Diem, Comrie 2002, Chen et al. 2004, So, Wang 2003)
Urban/Rural	>20% (n=3) <20% (n=1) Mean= ~40% (Range:13-65%)	(Bhugwant, Brémaud 2001, Diem, Comrie 2002, Chen et al. 2004, So, Wang 2003)

Relationship between O_3 and NO_x Spatial Variation

Table 4 summarizes observed correlations between measured concentrations of O_3 and NO_x, NO and NO_2 for short-term (daily, weekly), medium-term (monthly, seasonally) and long-term (annually) averaging durations. Negative correlations are consistently observed between O_3 and nitrogen oxides, with correlations ranging from -0.28 to -0.87 for NO_x, -0.36 to -0.78 for NO, and -0.27 to -0.96 for NO_2. Direct interpretation and comparison between studies is difficult, as studies vary by the number of sites and time periods examined.

Table 4. Correlations observed between O_3 and NO_x, NO and NO_2.

Relationship	NO_x	NO	NO_2
Short-Term (Daily/Weekly)	-0.51 (1)		-0.96 (2) -0.351 (3)
Medium-Term (Monthly/Seasonal)	-0.87 Winter (4) -0.75 Summer (4)	-0.748 Summer Daytime (5) -0.561 Summer Nighttime (5)	-0.54 (6) Monthly (6) -0.566 Summer Daytime (5) -0.774 Summer Nighttime (5) -0.516 (Jan); -0.634 Apr); -0.625 (Jul); -0.585 (Oct) (7) -0.74 (2)
Long-Term (Annual)	-0.28 to -0.39 (8) -0.39 (9) -0.83 Daytime (9)	-0.55 to -0.57 (8) -0.78 (10) -0.58 (11)	-0.22 to- 0.23 (8) -0.86 (4) -0.27 to -0.43 (11) -0.74 (6) -0.62 (12)

(1) (McConnell et al. 2006a); (2) (Bernard et al. 1999); (3) (Lee et al. 2004); (4) (Khoder 2009); (5) (Abdul-Wahab et al. 2005)); (6) (Tidblad et al. 2004b); (7) (Jiménez-Hornero et al. 2010); (8) (Wang et al. 2003); (9) (Pudasainee et al. 2006); (10) (Yuval, Broday 2006); (11) (Sousa et al. 2006); (12) (Frischer et al. 1999).

Conclusions: O_3 Intraurban Spatial Variability

Characteristics of metropolitan areas, such as population, traffic, industrial emission densities, topography and meteorology can affect the spatial distribution of O_3 (Liu, Rossini 1996). Since NO produced by combustion emissions quenches O_3 to form NO_2, O_3 concentrations are often lower in urban centers than in surrounding suburban and rural areas (Bates 2005). This phenomenon creates an inverse pattern between NO_x and O_3 in urban regions, with higher concentrations of NO_2 associated with lower concentrations of O_3 (Tidblad et al. 2004).

Air quality monitoring networks, while generally adequate at capturing or estimating maximum O_3 concentrations and regional patterns, are too sparse and have inadequate spatial resolution to capture local or neighborhood variation within urban areas (Liu, Rossini 1996). A recent critique of O_3 mapping efforts noted that interpolations from central site monitors are not justified by the spatial resolution of available data and have not been validated against a dense network of measurements that would be required to justify the assumption that the distribution of O_3 is homogeneous within communities (Diem 2003).

Based on the literature review, spatial variation is evident at all geographic scales (as seen in Table 3), however, the magnitude of variation was relatively small (<65%) compared to that seen for primary $PM_{2.5}$. The spatial variability in O_3 concentrations in 24 US metropolitan areas was also examined using correlation and COD measures between paired monitor sites (EPA, 2006). Mean O_3 concentrations were found to vary within individual urban areas by

factors of 1.4 to 4. This variability in O_3 concentrations is larger than that found for $PM_{2.5}$ concentrations using COD and comparable to that obtained for coarse PM (EPA, 2006). These results indicate that caution should be used when interpreting data from fixed-site O_3 monitors and that representative areas need to be considered when assigning values to approximate community-scale exposures.

Evaluating O_3 Intraurban Variability using Existing NO_2 Models

We explored the ability of existing city-specific NO_2 LUR models to examine the spatial representativeness of NAPS O_3 and $PM_{2.5}$ monitors. An inverse relationship is commonly observed between concentrations of O_3 and NO_x due to emissions of NO in urban areas reacting with O_3 to produce NO_2 (Tidblad et al. 2004). The nature of this relationship may potentially allow concentrations of O_3 to be modeled within cities where information is known about the spatial variability of NO_2. Similarly, vehicle emissions are a large source of both primary $PM_{2.5}$ and NO_2 and existing LUR NO_2 surfaces may be informative for determining the spatial variation in primary $PM_{2.5}$. Here, however, we only examine the feasibility of using LUR surfaces from Canadian cities to assess the spatial representativeness O_3 measurements. A review of the literature was conducted to identify information on the relationship between O_3 and NO_2 (shown previously in Table 4) and existing studies that have used NO_2 to model intraurban O_3 variability.

Studies Modeling Intraurban O_3 Spatial Variation using NO_x

A small number of studies have modeled intraurban O_3 concentrations using measured or modeled NO_x concentrations. Rodes and Holland (1981) developed a model (O_3=0.069-0.063*0.994*D + ε, where d=distance) from empirical data to estimate O_3 from NO at downwind distances from highways (the distance decay of O_3 and NO_x are shown in Figure 6). They concluded that during days with low (0-0.057ppm), and medium (0.058-0.084ppm) O_3 concentrations days, scavenging of O_3 occurs up to 388m from the roadway.

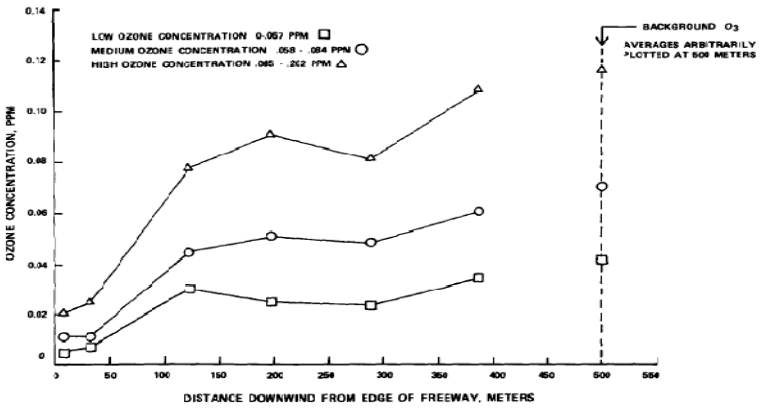

Fig. 2. Average ozone (O_3) concentrations.

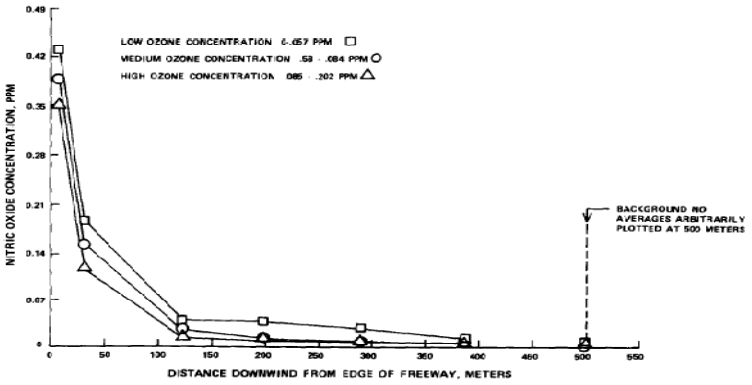

Fig. 3. Average nitric oxide (NO) concentrations.

Figure 6. Average O_3 and NO concentrations downwind of a highway (Rodes, Holland 1981).

McConnell et al. (2006) examined O_3 and NO_x interaction and found that O_3 declined in a predictable manner as modeled residential NO_x increased. Some non-linearity in the effect of NO_x was seen that that may have been due to much smaller residential NO_x estimates in one community (mean of 29ppb). Their regression model (shown below) indicates that residential NO_X reduces residential O_3 concentrations by 0.51ppb per 1 ppb of NO_X, and that a 10th-90th percentile increase in local NO_X results in a 7.5ppb decrease in local O_3 concentrations.

Home O_3 = 37.7 +0.91*(Central Site O_3) – 0.029*(Central Site NO_x) – 0.51* (Estimated Residential NO_x)

Tidblad et al. (2004) examined the relationship between NO_2 and O_3 in urban areas of Europe and highlighted the importance of sunshine duration on the relationship between O_3 and NO_x [O_3=(35+0.016*Sun)*exp((-0.022+5.0e-6*Sun)*(NO_x)]. Bernard et al. (1999) and Narasimhan et al. (2010) also found distance from NO_2 sources and sunlight to be an important determinant in the relationship between NO_2 and O_3.

Syri et al. (2001) examined the impact of urban NO_x concentrations on O_3 levels using a titration model and found that O_3 concentrations from single measurement points could be extrapolated with NO_x concentrations to improve spatial estimates of O_3 using:

$$[O_{3,U}] = K \times \frac{[O_{x,U}] - [O_{3,U}]}{[NO_{x,U}] - [O_{x,U}] + [O_{3,U}]}$$

where $O_{x,u}$ is the urban oxidizing components (O_3 and NO_x), and $O_{3,u}$ is a representative urban O_3 measurement, and $NO_{x,u}$ a representative urban NO_x measurement.

A number of studies have used LUR approaches to model O_3 and NO_x. Diem and Comrie (2002) used landuse characteristics in various multiple linear regression equations to improve O_3 mapping and Hooyberghs et al. (2006) demonstrated that interpolation methods can be augmented with geographic data, in this case population density, to account for O_3 titration. Abdul-Wahab et al. (2005) used spatiotemporal models to estimate O_3 from a mobile monitor using a number of predictor variables and co-pollutants and achieved R^2 values of 0.69 and 0.68 for daytime and nighttime O_3 prediction. Below are example predictive equations of O_3 (T is temperature, SR is solar radiation).

$$\text{Log } [O_3] = 1.628 - 0.00894*[NO] + 0.04316*T + 0.661*SR - 0.003952*[SO_2]$$

$$\text{Log } [O_3] = 5.26 - 0.0788*[NO_2] + 8.251x10^{-6}*[NO_2]^3 - 0.00969*[NO] + 1.338x10^{-5}*[NO_2]^2$$

An O_3 summer model was produced for New York City (NYC CAS , 2009) that incorporated predicted NO_2 concentrations using the equation [O_3 = 13.41 +0.79*(Fixed Site O_3) - 0.41*(NO_2)]. Figure 7 illustrates the inverse spatial relationship between predicted NO_2 and O_3.

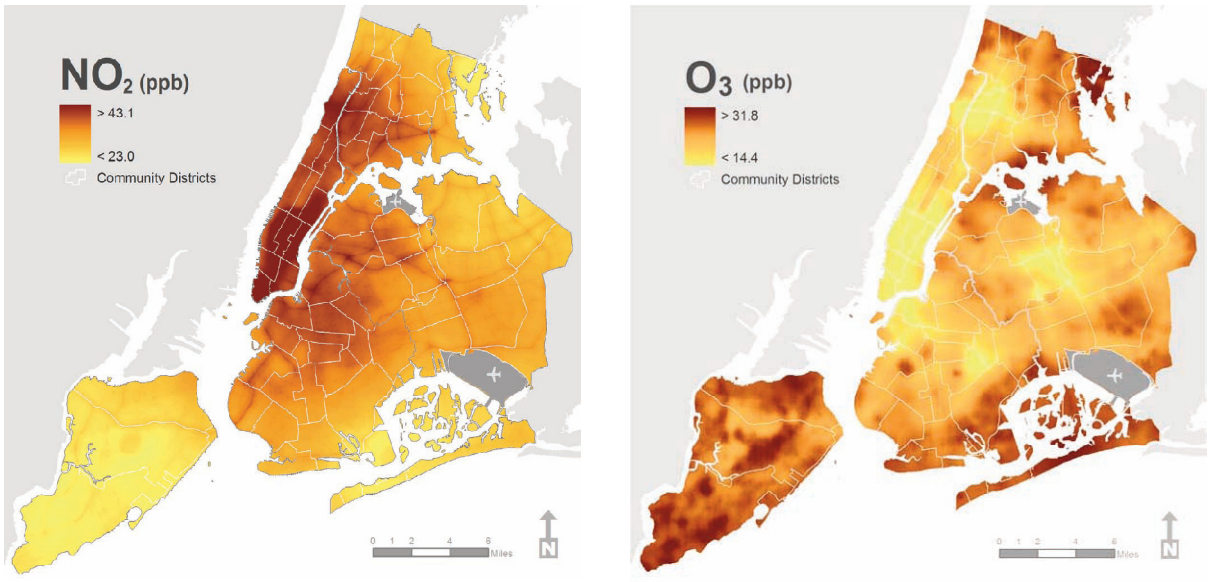

Figure 7. Predicted NO_2 and O_3 for New York City (NYC CAS, 2009).

Existing city-specific NO_2 LUR surfaces may be used to examine the spatial representativeness of NAPS O_3 monitors due to the relationships between NO_x and O_3 described above. To date there have been few attempts to use the relationship to predict O_3

exposure at fine scales, for example at residential locations. These types of predictions would be useful for health studies and regulatory purposes (McConnell et al. 2006). Here we explore the feasibility of using Canadian LUR NO_2 concentration surfaces to predict O_3 spatial variability.

Modeling using Existing Canadian Intraurban NO_2 Data

The majority of existing studies measuring or modeling concentrations of NO_2 in Canada use LUR methodology to model spatial distributions across urban areas. Results from these studies provide more detailed estimates of within-city variability of NO_2 than is typically captured by regulatory monitoring networks. Table 5 summarizes published Canadian LUR models for Victoria, Vancouver, Edmonton, Winnipeg, Toronto, Windsor, Sarnia, Hamilton and Montreal. Other city-specific LUR models are currently being conducted for Health Canada in Calgary, Ottawa, London and Halifax. Appendix 1 illustrates the resulting spatial NO_2 surface for each city-specific LUR model and the locations of NAPS O_3 and $PM_{2.5}$ monitors.

Table 5. Summary of existing intraurban NO_x, NO_2, and NO monitoring data for Canadian Cities.

Investigator, Year	Study Location	Monitoring Date(s)	Measurement Type(s)	Measurement Duration(s)	Sites (n)	Domain (km^2)	Mean (SD) (ppb)	Range (ppb)	IQR (ppb)	R^2
Henderson et al. (2007)	Vancouver, BC	Feb 24 – Mar 15 Sept 8 – 26, 2003	Ogawa Passive Diffusion Samplers	2 weeks	116	2200	16.2 (5.6)	4.8 – 28.0		0.56
Poplawski et al. (2009)	Victoria, BC	Jun 22/23 – Jul 6/7, 2006	Ogawa Passive Diffusion Samplers	2 weeks	40		4.9 (2.6)	0.4 – 10.3		0.61
Allen et al. (2011)	Edmonton, AB	Jan 27 – Feb 1 Apr 27 – May 11, 2008	Ogawa Passive Diffusion Samplers	2 weeks	50		15.4 (3.1)	7.3 – 26.0		0.81
Allen et al. (2011)	Winnipeg, MB	Oct 28 – Nov 11, 2007 Mar 16 – 30, 2008 June 15 – 29, 2008	Ogawa Passive Diffusion Samplers	2 weeks	50		8.5 (2.9)	1.7 – 12.4		0.84
Crouse et al. (2009)	Montreal, QC	Nov/Dec 2005 Apr/May 2006 August 2006	Ogawa Passive Diffusion Samplers	2 weeks	133	600	N-D 12.6 (2.6); A-M 14.0 (4.3); A 8.9 (3.1)	2.6 – 31.5		0.80
Gilbert et al. (2005)	Montreal, QC	May 2003	Ogawa Passive Diffusion Samplers	2 weeks	67		11.6 (3.0)	4.9 – 21.2	3.6	0.52
Su et al. (2009)	Toronto, ON	Spring 2004	Ogawa Passive Diffusion Samplers	2 weeks	100		10.15 (3.12)	4.92 – 19.31	3.53	0.79
Jerrett et al. (2007)	Toronto, ON	Sept 9 – 25, 2002	Ogawa Passive Diffusion Samplers	2 weeks	95	633	32.2 (9.2)	17.6 – 61.1		0.69
Wheeler et al. (2008)	Windsor, ON	Feb, May, Aug and Oct, 2004	Passive 3M #3500 Badges	2 weeks	54	235?	12.4 (2.9)	6.9 – 20.2		0.77
Sahsuvaroglu et al. (2006)	Hamilton, ON	October 2002	Ogawa Passive Diffusion Samplers	2 weeks	107	1372	14.6 (3.7)	8.0 – 28.1		0.76
Atari et al. (2008)	Sarnia, ON	October 2005	Ogawa Passive Diffusion Samplers	2 weeks	37		10.7 (3.0)	5.7 – 16.7		0.79

The literature demonstrates a large inverse relation between NO_2 and O_3; however, the magnitude varied, for example from -0.54 to -0.77 for month and seasonal averages and -0.22 to -0.74 for annual averages. In addition, while a few models were indentified that have incorporated spatial patterns of NO_2 to predict O_3 they include other model parameters and local intercepts that cannot be directly applied to Canadian cities. We therefore developed empirical models using NAPS O_3 measurements.

The relationship between NO_2 and O_3 in Canada was first explored using collocated NAPS NO_2 and O_3 monitors. In 2008 there were 132 NAPS stations with both O_3 and NO_2 monitors. The correlation between average yearly O_3 and NO_2 measurements was -0.60, monthly averages -0.44, and daily averages -0.44. For summer data only (May-September) monthly correlations were -0.29 and daily -0.22. Figure 8 illustrates monthly correlations between collocated NAPS O_3 and NO_2 measurement for 2006, 2007, and 2008 and indicates large correlation differences by month. This suggests that NO_2 may be a poor predictor of O_3 in the summer, particularity July, or that more information in needed in the summer to model O_3 from NO_2.

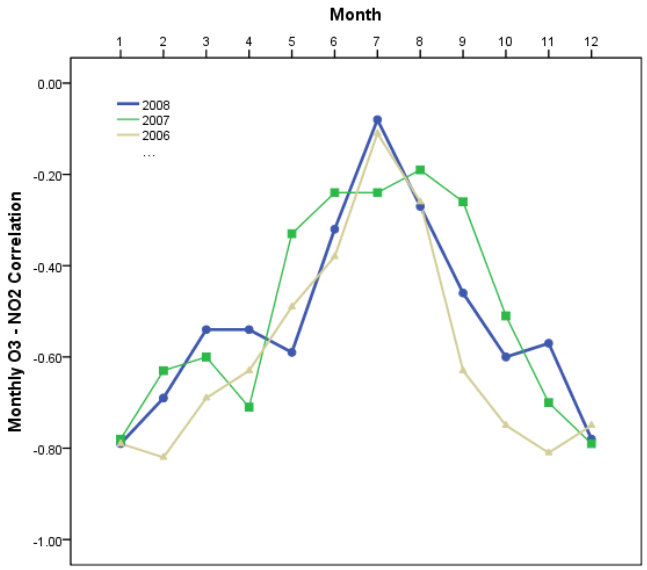

Figure 8. Correlation between average monthly O_3 and NO_2 for 2006, 2007, and 2008.

The low correlations between NO_2 and O_3 in July may be due to the relationship between these two pollutants breaking down during high O_3 concentrations. Figure 9 and 10 illustrate the relationships between NO_2 and O_3 in January and July, with July indicating a non-linear relationship where higher O_3 concentrations may be related to an increase in NO_2 concentrations.

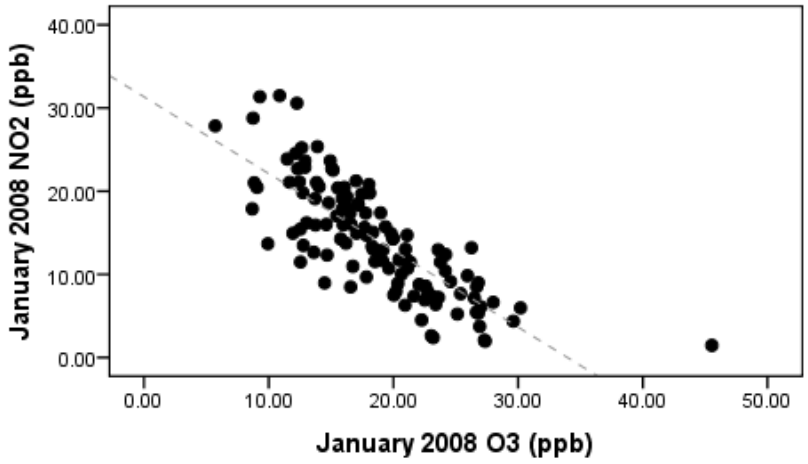

Figure 9. Scatter plot between collocated NO_2 and O_3 (ppb) NAPS measurements in January 2008.

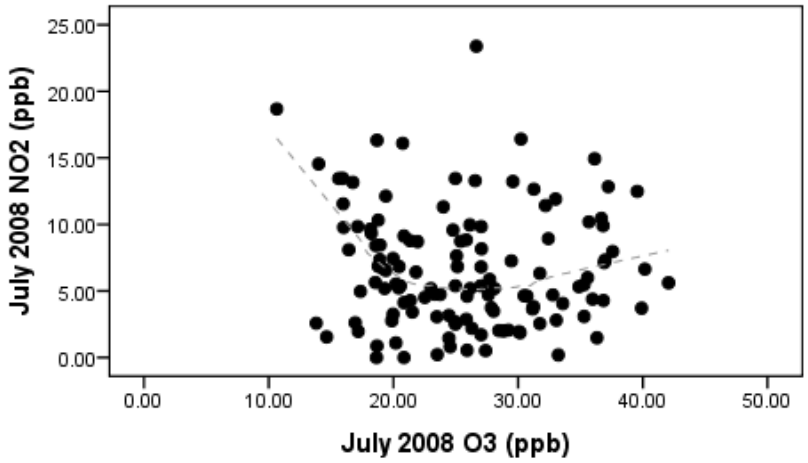

Figure 10. Scatter plot between collocated NO_2 and O_3 (ppb) NAPS measurements in July 2008.

Due to NAPS monitoring siting guidelines (which restrict monitors from being placed close to major roads) the influence of NO_2 on O_3 scavenging may not be seen in the NAPS data. We restricted the correlation analyses to collocated NO_2 and O_3 monitors that were within 100m and 400m of a expressway or highway (using the derived monitor siting characteristics provided in the accompanying Excel table). A maximum distance of 400m was selected as Rodes and Holland (1981) found that during low and medium O_3 days, scavenging occurred up to 388m from the roadway. Figure 11 illustrates the resulting monthly correlations. It can be

seen that for monitors located within 100m of an expressway or highway the correlations between NO_2 and O_3 are larger than that for all monitors. In July the correlation is -0.50 compared to -0.08 for all monitors. On average the correlation between NO_2 and O_3 from collocated NAPS stations within 100m of an expressway or highway was -0.71 compared to -0.52 for all monitors. This suggests that models developed from all NAPS O_3 monitors may not fully capture, or may underestimate, the influence of NO_x scavenging on O_3 next to major roads.

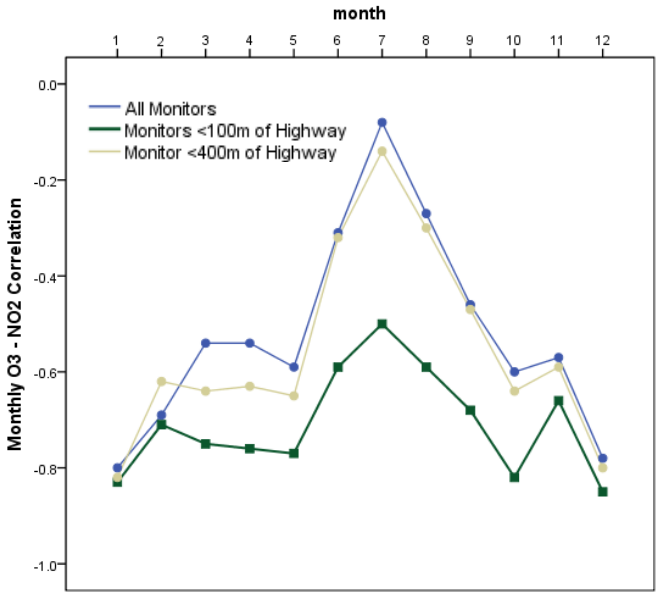

Figure 11. Correlation between average monthly O_3 and NO_2 for 2008 for all NAPS monitors and monitors location <100m and <400m from an expressway or highway.

Ideally, models designed to predict within-city O_3 spatial variation would incorporate both NAPS O_3 measurements (to account for regional differences and high O_3 episodes) as well as NO_2 LUR models to predict local-scale O_3 spatial variation - similar to the approach taken by McConnell et al. (2006). For example, predictions of summer average O_3 concentrations could be based on the following equation:

$$O_3^{i,j} = IDW(NAPS\ O_3) - 0.592(LUR\ NO_2)$$

where O_3 is the predicted concentration at location i,j; IDW(NAPS O3) is the regional predicted O_3 concentration at location i,j using Inverse Distance Weighting (IDW) of NAPS O_3 measurements; and LUR NO_2 is the predicted NO_2 concentration from the LUR model. Unfortunately, none-NAPS O_3 data were not available to build and test such models.

The utility of using NO_2 spatial patterns to estimate O_3 spatial patterns was examined by regressing the city-specific NO_2 LUR model estimates on NAPS O_3 data. A limited number of NAPS O_3 monitors are located in cities within LUR models and there were only 39 O_3 monitoring stations with sufficient monitoring data during the coverage year of each city-specific LUR model. We assumed sufficient NAPS monitoring coverage when there was >15 days of valid measurements per month and ≥10 months per year. Linear regression models were created with the LUR NO_2 yearly estimates assigned to each NAPS monitoring site and the average O_3 concentration for the corresponding LUR modeling year (see Table 5) and time-period. First, annual models were developed. Model 1, including only NO_2 estimates from city LUR models, explained 44% of the annual O_3 spatial variation. Incorporating a dummy variable for city increased the R^2 to 0.84, suggesting a large component of O_3 variation seen in NAPS measurements occurs between cities. Table 6 summarizes the two models.

Table 6. Models predicting NAPS annual average O_3 with LUR NO_2 estimates.

	Parameter Estimate	Standard Error	P
Model 1 R^2=0.44, RMSE=3.00			
Intercept	27.835	1.716	<0.001
NO2_LUR	-0.677	0.122	<0.001
Model 2 R^2=0.84, RMSE=1.58			
Intercept	37.177	1.944	<0.001
NO2_LUR	-0.639	0.074	<0.001
Toronto	-8.305	1.742	<0.001
Montreal	-10.485	1.683	<0.001
GVRD	-11.896	1.633	<0.001
Edmonton	-5.734	1.830	0.004
Winnipeg	-8.529	1.977	<0.001
Victoria	-6.537	1.954	0.003
Sarnia	ref	-	-

Figure 12 illustrates the scatter plot of annual NAPS O_3 measurements and LUR NO_2 measurements. One outlier (NO_2=29 ppb) was left in the model as this monitor is located near traffic sources and likely is influenced by O_3 scavenging. The model parameters in Table 6 did not change significantly when this point was removed; however the R^2 was reduced (Model1 R^2=0.33; Model2 R^2=0.82).

Model 2 was applied to the city-specific NO_2 LUR surfaces to estimate the spatial distribution of annual O_3. Appendix 2 illustrates the resulting O_3 spatial surfaces and the location and value of the corresponding NAPS annual O_3 concentrations.

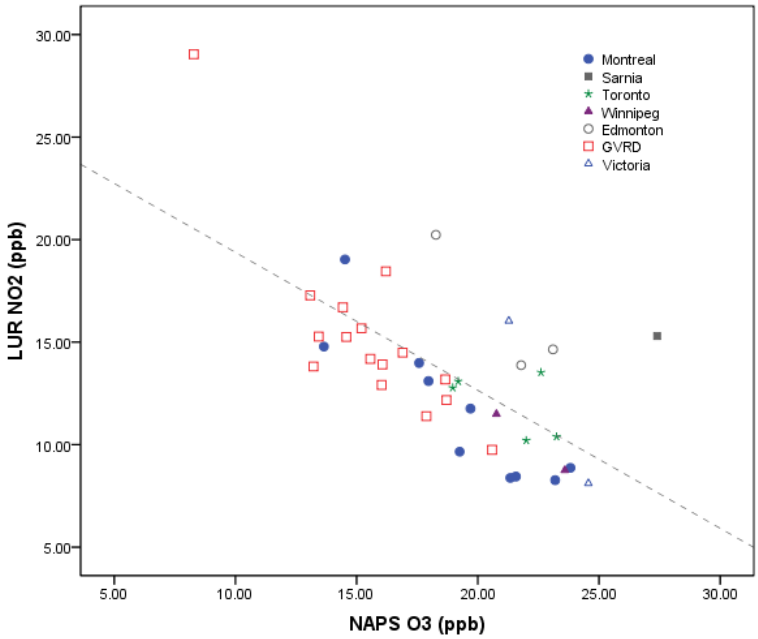

Figure 12. Scatter plot of LUR NO_2 (ppb) and corresponding annual NAPS O_3 (ppb) by city.

Models of O_3 spatial variability during summer months (when concentrations are highest) are of more interest than annual models. The NO_2 LUR models represent annual averages; however, the spatial patterns should be consistent in the summer as model predictors do not change throughout the year (i.e. the NO_2 concentration will shift up or down but the spatial pattern will not change). We therefore explored the application of the LUR models to predict summer (May-September) O_3 patterns as well as summer daylight (7am-9pm) O_3 spatial patterns. Table 7 illustrates the results of these four models and Figure 13 shows the scatter plots between LUR NO_2 estimates and the corresponding summer and summer daylight NAPS O_3 measurements. Models incorporating only LUR NO_2 (Model 1) predicted slightly less of the O_3 variability (R^2=0.25 and 0.24) than the annual model. Both models (summer and summer daylight) incorporating a dummy variable (Model 2) had R^2 values similar to the annual average model, again suggesting that the largest portion of O_3 variation is occurring between cities. Importantly, the NO_2 coefficients in all models were significant and relatively stable (annual LUR NO_2 coefficient=-0.639, summer=-0.592, summer daylight=-0.753), suggesting the association between NO_2 and O_3 can be applied in different seasons to account for O_3 scavenging by NO_x. Figures 14 through 20 illustrate the resulting summer daylight O_3 surfaces for each city.

Table 7. Models predicting NAPS summer, and summer daytime average O_3 with annual LUR NO_2 estimates.

	Parameter Estimate	Standard Error	P
Summer			
Model 1 R^2=0.25, RMSE=3.88			
Intercept	29.566	2.221	<0.001
NO2_LUR	-0.576	0.157	<0.001
Model 2 R^2=0.84, RMSE=1.80			
Intercept	43.391	2.209	<0.001
NO2_LUR	-0.592	0.084	<0.001
Toronto	-9.534	1.991	<0.001
Montreal	-14.854	1.912	<0.001
GVRD	-15.825	1.855	<0.001
Edmonton	-8.030	2.080	0.001
Winnipeg	-14.478	2.246	<0.001
Victoria	-14.141	2.221	<0.001
Sarnia	ref	-	-
Summer Daylight			
Model 1 R^2=0.24, RMSE=4.21			
Intercept	34.28	2.406	<0.001
NO2_LUR	-0.616	0.171	<0.001
Model 2 R^2=0.79, RMSE=2.23			
Intercept	51.057	2.735	<0.001
NO2_LUR	-0.753	0.104	<0.001
Toronto	-10.749	2.465	<0.001
Montreal	-17.601	2.368	<0.001
GVRD	-16.013	2.297	<0.001
Edmonton	-9.332	2.575	<0.001
Winnipeg	-17.466	2.781	<0.001
Victoria	-16.485	2.749	<0.001
Sarnia	ref	-	-

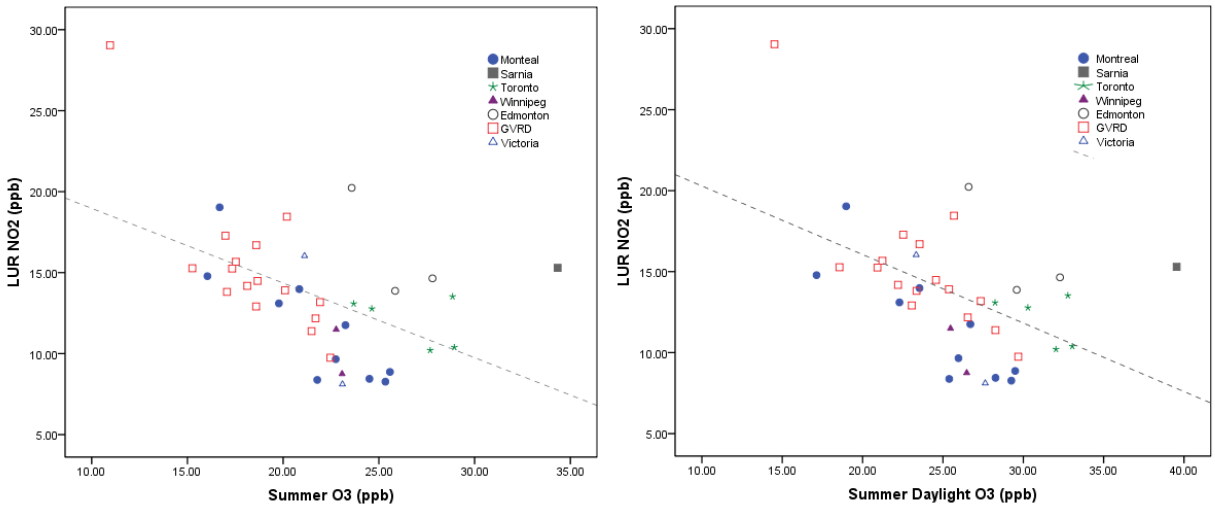

Figure 13. Scatter plot of LUR NO_2 (ppb) and corresponding annual NAPS O_3 (ppb) by city.

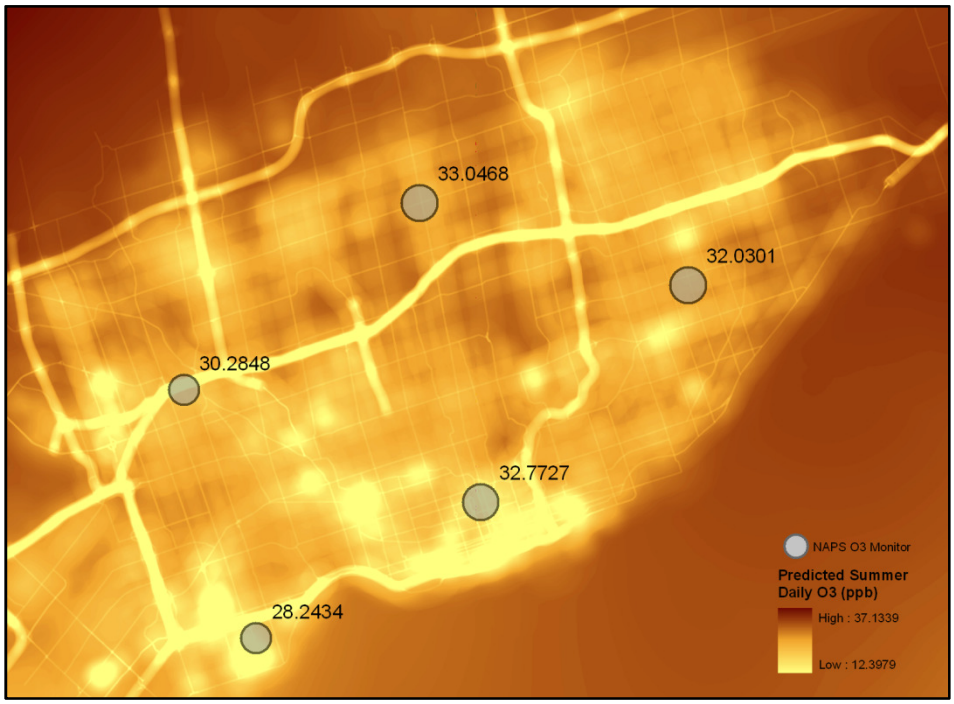

Figure 14. Predicted 2006 summer daylight spatial concentrations of O_3 in Toronto.

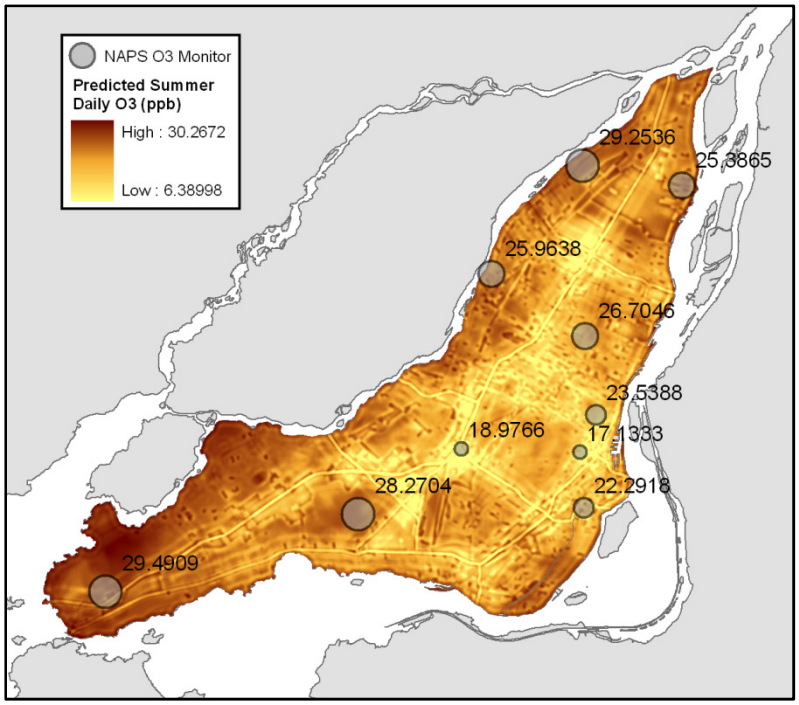

Figure 15. Predicted 2006 summer daylight spatial concentrations of O_3 in Montreal.

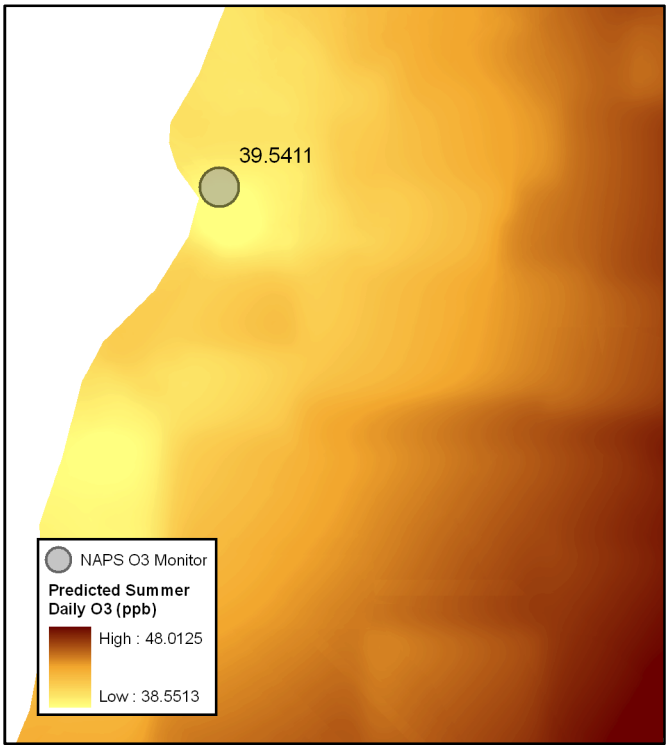

Figure 16. Predicted 2005 summer daylight spatial concentrations of O_3 in Sarnia.

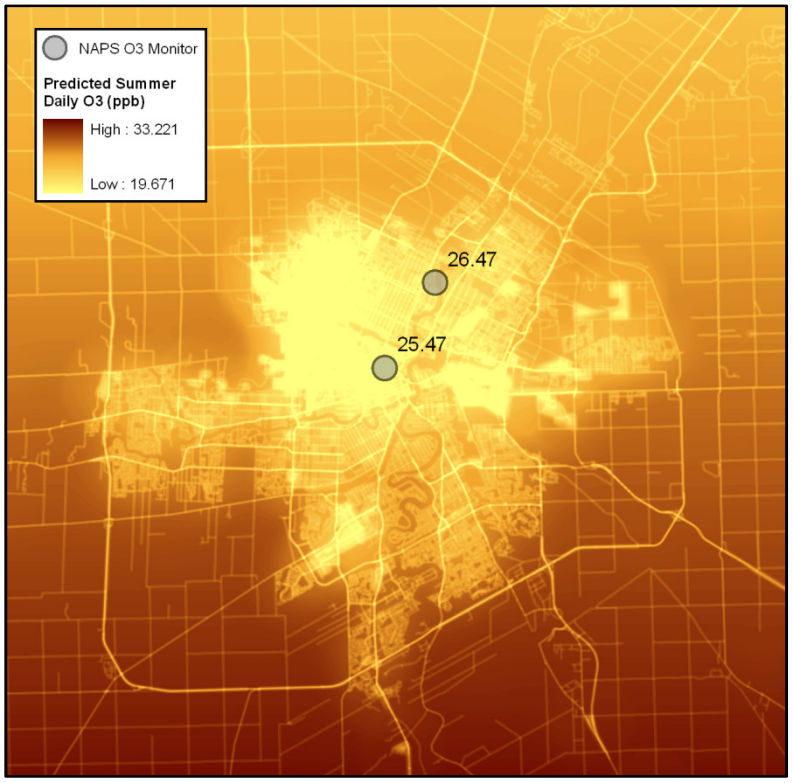

Figure 17. Predicted 2008 summer daylight spatial concentrations of O_3 in Winnipeg.

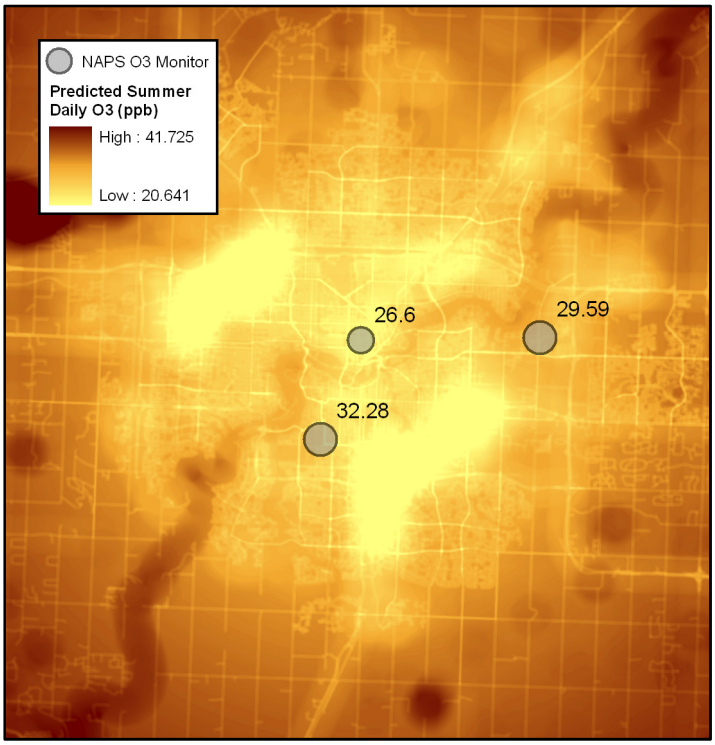

Figure 18. Predicted 2008 summer daylight spatial concentrations of O_3 in Edmonton.

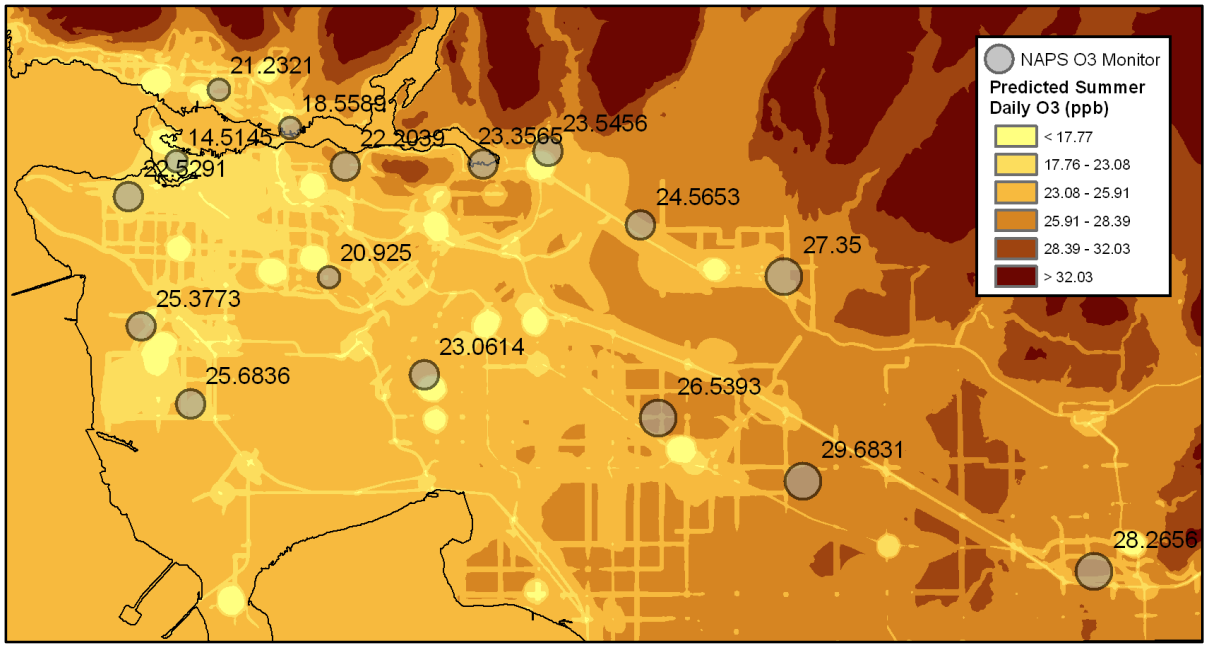

Figure 19. Predicted 2003 summer daylight spatial concentrations of O_3 in Vancouver

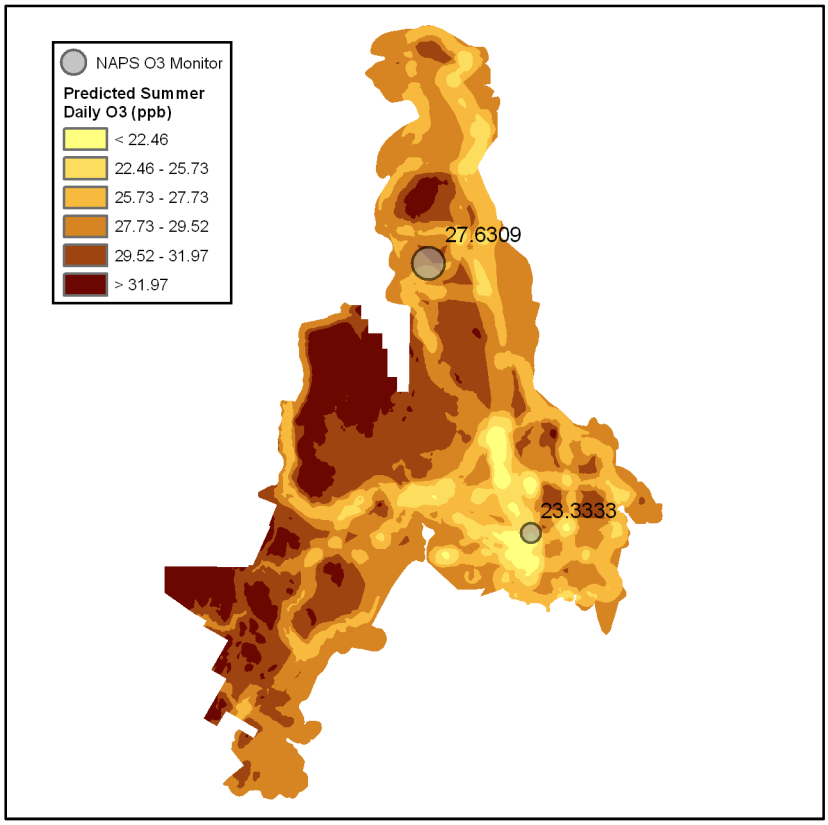

Figure 20. Predicted 2003 summer daylight spatial concentrations of O_3 in Vancouver

No independent O_3 data were available to evaluate these O_3 spatial models. We therefore conducted a leave-one-out analysis where each model (annual, summer, summer daylight) was repeatedly parameterized on N - 1 data points and then used to predict the excluded monitor measurement. The differences between the predicted and measured values at each monitor can be used to estimate model prediction and error.

The results of the leave-one-out analyses for the annual, summer, and summer daylight models are illustrated in Figures 21, 22 and 23. The R^2 for the annual model using leave-one-out analysis was 0.70. The mean (+/-SD) prediction error was 0.3 (2.2) ppb with a range of -4.1 to 8.5 ppb. For the summer model, the R^2 was 0.55 (the decrease in R^2 is the result of one outlier) and the mean (+/-SD) prediction error was 0.4 (3.0) ppb with a range of -4.3 to 14.5 ppb. The summer daylight model R^2 was reduced to 0.45 (decrease in R^2 is the result of the same outlier as in the summer model) and the mean (+/-SD) prediction error was 0.5 (3.6) ppb with a range of -5.9 to 17.5 ppb. The NAPS monitor with the poor predictions (outlier in all models) is Station ID 61004, located in Sarnia. The high density of industrial emissions in Sarnia may be responsible for the poor model performance in this area.

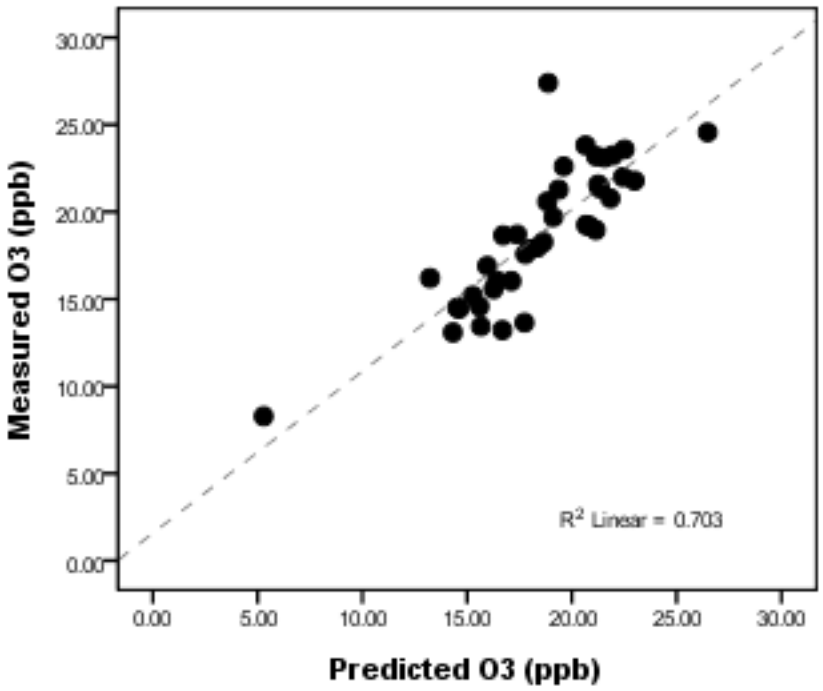

Figure 21. Measured and predicted annual O_3 (ppb) using leave-one-out evaluation.

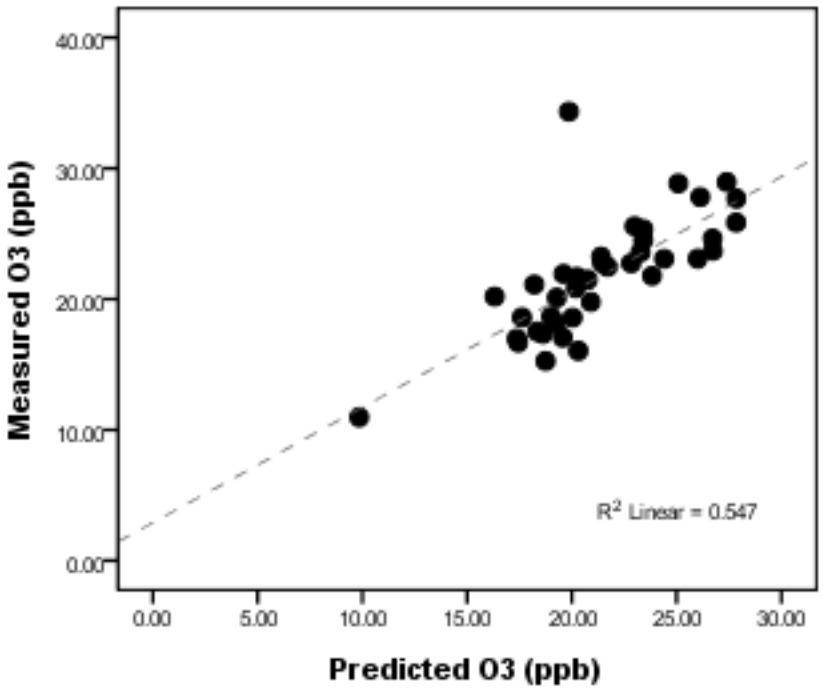

Figure 22. Measured and predicted summer O_3 (ppb) using leave-one-out evaluation.

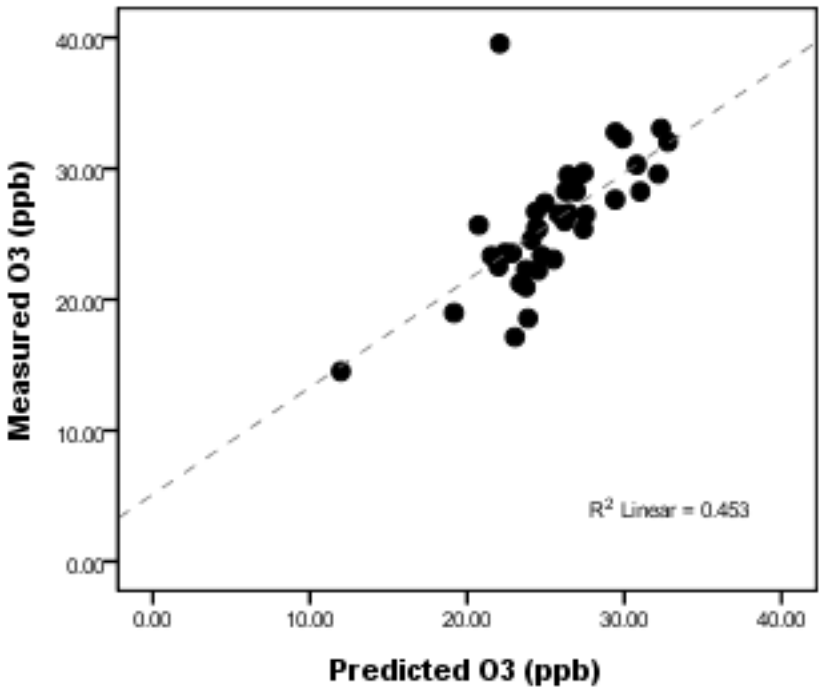

Figure 23. Measured and predicted summer daylight O_3 (ppb) using leave-one-out evaluation.

Conclusions: Creating Spatial O_3 Models for Existing NO_2 LUR Models

A small number of studies were identified in the literature that have attempted to model within-city spatial variability of O_3 using local NO_2 data (either measured or modeled). The modeling approach demonstrated here capitalizes on existing NO_2 LUR models in Canada to examine the within-city spatial variation of O_3. The annual model indicates that O_3 concentrations decrease by 0.64 ppb per 1 ppb increase of annual NO_2, by 0.58 ppb per 1 ppb increase of average summer NO_2, and by 0.75 ppb per 1 ppb increase of average summery daylight NO_2. A large portion of the difference in NAPS O_3 measurements was observed between cities, with smaller within-city differences. This is likely due in part to NAPS monitors not capturing fine-scale O_3 gradients around major roads and high traffic areas. For example, the range of O_3 summer daylight measurements from NAPS in Montreal was 17.1 - 29.5ppb, while the modeled O_3 range was 6.4 - 30.3pbb.

Once a spatial surface of O_3 is created it can be used to determine a "representative area" for each NAPS O_3 monitor. Figure 24a,b illustrates this concept using the NO_2 LUR models for two Victoria NAPS monitors. Representative areas are defined as regions with NO_2 or O_3 concentrations within 5 ppb of the monitors' annual mean. Percentage differences, rather than absolute concentration differences, could also be used. While this report is focused on $PM_{2.5}$ and O_3, it is recommended that representative areas be created for other NAPS pollutants. This, at the very least, will demonstrate the importance of considering spatial variability and measurement representativeness when using NAPS measurement data and would also be useful in siting additional monitors in the future. Figures 25 and 26 illustrate modeled representative areas for two NAPS O_3 monitors in Vancouver and Montreal.

The models developed here are a first step and are meant to illustrate the feasibility of using existing NO_2 LUR models to examine the spatial variation of O_3. While this modeling approach had good model evaluation using the leave-one-out evaluations, there are a number of limitations that must be addressed. The spatial O_3 surfaces created are based on 39 NAPS monitors, and due to current monitor siting criteria very few monitors are located near major roads. Important variables that influence O_3 spatial variation are also not included in these models, such as elevation and green cover. In Vancouver and Victoria this led to the highest O_3 concentrations located in mountain regions, which may be incorrect. Future modeling should incorporate these variables and other important predictors, such as meteorological conditions.

From collocated NAPS data it appears that the strong inverse relationship between NO_2 and O_3 breaks down in high O_3 concentrations, potentially at concentrations greater than 30ppb. During high episodes, O_3 will be elevated through-out the region, overwhelming the influence of

localized titration and limiting within-city spatial variation. It is important to note that current O_3 monitors are sited to avoid traffic influences; and the associations found using NAPS data alone may not capture the scavenging of O_3 in close proximity to major roads. Deterministic approaches drawing on the literature, monitoring siting and landuse characteristics, and new monitoring data that captures intraurban spatial variation of O_3 are needed.

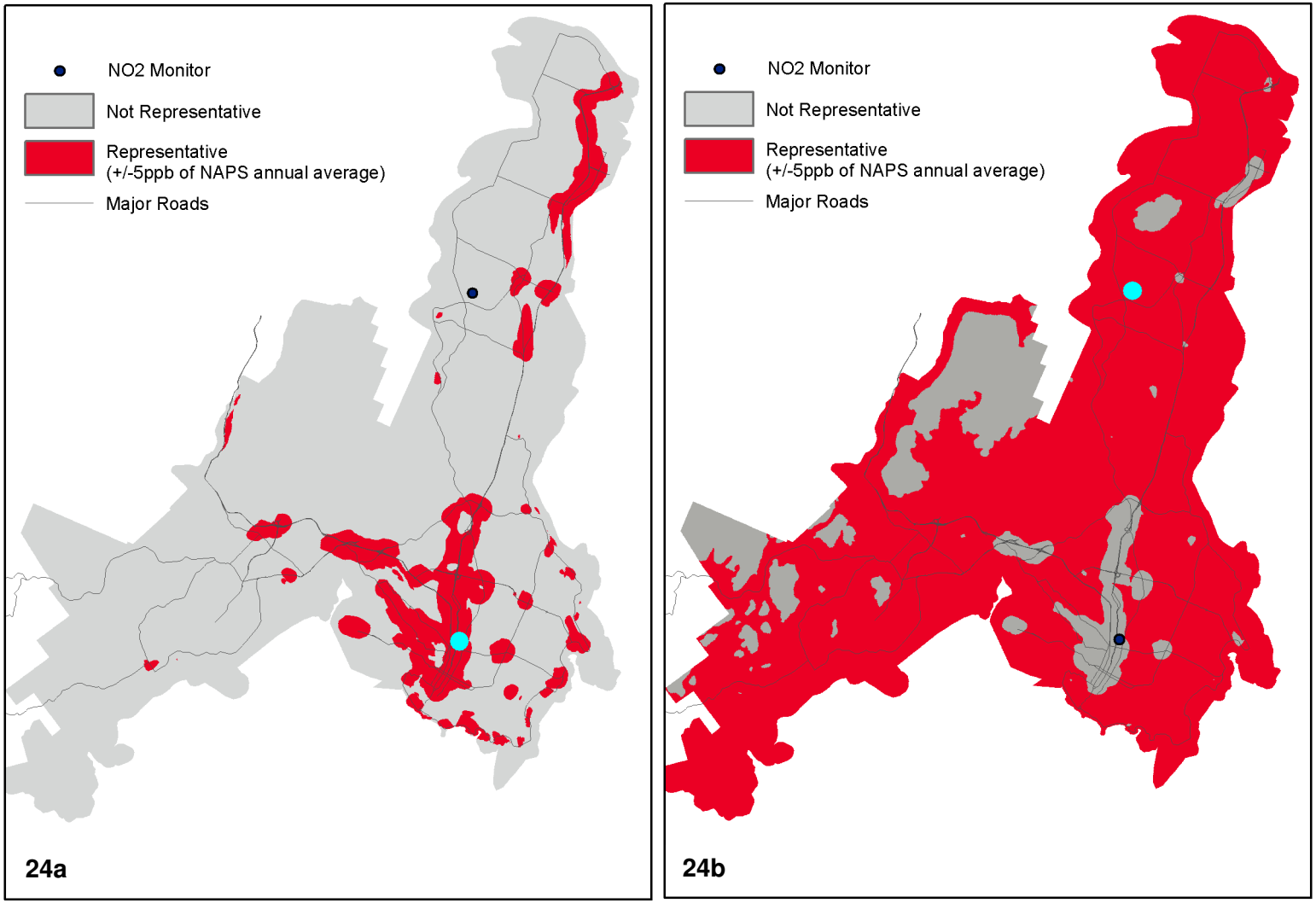

Figure 24. Spatial representativeness of 2006 annual average NO_2 levels (ppb) from NAPS monitor 100314 (20a) and 100304 (20b) (representative areas are defined as regions with NO_2 concentrations within 5 ppb of the monitors' values (NAPS station 100313=5.1ppb; NAPS station 100304=11.8 ppb)

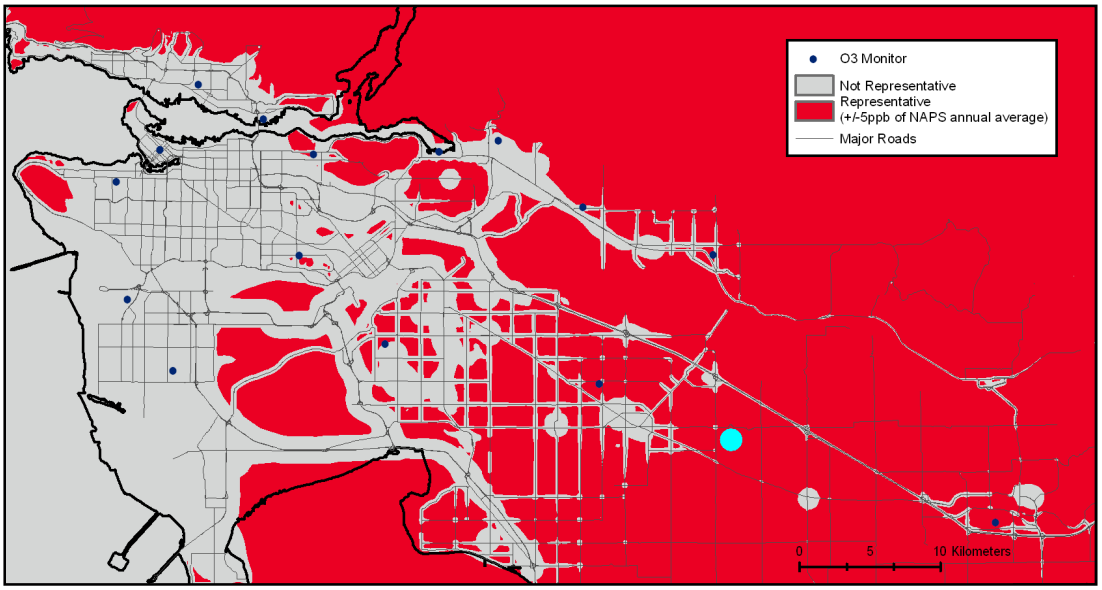

Figure 25. Spatial representativeness of 2003 annual average O_3 levels (ppb) from NAPS monitor 101301(representative area is defined as regions with O_3 concentrations within 5 ppb of the monitors' annual average (21.7ppb).

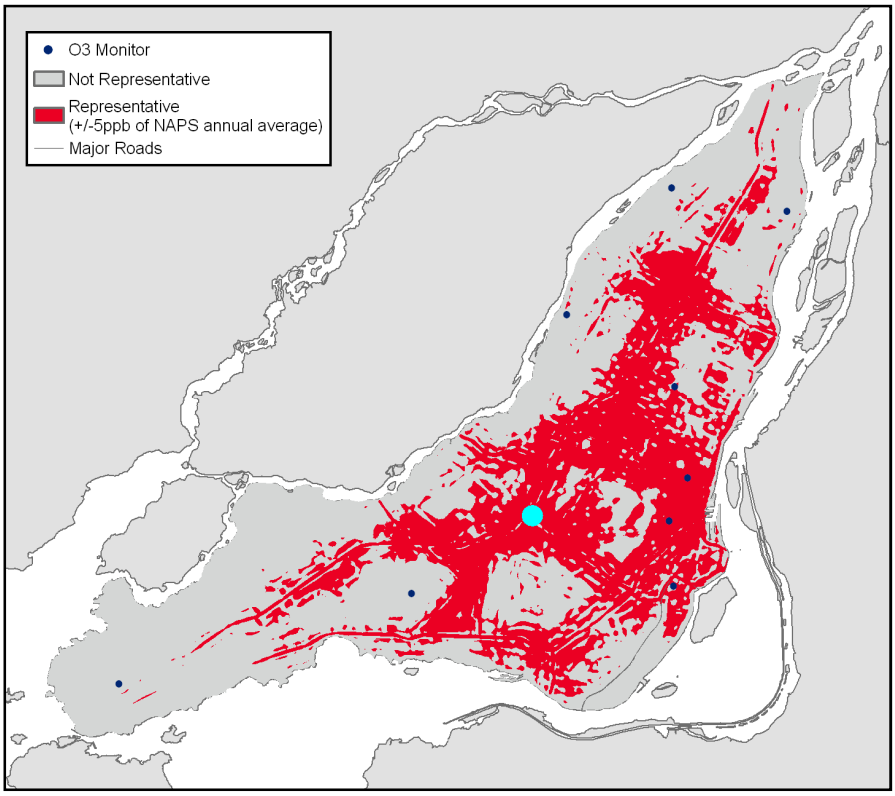

Figure 26. Spatial representativeness of 2006 annual average O_3 levels (ppb) from NAPS monitor 50109 (representative area is defined as regions with O_3 concentrations within 5 ppb of the monitors' annual average (14.5 ppb).

Landuse Characteristics of the NAPS Network

Currently, NAPS monitor siting is characterized using subjective classifications that are intended to represent the dominant landuse within a square kilometer of each monitor. To our knowledge, these codes have not been revised, edited or audited since each station was established. Monitor classification may change from one code to another and NAPS recommends checking each station designations before use (from: http://www.etccte.ec.gc.ca/NapsAnnualRawData /Main.aspx). Table 8 summarizes the current NAPS classifications and frequency of monitors classifications for $PM_{2.5}$ and O_3.

Table 8. NAPS $PM_{2.5}$ and O_3 monitor classifications and frequencies.

Monitor Classifications	$PM_{2.5}$ Monitors	O_3 Monitors	$PM_{2.5}$ and O_3Monitors
A = Agricultural Rural	18 (8.8%)	23 (11%)	24 (9.3%)
C = Commercial	51 (25%)	47 (22.4%)	58 (22.6%)
F = Forested Rural	14 (6.9%)	18 (8.6%)	18 (7%)
I = Industrial	12 (5.9%)	5 (2.4%)	12 (4.7%)
R = Residential	99 (48.5%)	91 (43%)	118 (45.9%)
U = Undeveloped Rural	10 (4.9%)	26 (12.4%)	27 (10.5%)
All Sites*	***204***	***210***	***257***

*one site (NAPS ID: 102602) missing monitor classification.

The representativeness of measurements from NAPS stations is closely tied to monitor siting. Detailed landuse and source characteristics around each monitoring site are therefore needed to examine measurement representativeness.

We used a geographic information system (GIS) and readily available national geographic datasets to derive landuse characteristics around each $PM_{2.5}$ and O_3 monitor. NAPS monitoring site information was provided by Celine Audette (Environment Canada) on December 16th 2010. Included in the following analyses were any $PM_{2.5}$ and O_3 monitor that was active (n=243) or new/proposed (n=13) (2 stations did not have status information and were included). This resulted in 258 NAPS monitors (two station locations were corrected (ID 30501 and 100304). Table 9 summaries the geographic variables derived for each station, and the data sources and geoprocessing tools used for site characterization.

Table 9. Data sources, variables and geoprocessing of NAPS landuse characteristics.

Variables	Data, Source and Format	Geoprocessing
• Population within 0.5, 1, 2, 5, 10, 25 50 km of NAPS Monitor	• Data: 2006 Statistics Canada street block points.	• Spatial join - sum population counts in all points falling with buffer distance.
• Distance to freeway or highway	• Data: 2007 DMTI Road Network (Carto 1 and 2 road classifications)	• Near tool in ArcGIS
• Distance to major road	• Data: 2007 DMTI Road Network (Carto 1,2,3,4 road classifications)	• Near tool in ArcGIS
• Distance to any road	• Data: 2007 DMTI Road Network (All Carto classifications)	• Near tool in ArcGIS
• Length of freeway's and highway's within 0.5, 1, 2, 5, 10km	• Data: 2007 DMTI Road Network (Carto 1 and 2 road classifications)	• Geospatial Modeling Environment (sumlinelengthsinpolys)
• Length of major roads within 0.5, 1, 2, 5, 10 km	• Data: 2007 DMTI Road Network (Carto 1,2,3,4 road classifications)	• Geospatial Modeling Environment (isectpolypoly)
• Length of all roads within 0.5, 1, 2, 5, and 10 km	• Data: 2007 DMTI Road Network (All Carto classifications)	• Geospatial Modeling Environment (isectpolypoly)
• Landuse area (commercial, water, residential, parks, open industrial, government/ institutional) within 0.5, 1, 2, 5, 10 km	• Data: 2007 DMTI landuse • Available for urban areas of Canada only.	• Geospatial Modeling Environment (isectpolypoly)
• Elevation	• Data: GTOPO30 global DEM ~1kmx1km	• Extract values to point
• # of Large industrial emission sources within 0.5, 1, 2, 5, 10 km	• NPRI 2009 Database (emissions to air)	• Spatial join - sum population counts in all points falling with buffer distance.
• Tonnes of air emissions released from large industrial sources within 0.5, 1, 2, 5, 10 km	• NPRI 2009 Database (emissions to air)	• Spatial join - sum population counts in all points falling with buffer distance.

Derived NAPS Monitor Characteristics

An Excel file is provided with all landuse characteristics described in Table 9 assigned to each monitoring station for buffer distances of 0.5, 1, 2, 5, 10 km.

Currently, NAPS monitors do not have a classification representing impact from vehicle emissions. Figure 27 and 28 illustrate the distribution of distances from $PM_{2.5}$ and O_3 monitor stations to the nearest Freeway or Highway and the nearest major road. 73 monitors are within 500 m of a freeway or highway (defined using DMTI road classifications 1 and 2), 61 within 400

m, 54 within 300 m, 36 within 200 m, 17 within 100m and 11 within 50m respectively. In terms of monitor proximity to major roads (defined using DMTI road classifications 1,2,3,4), 163 monitors are within 500 m of a freeway or highway,147 within 400 m, 129 within 300 m, 108 within 200 m, 60 within100 m and 33 within 50 m, respectively. Road length within circular buffer distances of each monitor were also calculated.

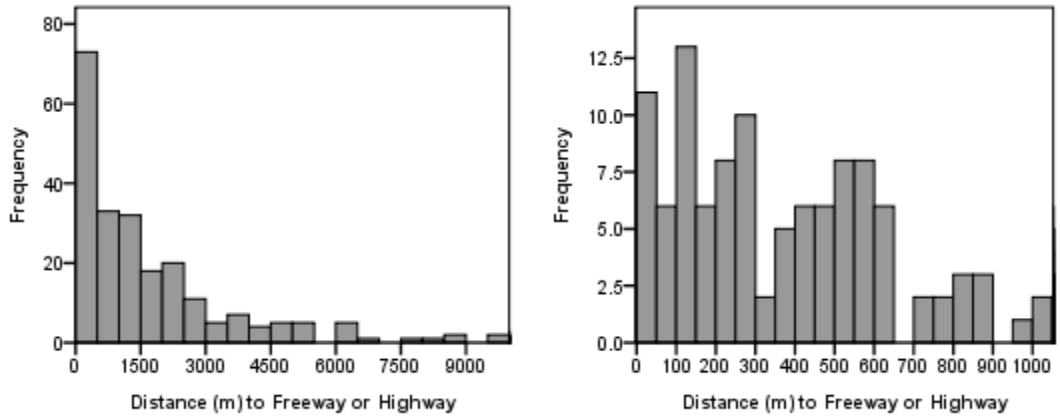

Figure 27. Distribution of distances to freeways or highways (DMTI Carto 1,2) within 10 and 1 km of $PM_{2.5}$ and O_3 monitoring stations

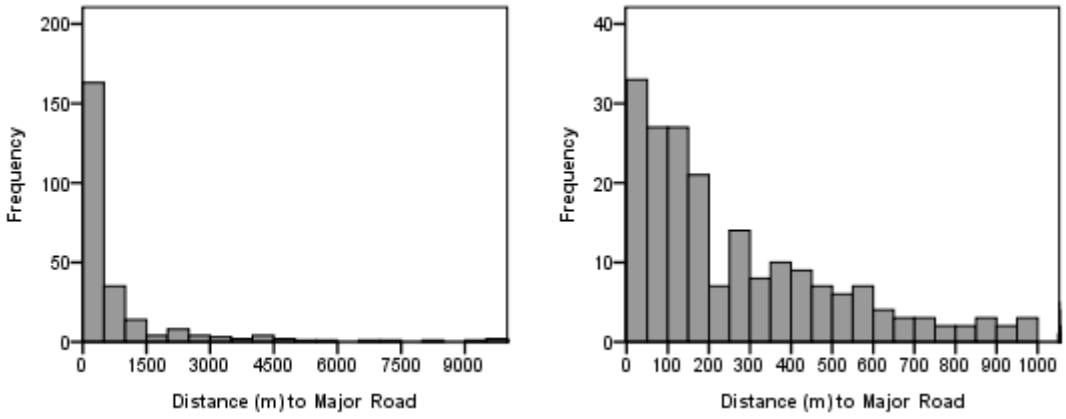

Figure 28. Distribution of distances to major roads (DMTI Carto 1,2,3,4) within 10 and 1 km of $PM_{2.5}$ and O_3 monitoring stations

Proximity of NAPS $PM_{2.5}$ and O_3 monitors to large industrial emissions sources was examined using the 2009 NPRI database (http://www.ec.gc.ca/inrp-npri). The total number of facilities releasing substances to air and the total emissions of substances released to air were calculated for different buffer distances. There were 53 monitors within 500m of an NPRI

emitter, 55 monitors within 1 km, 140 monitors within 2 km, 191 within 5 km, and 208 within 10 km. The total tonnes of emissions within each buffer distance were also calculated. Figure 29 illustrates the number of NPRI facilities reporting air emissions within 1 and 5 km of monitoring sites.

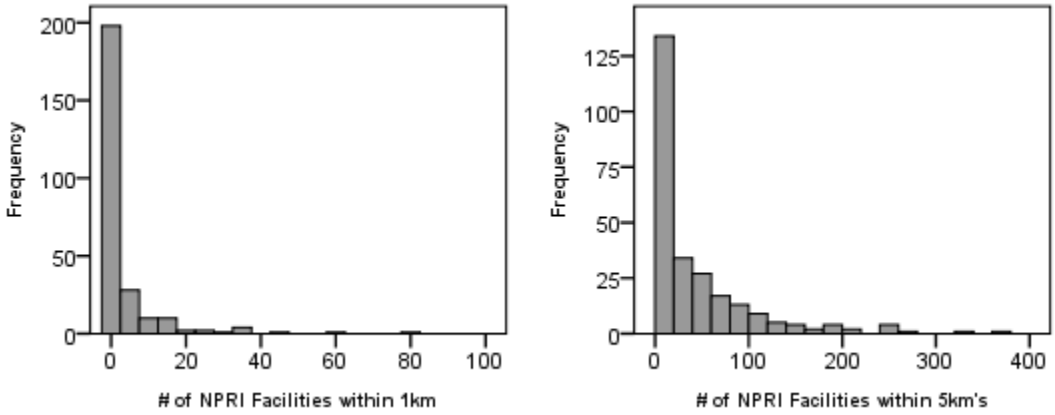

Figure 29. Distribution of number of NPRI facilities reporting air emissions within 1 and 5 km of $PM_{2.5}$ and O_3 NAPS monitors.

Landuse characteristics around monitoring sites were derived using 2007 DMTI landuse data. The landuse dataset is available only in urban areas. Of the $PM_{2.5}$ and O_3 monitoring sites examined, 215 sites (83%) were covered by DMTI landuse data, while 43 sites (17%) were not covered. The area (m^2) of different landuse classifications, including water, open, park, residential, commercial and industrial, were calculating for various buffer distances around each monitoring site.

Population density around monitors was examined using 2006 census block point counts. Population density is an important indicator of emission sources, for example from residential heating, and is also an indicator of monitor population coverage. The mean (SD) population counts within 0.5 km, 1 km, 2 km, 5 km, and 10 km of $PM_{2.5}$ and O_3 monitors are 1203 (1959), 4619 (7034), 15770 (23743), 69496 (106385) and 199901 (330929) respectively. Figure 30 illustrates the distribution of population counts within 0.5 and 5 km of monitoring sites.

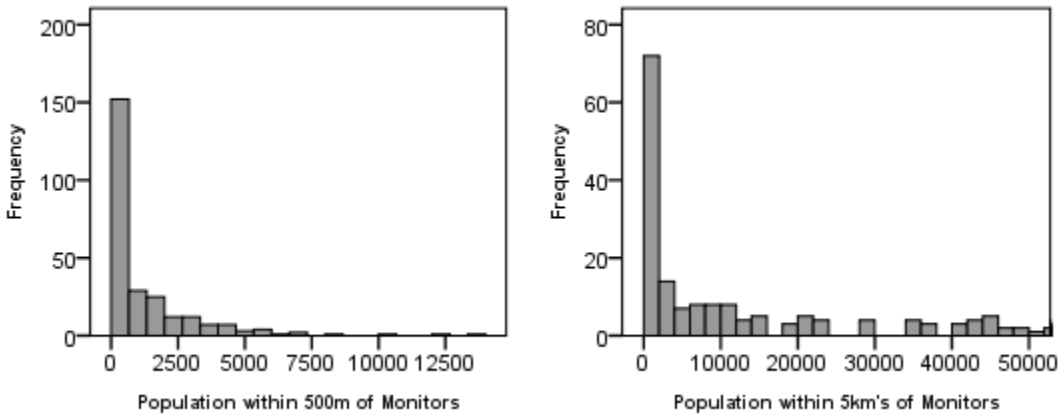

Figure 30. Distribution of population counts within 0.5 and 5 km of $PM_{2.5}$ and O_3 NAPS monitors.

With regard to population coverage of $PM_{2.5}$ and O_3 monitors, an estimated 307,918 individuals (0.97% of the Canadian population) are within 0.5 km of a monitor, 1,154,669 individuals (3.65%) are within 1 km, 11,932,114 individuals (37.74%) are within 5 km, 19,069,846 individuals (60.32%) are within 10 km, 25,026,216 individuals (79.16%) are within 25 km, and 28,241,117 individuals (89.33%) are within 50 km. Figure 31 illustrates the locations of individuals not located within 50km's of a O_3 or $PM_{2.5}$ NAPS monitoring station. Very small differences between NAPS $PM_{2.5}$ and O_3 coverage exist, for example, 13,433,728 and 13,286,608 individuals are not located within 10 km of a $PM_{2.5}$ and O_3 monitor respectively.

The population coverage of the NAPS network using simple proximity measures indicates that the majority of the Canadian population lives within 50km of a $PM_{2.5}$ or O_3 monitoring station and populations not within 50 km are located primarily in rural locations of Canada (as shown in Figure 31). When we restrict to populations living within 10 km of a NAPS station, coverage decreases to 60% of the Canadian population - but is still relatively high in-terms of coverage. It is important to note that simple proximity is not the best surrogate for exposure as different NAPS site may represent different areas of representativeness or exposure situations (as was seen previously in Figures 32, 33 and 34).

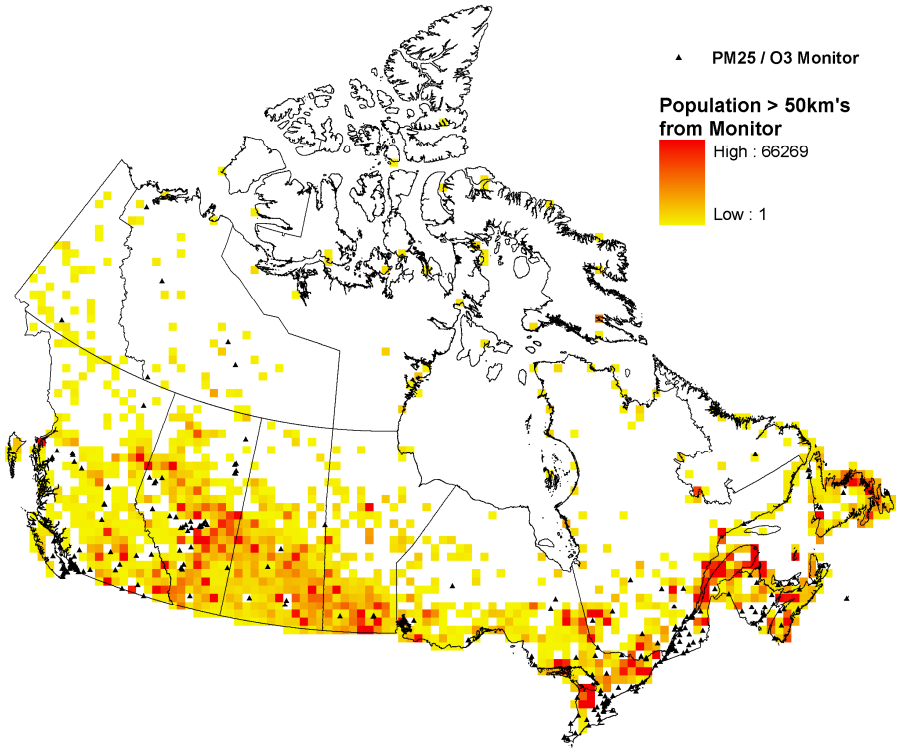

Figure 31. Individuals (n=3,371,780) located > 50 km from $PM_{2.5}$ or O_3 NAPS monitors.

Comparison of Derived GIS Characteristics with Existing Monitor Classifications

The monitor characteristics derived in a GIS were compared against the existing NAPS classifications. Figure 32 through 35 illustrate derived landuse characteristics for different buffer distances (i.e. 0.5, 1, 2, 5 and 10 km) by existing NAPS classifications (i.e. Agricultural, Commercial, Forested, Industrial, Residential and Undeveloped). Overall, there is generally poor agreement between existing NAPS landuse classifications and landuse area derived in a GIS. For example, Figure 32 shows that there are larger areas of residential landuse present in the existing commercial classification compared to the residential classification, and that the industrial classification also contains large areas of residential landuse, especially in buffers >500 m. Similar results are seen for industrial and commercial landuse in Figure 34 and 35. When the number of industrial emission sources and amounts from NPRI are examined there are similar misclassifications, with the current NAPS residential and commercial classes containing large industrial emissions (Figure 35). Current classification for undeveloped, forested and agricultural seem to correspond better to derived variables.

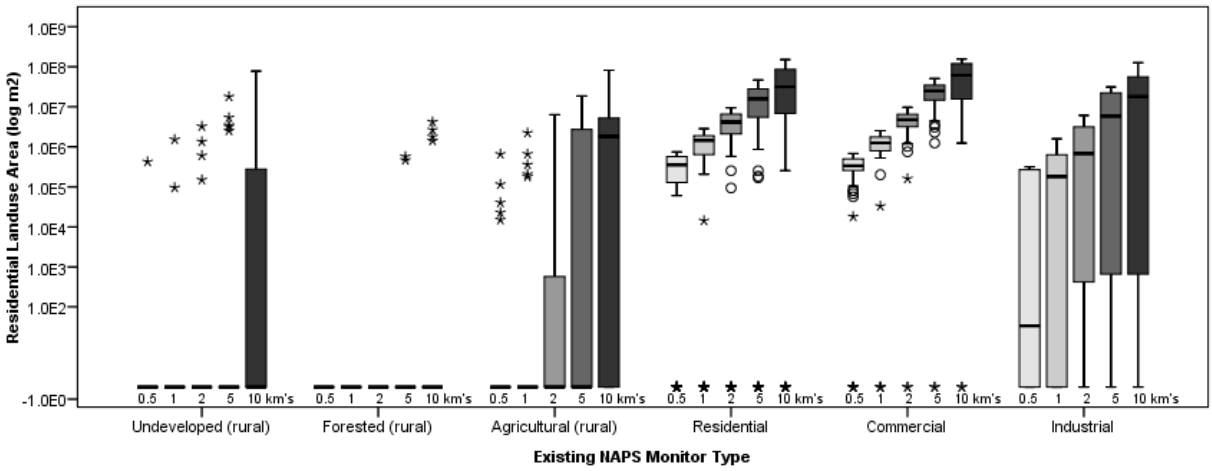

Figure 32. Derived residential landuse area (log m2) by 0.5, 1, 2, 5, 10 km buffers for monitoring sites by existing NAPS siting classifications.

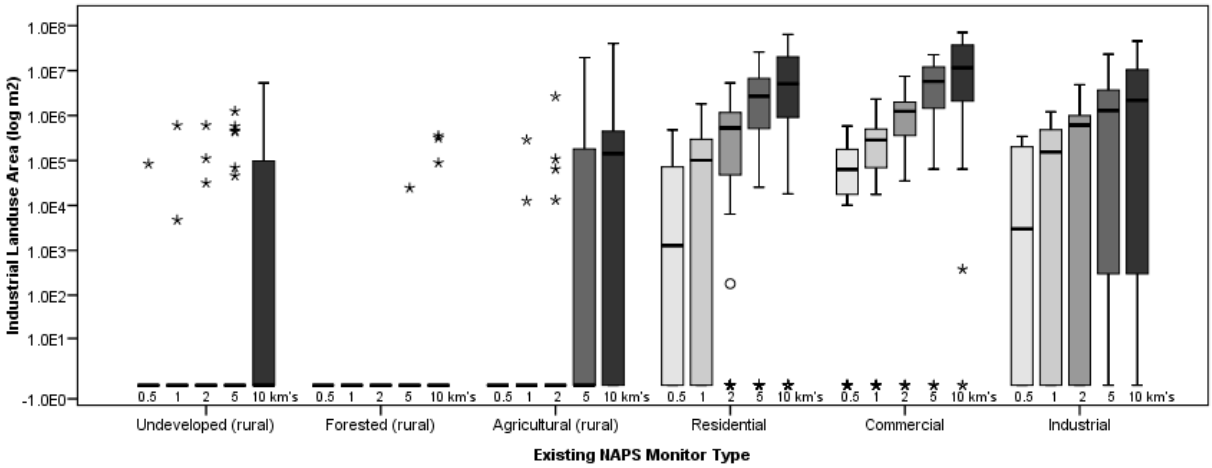

Figure 33. Derived industrial landuse area (log m2) by 0.5, 1, 2, 5, 10 km buffers for monitoring sites by existing NAPS siting classifications.

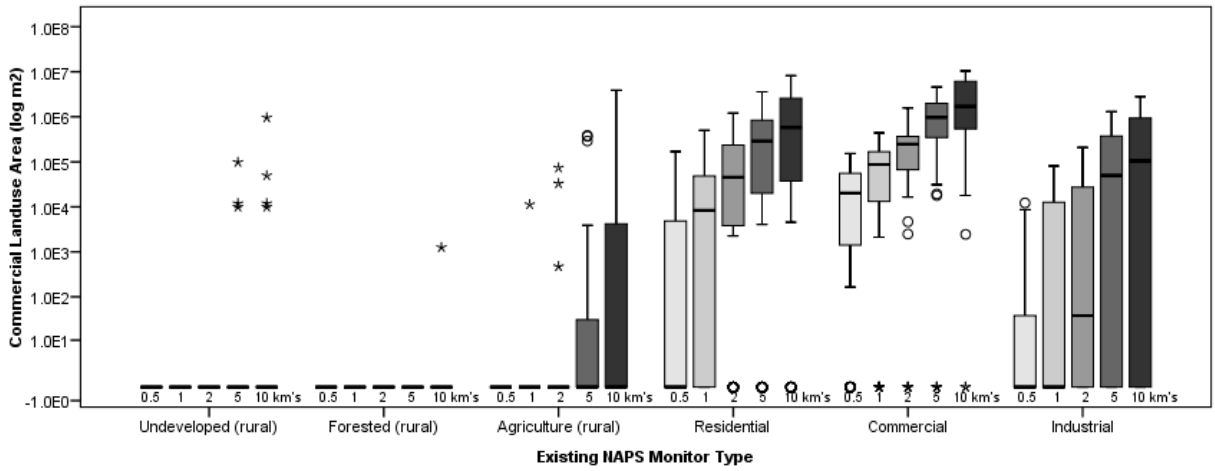

Figure 34. Derived commercial landuse area (log m2) by 0.5, 1, 2, 5, 10 km buffers for monitoring sites by existing NAPS siting classifications.

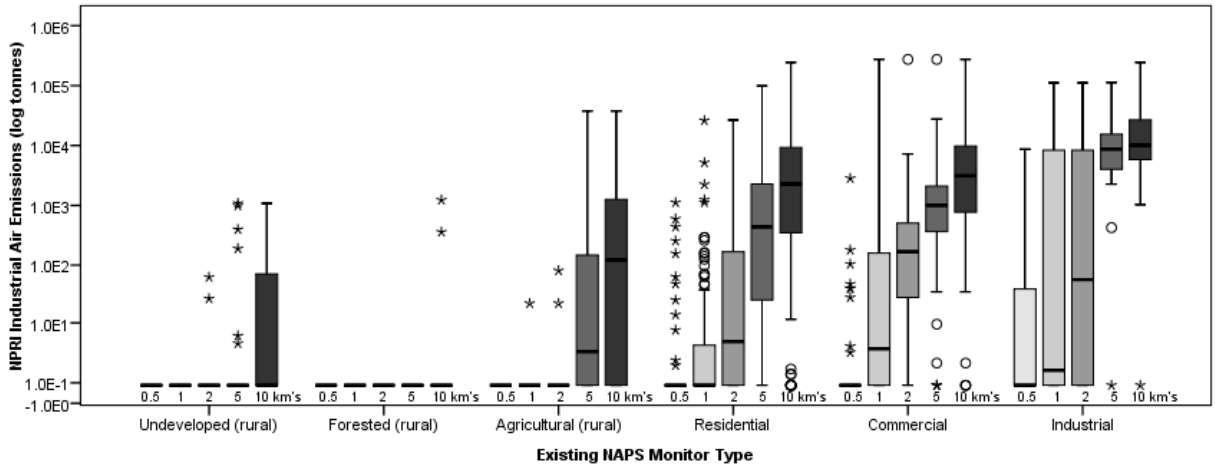

Figure 35. Derived NPRI industrial emissions (log tonnes) by 0.5, 1, 2, 5, 10 km buffers for monitoring sites by existing siting classifications.

Conclusions: NAPS Monitor Siting Characteristics

The derived landuse characteristics for each monitor site correspond poorly to the current NAPS classification, which is based upon expert judgement. Besides being representative of a certain area, a monitoring station may also be representative of a certain emission source, such as industrial or traffic. If detailed data on monitor siting is available for each station, it can provide information and guidance for use of monitoring data for different purposes. The current NAPS monitor classifications (undeveloped, forested, agricultural, residential, commercial, industrial) are too general for this purpose and do not provide detailed

information to inform the spatial representativeness of monitor measurements. The specificity of monitor classifications could be improved to represent specific emission sources, such as traffic and industrial emissions, using standardized classification approaches.

More detailed siting characteristics could be derived for NAPS monitors and added to the variables reported here. Landuse for monitors not covered by DMTI landuse data could be derived from Land Cover, Circa 2000 (http://www.geobase.ca/); the amount of vegetation surround each monitor could be derived using Landsat data and the normalized difference vegetation index (NDVI); detailed topographic characteristics could be derived from fine-scale digital elevation models (DEM's); and meteorological conditions, such as an annual wind rose, could be provided with each monitoring station. While developing siting characteristics from standardized national data is advisable, it is important to recognize the limitations of accurately assessing emission sources with readily available data. For example, industrial sources cover a wide range of processes, substances, and sources (e.g. single stack vs. fugitive emissions) and vehicle traffic counts and profiles are not typically available to assess specific emissions. Land use may change around monitors over time and it is recommended that NAPS periodically update monitor siting characteristics.

Monitor classifications, while useful for monitor interpretation, may be misleading as the monitor siting characteristics of most importance are likely to change with different questions and monitor data uses. Provided with this report is an Excel spreadsheet with all derived characteristics for $PM_{2.5}$ and O_3 NAPS monitors. This information could be used to assess monitor representativeness on a case-by-case basis that would allow users of the NAPS data to more fully understand the siting characteristics of the monitors and any potential implications for the use of the data.

Measurement and Modeling Methods to Assess the Spatial Variability of NAPS Monitors

Ideally, the spatial representativeness of NAPS monitors would be assessed using measurements at various locations in space and time around each monitor. There are various monitoring approaches that could be used, including active, passive and mobile monitors. Complete monitoring coverage (i.e. measuring pollutant concentrations at all points within an airshed) is not feasible and modeling methods, such as dispersion, LUR, satellite derived estimates and interpolation, can also be used to assess spatial representativeness of monitor measurements. Here we briefly introduce each method and summarize their strengths and weaknesses in Table 10. The goal is not to provide a comprehensive list of specific monitoring

equipment available or detailed descriptions of modeling technique, but rather to summarize their strengths and weaknesses for assessing the spatial representativeness of NAPS measurements in the future.

Measurement Methods

A number of monitoring methods are available that could be used to examine the spatial representativeness of NAPS O_3 and $PM_{2.5}$ measurements, including both passive and active monitors and mobile monitors.

Passive monitors, which produce an integrated measurement over the entire monitoring period (generally ranging from 24 hrs to several weeks), operate on the principles of diffusion, where air pollutants being monitored will diffuse or permeate through a semi-permeable membrane and collect on a filter that has been chemically treated to adsorb targeted compounds. This type of monitoring has been used extensively in Canada for LUR based modeling which can provide information on spatial patterns at a resolution of tens of meters. Ogawa samplers measuring NO and NO_2 are most common, but Ogawa passive ozone diffusion samplers (Ogawa & Co. USA Inc.) are also available. In a study by the EPA (2003) excellent correlations were found between passive O_3 samplers and federal reference method O_3 monitors for 24-hr samples. In addition to ozone and NO_x, passive samplers have been widely used for measurement of SO_2 and VOCs. Currently, passive monitoring techniques for PM are not readily available and those that do exist are focused on the coarse fraction. Passive samplers are cost effective and a large number can be distributed to identify fine-scale pollutant variability within a city. The temporal resolution will be limited to the period of monitoring and often fixed-site monitors are used to extrapolate the monitor period to other temporal periods, the most common being annual averages.

Active monitoring methods require that aid of a pump and are based on the measurement of a pollutant within a quantified volume of air. A number of portable active monitors are available for both O_3 (e.g. Timed Exposure Diffusion (TED) ozone sampler) and $PM_{2.5}$ (e.g. Harvard Impactors). While significantly less expensive than continuous monitors or filter samplers routinely deployed in NAPS, equipment costs are still substantial enough that this limits deployment. Monitoring therefore needs to occur at different locations at different points in time. This leads to good temporal coverage but poor spatial coverage and measurements taken at different times will need to be standardized to examine spatial patterns.

Mobile sampling, defined as monitoring methods that collect measurements while traveling through space, has gained popularity during the past decade (Xu et al. 2007). The

Canadian Regional and Urban Investigation System for Environmental Research (CRUISER) is an example of a state of the art mobile monitoring laboratory. The cost of such facilities are very high, but one mobile lab can be deployed to many locations and can respond to changes in wind direction to optimize data collection. Simpler mobile monitoring techniques, such as those incorporating nephelometers, aethalometers or soot photometers within moving vehicles, have also be shown to be useful in identifying spatial patterns in woodsmoke $PM_{2.5}$ (Larson et al. 2007; http://www.nyserda.org/programs/Research_Development/NESCAUM_factsheet.pdf) and in BC from vehicle emissions (Larson et al, 2009). These mobile monitoring protocols typically include one or more fixed-site measurement locations where temporal (with and between-day) variability in the measured pollutant is determined and used to adjust the mobile measurements, resulting in high-resolution (e.g. <100m) information on pollutant spatial patterns. A mobile monitor, however, cannot measure in multiple locations simultaneously and there is a need to determine how well a discrete set of measurements at a given location accurately represent spatial gradients (Xu et al. 2007).

Modeling Methods

Dispersion modeling is a long-established field, and a wide range of models have been developed, from simple plume dispersion models used for a single point or line source, to numerical chemical transport models that incorporate interactions among numerous pollutants and produce three-dimensional spatial estimates for relatively large regions. Input data required for these models include, at a minimum, source characteristics and emissions, meteorological information and locations of receptors at which pollutant concentrations will be estimated. More advanced models include parameters on chemical interactions to characterize the formation of secondary pollutants and require a high level of training to properly run. At present these models (e.g. CMAQ http://www.cmaq-model.org/) run at spatial resolutions of 10 x 10 km or larger, although there are some examples of nested grid models being implemented at spatial resolution of 4 x 4 km, still generally a more coarse spatial resolution than required for within-city pollution variability. Line source dispersion models are routinely used for high-resolution spatial air pollution estimation in Europe (e.g ADMS http://www.cerc.co.uk/environmental-software/ADMS-Roads-model.html; AIRVIRO http://www.smhi.se/airviro; CARhttp://www.integrated-assessment.eu/resource_centre/car_international). In North America, CALINE (http://www.dot.ca.gov/hq/env/air/index.htm) and MOVES (http://www.epa.gov/otaq/models/moves/index.htm) are commonly used to estimate impact of

emissions from single roads but have not been routinely used on a city-wide basis as is common for the European models.

Land use regression is a method of spatial modeling that has become very popular in air pollution health research. Typically, 40 or more passive monitors are deployed throughout the study area, with locations chosen to represent the full range of pollutant concentrations. For each measurement location, predictor variables (e.g. proximity to and length of roads by road class, area of residential or commercial development, population density, etc.) are derived in a GIS and models of measured concentrations produced. Models can then be applied in a GIS to estimate pollutant levels at unmonitored study locations, usually at fine spatial resolutions (5-50 m). Predictor variables used in LUR models are mostly static and annual average concentration surfaces are typically produced; however, some studies have attempted to adjust for shorter term variations in pollution levels by adjusting the annual average up or down using fixed-site monitoring data. The spatial pollutant pattern, however, remains fixed.

Satellite derived pollutant estimates are becoming increasingly important in air pollution research, due to their complete spatial coverage and cost effectiveness. A number of studies have evaluated different remotely sensed concentrations of $PM_{2.5}$ (e.g. Van Donkelaar et al. 2006) and gaseous pollutants (Martin 2008) and found moderate to good associations with ground level monitoring data. Currently, the spatial resolutions of satellite estimates are 4 to 10 kilometers or more, limiting their use in examining intraurban pollution patterns. This will likely change and several studies have used satellite derived pollutant estimates with other local-scale information to improve the spatial resolution of pollution estimates (Liu et al. 2009). In terms of temporal resolution, many satellites have sun-synchronous orbits (i.e. they are overhead at about the same local time on every pass) and provide measurements at the same time each day. Pollution levels derived from these data need to be related to the hourly, daily, seasonal or annual averages at that same location. An exception is the GOES/GASP product, which is collected from a geo-synchronous satellite, and therefore collects data over the same location every 30 minutes; however, the data are not as precise as those gathered by other satellites with less temporal coverage (MODIS or MISR) (Prados et al. 2007). A key advantage of satellite-based estimates is the continuous spatial coverage, thereby reducing reliance on extensive ground-level monitoring. Disadvantages can include the relatively coarse spatial and temporal resolution of the data, the interference of cloud, snow and ice, and the difficulty in determining how much of the pollutant is actually at ground-level (as pollutant measurements are often from the entire air column). A more recent development is the use of satellite-based

estimates in combination with geographic predictors in high-resolution LUR models (Hystad et al, 2011).

There are several interpolation techniques that can be used to estimate pollution values between monitor locations; however, the utility of these methods for capturing fine-scale pollution gradients is limited by the number of measurement points available. For kriging, an interpolation technique that produces an error estimate, the general rule is that at least 50 measurement points are needed to reliably estimate the spatial autocorrelation functions (Hengl et al. 2007). Pollution variability over distances of several hundred meters, such as gradients related to traffic sources, would require a large number of appropriately located monitors for interpolation techniques to capture such small scale gradients.

Conclusions: Intraurban Measurement and Modeling Methods

While a number of measurement and modelling methods are available to assess the spatial representativeness of NAPS monitors they all have inherent trade-offs, particularly with respect to spatial and temporal coverage. Table 10 summarizes the advantages and disadvantages of each approach.

Passive monitors are cheap and can be distributed in large quantities to capture spatial pollution patterns, while active (continuous) monitors are expensive and rarely will a large number be available for simultaneous monitoring. Active monitors, however, have high measurement precision and good temporal resolutions. Fixed site monitors can be used to standardize measurements made at different time-periods to identify spatial patterns, but the influences of meteorology and emission changes are difficult to rule out entirely. Mobile monitors are a fusion of a lot of the advantages of passive and active samples; however, mobile monitors can also be expensive and need to used in targeted campaigns. Cheaper mobile monitoring methods, such as those using nephelometers or aethalometers within vehicles, have been used to identifying spatial patterns in $PM_{2.5}$ and wood smoke. These measurements also have to be temporally standardized to allow for spatial comparisons.

A number of modeling methods are available to estimate spatial pollution patterns. Some of these methods (LUR and interpolation) are dependent on data availability and their predictions will be affected by the number and quality of measurement used to create the models. Dispersion models are well established in Canada but have not been routinely implemented over entire cities in North America as in common in Europe. These models also require detailed traffic information that may not be readily available on a highly time-resolved

basis. Satellite derived pollution estimates are also limited in their ability to capture fine-scale pollution gradients but are useful for regional scale phenomena.

Table 10. Advantages and disadvantages of measurement and modelling methods available to assess the spatial representativeness of $PM_{2.5}$ and O_3 NAPS monitors.

Method	Advantage	Disadvantage
Measurement		
Passive samplers	• Low cost. • Large # of samplers can be distributed to capture fine-scale spatial patterns. • Ease of monitor setup and analysis.	• Time invariant (1 integrated measurement). • Low measurement precision. • Measure period ideally between 1-4 weeks. • Not available for PM
Active samplers	• Good temporal resolution. • High measurement precision.	• Poor spatial resolution. • Relatively high cost per monitor – limits monitor deployment. • Often requires continuous power supply.
Mobile monitors	• Flexibility in spatial coverage of monitoring. • Good temporal resolution. • High measurement precision.	• High cost for extensive mobile laboratories. • May be disproportionately affected by near-source impacts
Model		
Dispersion	• Good temporal and spatial resolution. • # of different models availability. • Can model multiple pollutants. • Can evaluate different emissions scenarios. • Low cost	• Dependent on emission parameters. • Specific emissions parameters are often not available. • Model operation requires considerable time and resources. • Requires model evaluation
Landuse regression	• Good spatial resolution. • Method widely used to capture within-city pollution patterns. • Low cost.	• Poor temporal resolution. • Spatial pattern limited to static predictor variables. • Complex pollution sources/interactions difficult to capture.
Satellite estimates	• Continuous spatial coverage. • Daily measurements. • Low cost.	• Coarse spatial resolution. • Estimates affected by land cover and clouds. • Precision of estimates may vary geographically and temporally. • Not available for all NAPS pollutants
Interpolation of NAP monitors	• Low cost. • Easy to implement.	• Utility dependent of # of monitors available. • Rarely are enough NAPS monitors available to capture fine-scale pollution gradients.

No one measurement or modeling approach can fully assess the temporal and spatial representativeness of NAPS $PM_{2.5}$ and O_3 monitors. Multiple methods are required and the mix of methods will change depending on the project goals. For example, if spatial representativeness is of primary concern then passive samplers and LUR models are best suited. If temporal representativeness is of primary concern then active monitoring methods are most suitable. Ideally, a combination of these approaches would be used to determine a spatial area of representativeness for each monitor and the temporal stability of this area. In the future, data fusion approaches in which information from multiple courses (e.g. satellite-based estimates, chemical transport models, measurements, geographic predictors) are likely to be more common and have promise to provide high resolution spatiotemporal estimates throughout the country.

Conclusions and Recommendations

Our review of the literature showed that small within-city spatial gradients exist for $PM_{2.5}$ mass, but that the extent of variability is case-specific. Very large within-city spatial differences were identified for components representative of primary $PM_{2.5}$ (EC, BC, UFPM) and the use of $PM_{2.5}$ mass is not adequate for capturing the spatial variation of primary $PM_{2.5}$. It is recommended that NAPS identify specific $PM_{2.5}$ components of most concern and more fully characterize their spatial representativeness as well as the resulting population coverage and exposure. For example, recent health research has identified EC and UFP as potentially important indicators of motor vehicle-related air pollution health effects.

Intraurban O_3 variation is likely larger than intraurban $PM_{2.5}$ variation; however, much smaller than that seen for primary $PM_{2.5}$. Due to interactions between NO_x and O_3 there also exists large spatial gradients next to busy roads and in high traffic areas. We provide examples of the use of existing NO_2 LUR models to estimate this fine-scale O_3 spatial variation; however, it is recommended that further models be developed using additional O_3 measurements and other predictor variables. How O_3 concentrations influence the relationship between NO_2 and O_3 also needs to be further examined for such estimates to improve.

Statistical methods could be applied to data from the NAPS monitoring network to examine intraurban variability when multiple monitors exist within a metropolitan area. Currently, 18 metropolitan areas have greater than two O_3 monitors and 23 metropolitan areas have greater than two $PM_{2.5}$ monitors. Correlations and coefficients of divergence should be calculated for monitor pairs using hourly, daily, monthly and yearly averages to identify areas where additional monitoring may be required.

NAPS monitor classifications could be refined to better provide information on source influences or representative area. Minimally, a traffic influenced code should be added to the current NAPS monitor classification. In addition, the full description of monitoring siting characteristics should be made available with NAPS monitoring data as the siting characteristics of most importance will change based on uses. For this reason, simple monitor classifications can be misleading.

Ideally, an area of representativeness around each NAPS monitor should be created (as demonstrated in Figures 20-22) for each NAPS pollutant, using either monitoring or modeling approaches. The representativeness of NAPS monitors likely varies widely and geographic proximity will not be a good surrogate for pollutants with fine spatial variability. The implications of variable areas of representativeness of monitors for population exposure assessment based on NAPS monitors should be examined.

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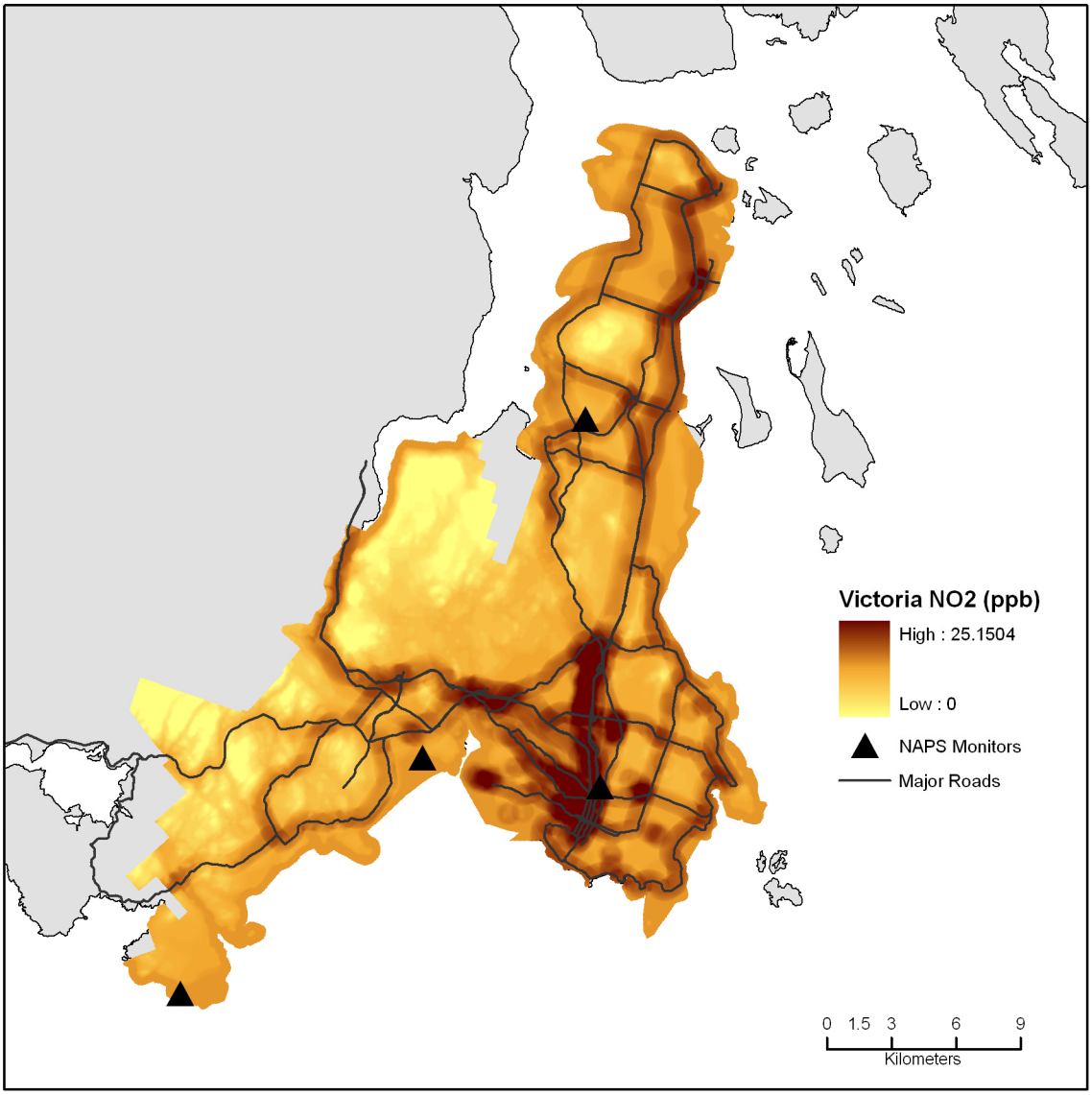
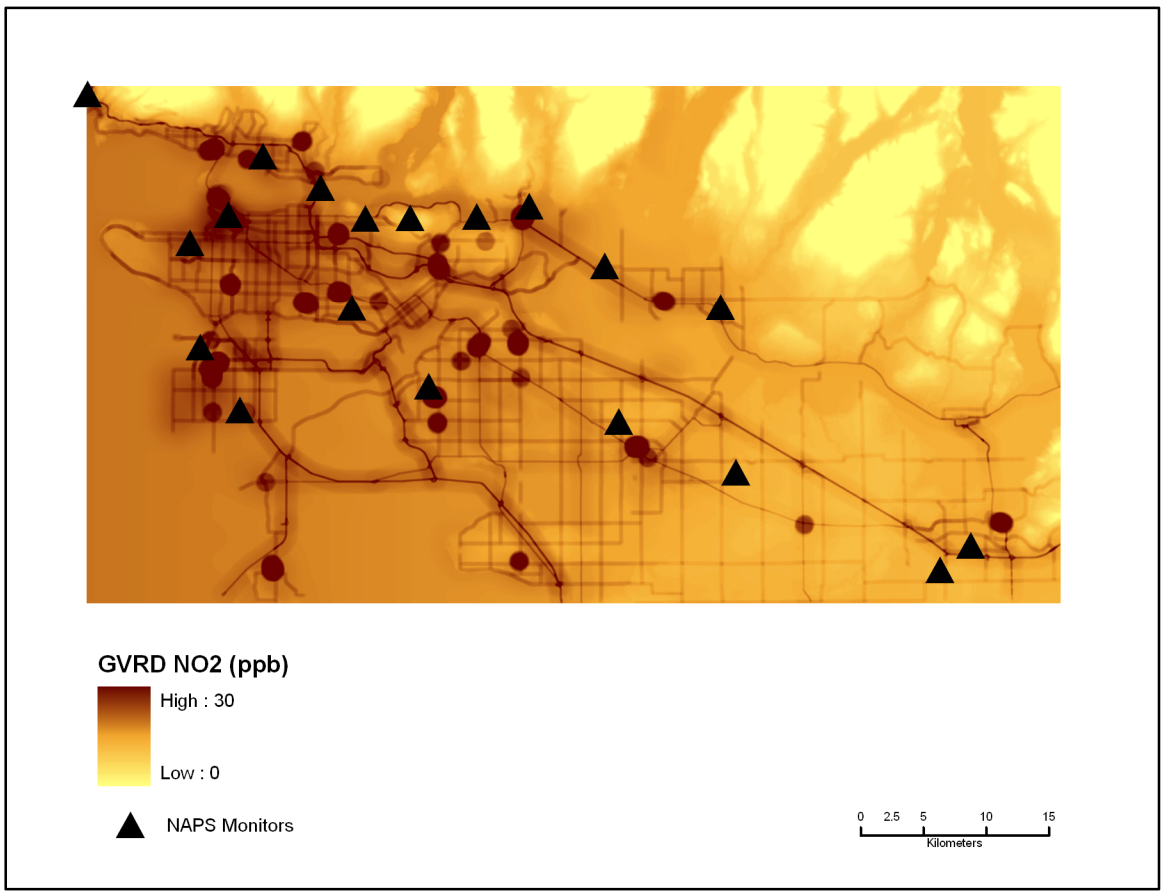
Appendix

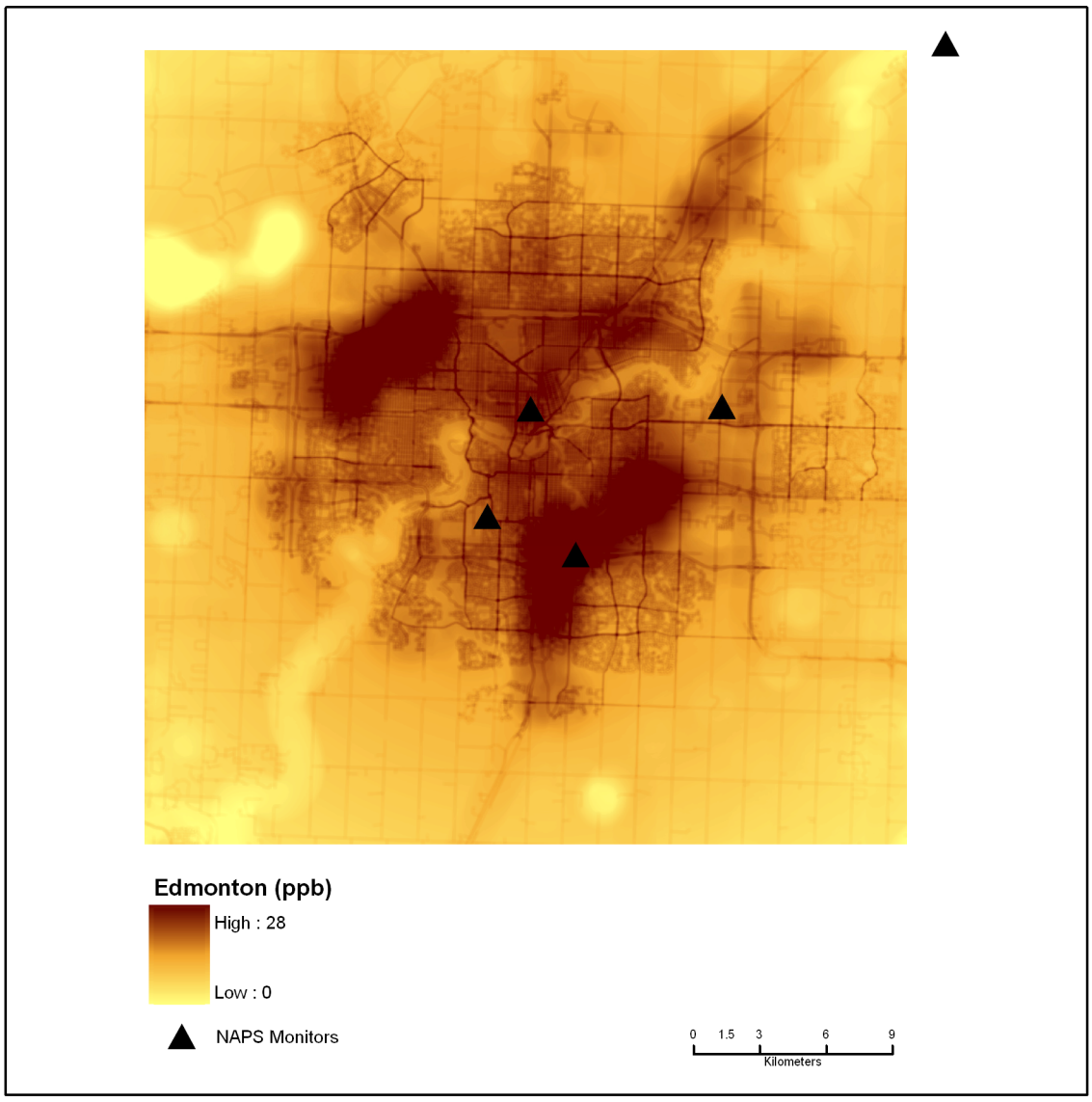
Appendix 1. City-Specific NO_2 landuse regression models in Canada.



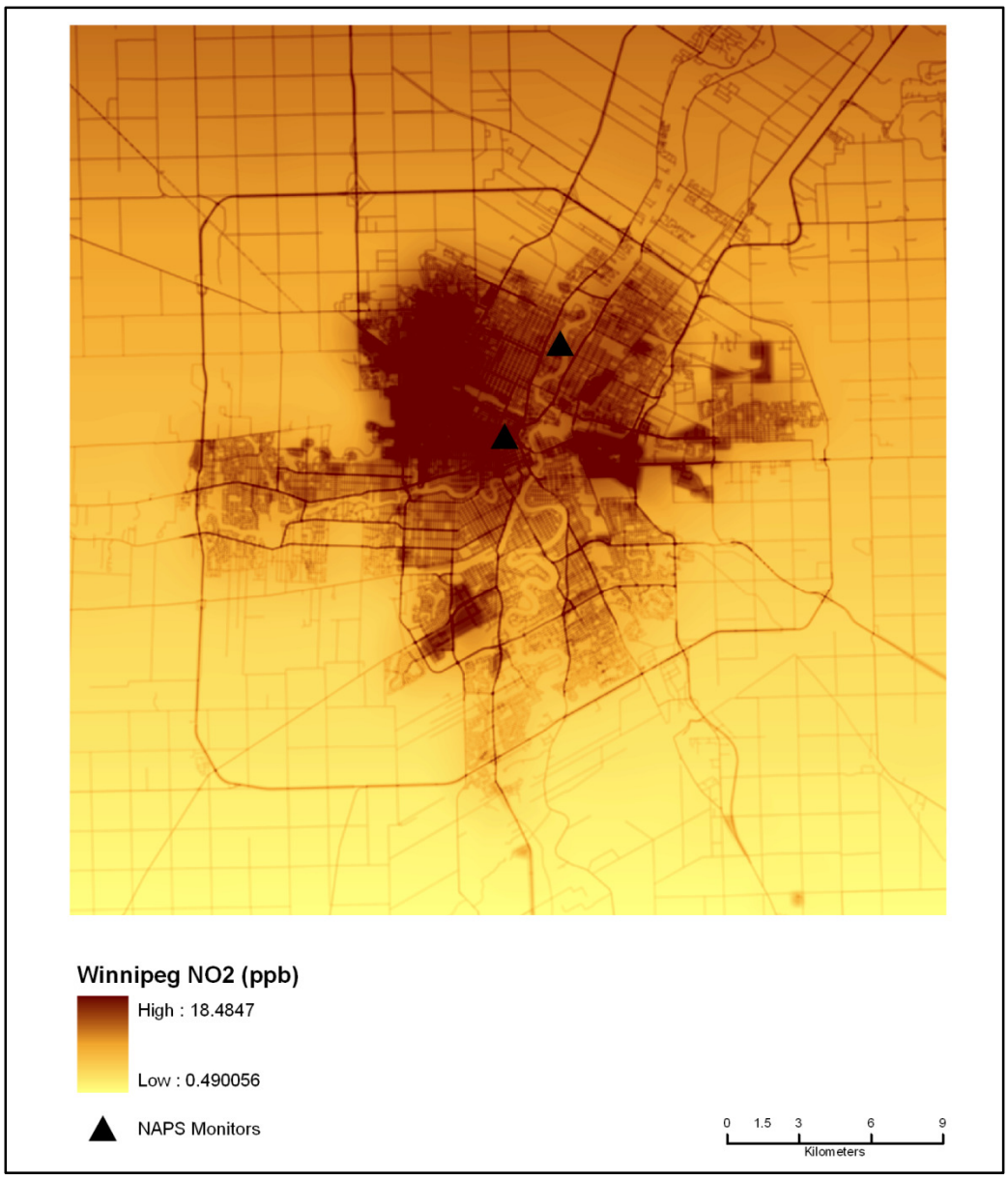
Appendix 1a. Victoria 2006 NO_2 LUR model (from Poplawski et al. 2008)) and NAPS O_3 and $PM_{2.5}$ monitoring stations.



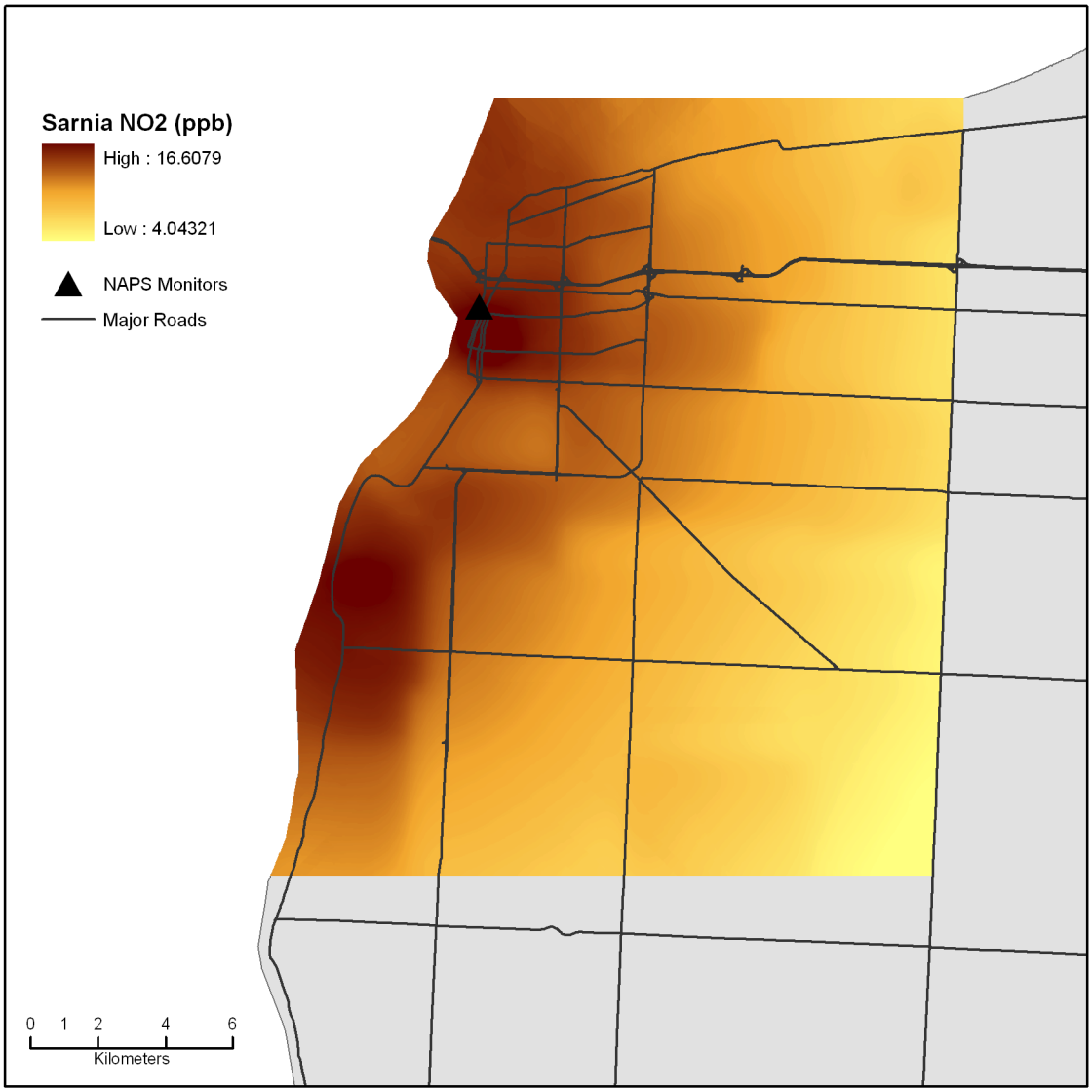
Appendix 1b. Vancouver 2003 NO_2 LUR model (from Henderson et al. 2007)) and NAPS O_3 and $PM_{2.5}$ monitoring stations.



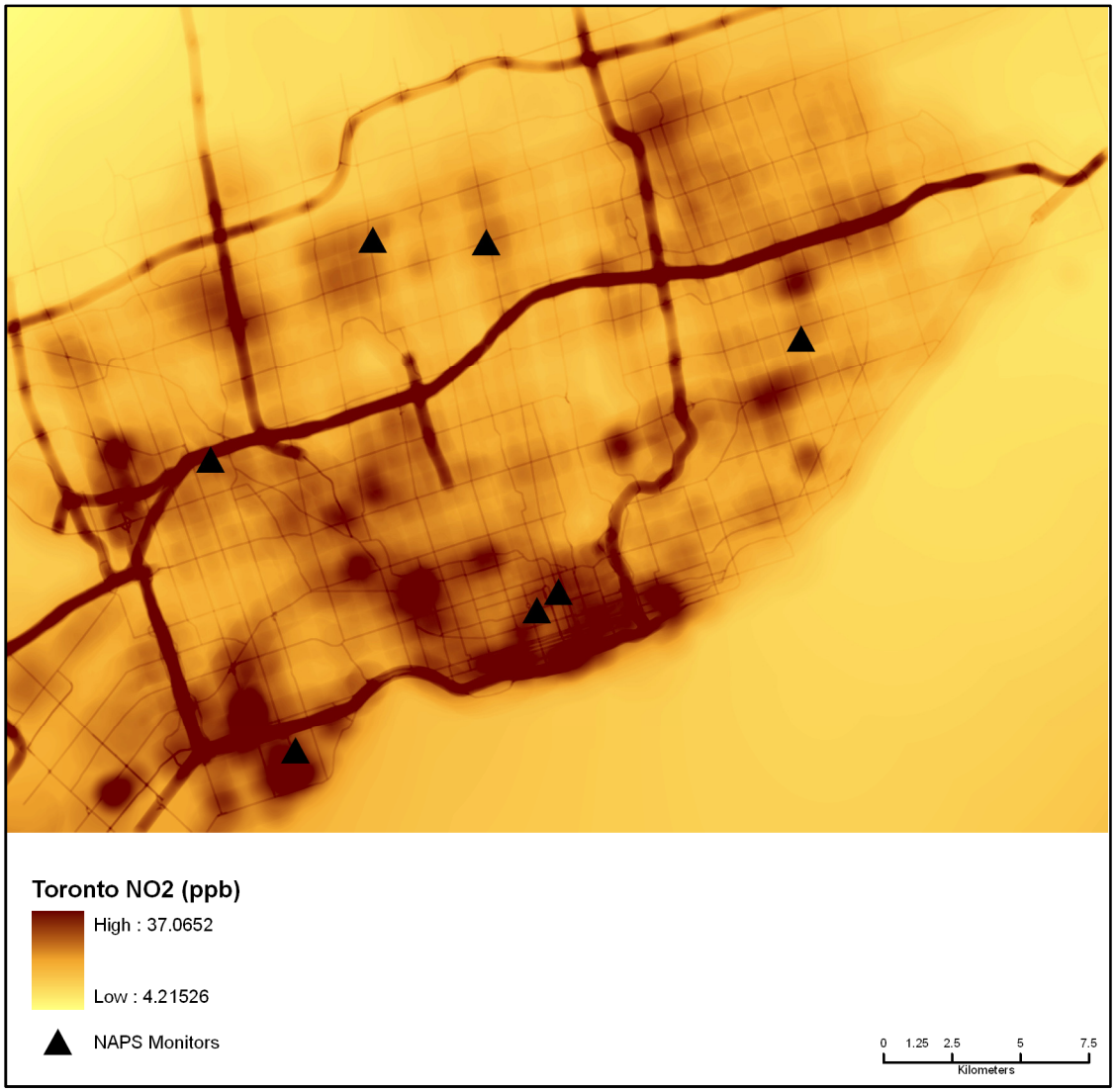
Appendix 1c. Edmonton 2008 NO_2 LUR model (from Allen et al. 2010) and NAPS O_3 and $PM_{2.5}$ monitoring stations.



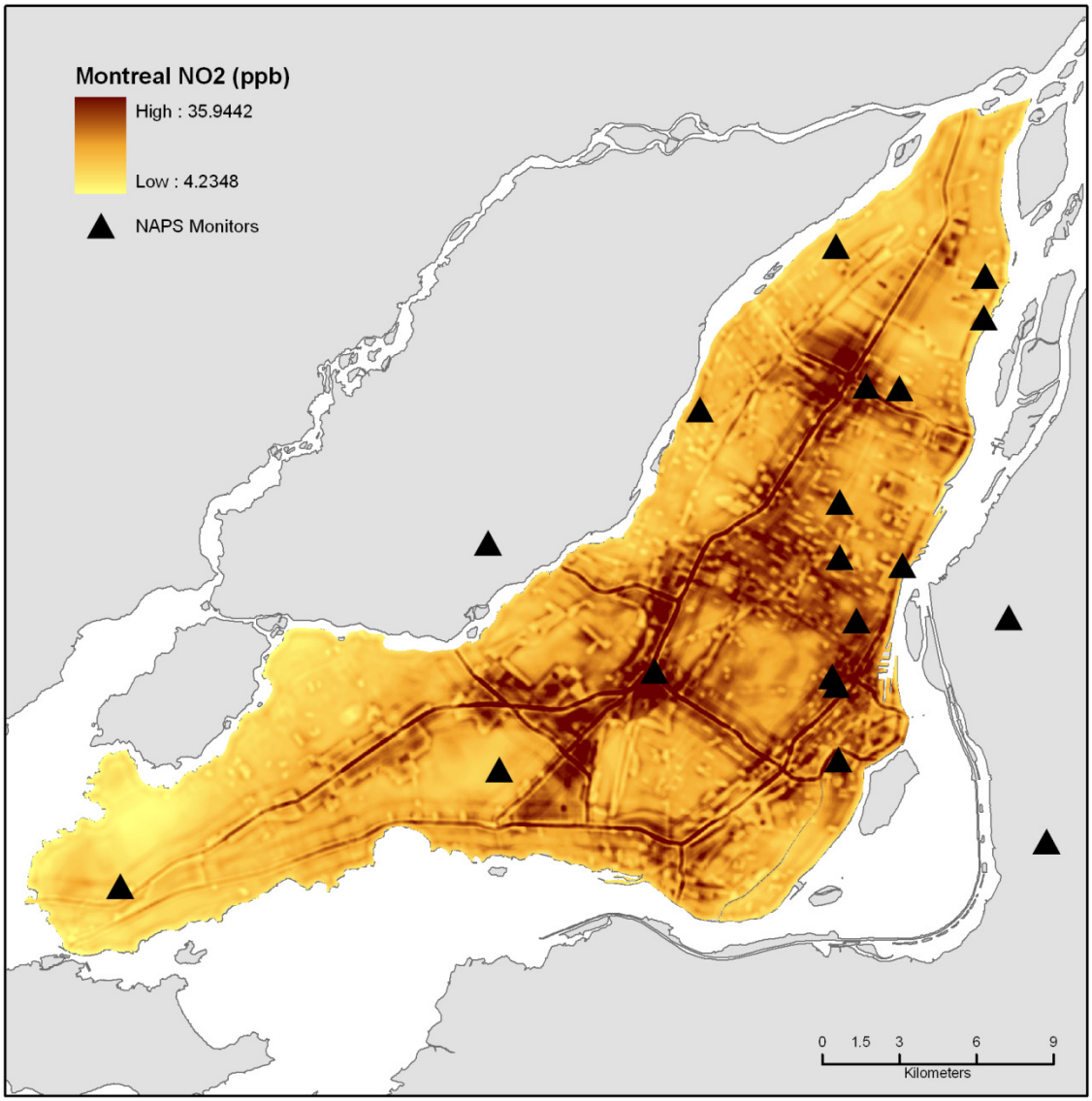
Appendix 1d. Winnipeg 2008 NO_2 LUR model (from Allen et al. 2010) and NAPS O_3 and $PM_{2.5}$ monitoring stations.



Appendix 1e. Sarnia 2005 NO_2 LUR model (from Atari et al. 2008) and NAPS O_3 and $PM_{2.5}$ monitoring stations.

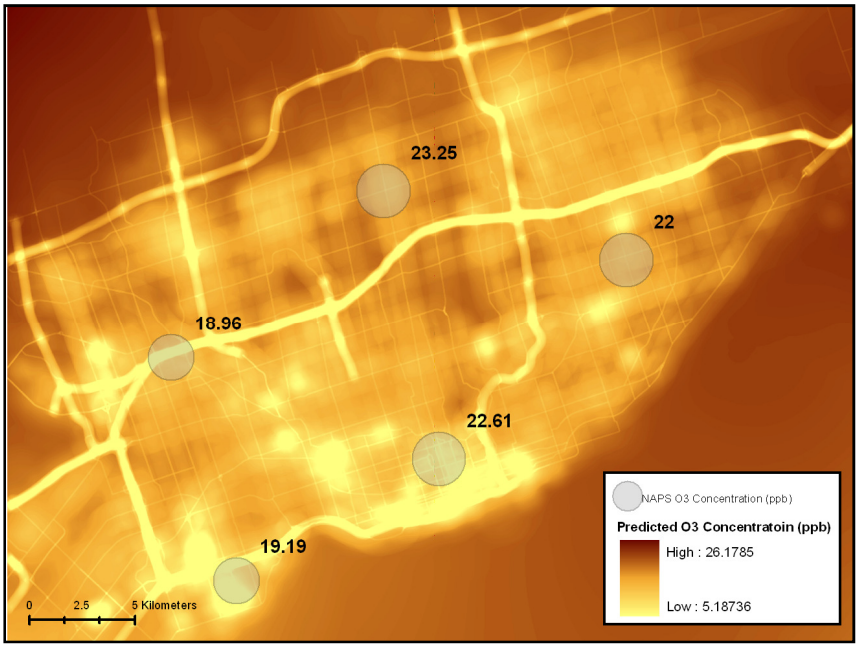


Appendix 1f. Toronto 2006 NO_2 LUR model (from Jerrett et al. 2007) and NAPS O_3 and $PM_{2.5}$ monitoring stations.

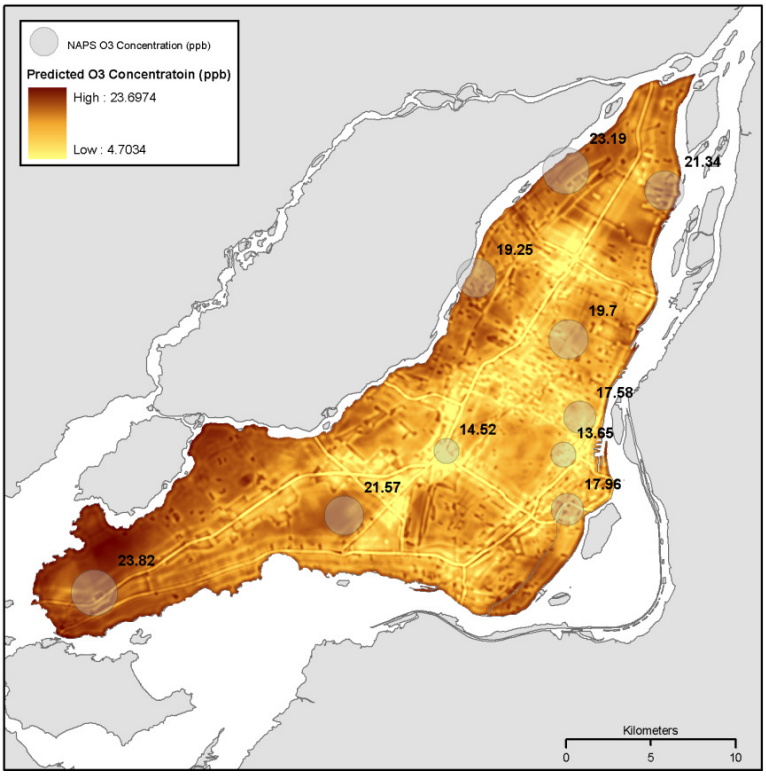


Appendix 1g. Montreal 2006 NO_2 LUR model (from Crouse et al. 2009) and NAPS O_3 and $PM_{2.5}$ monitoring stations.

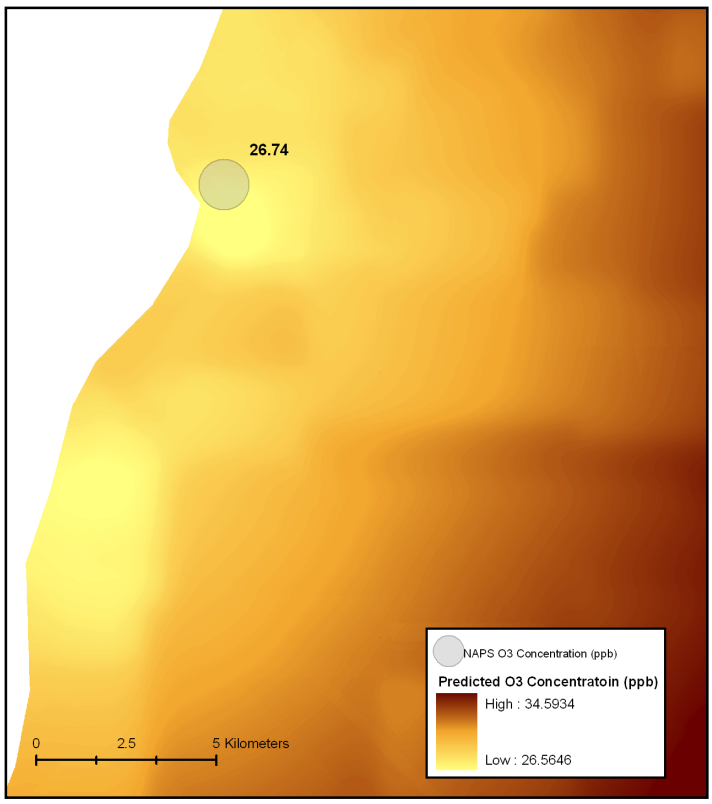
Appendix 2. City-Specific annual average O_3 model predictions.



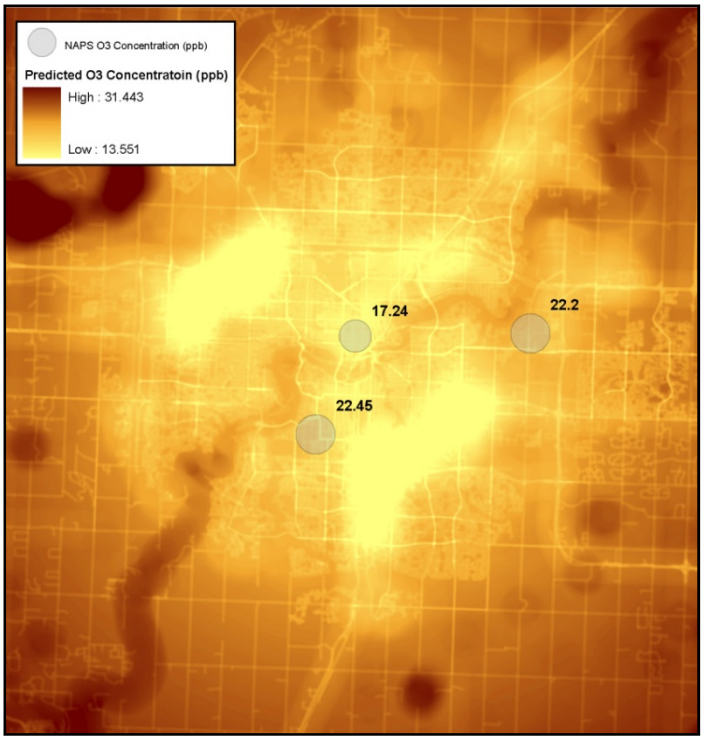
Appendix 2a. Predicted 2006 spatial concentrations of O_3 in Toronto.



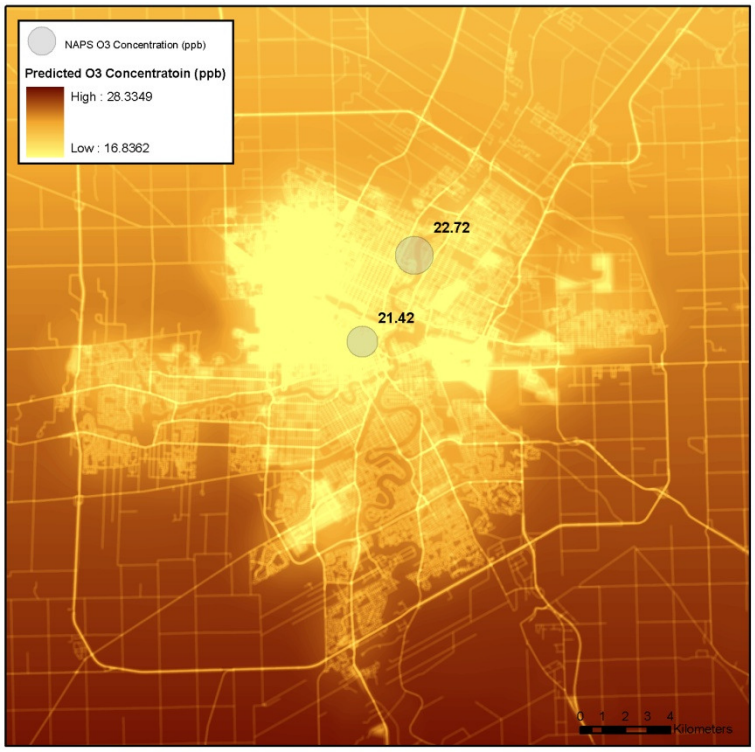
Appendix 2b. Predicted 2006 spatial concentrations of O_3 in Montreal.



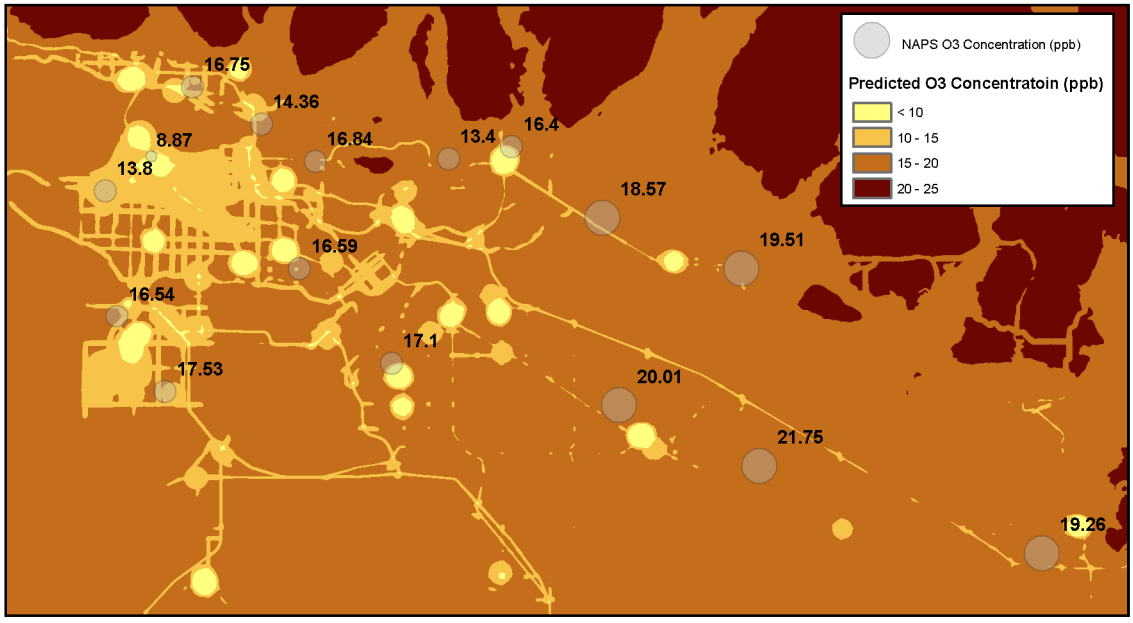
Appendix 2c. Predicted 2005 spatial concentrations of O_3 in Sarnia.



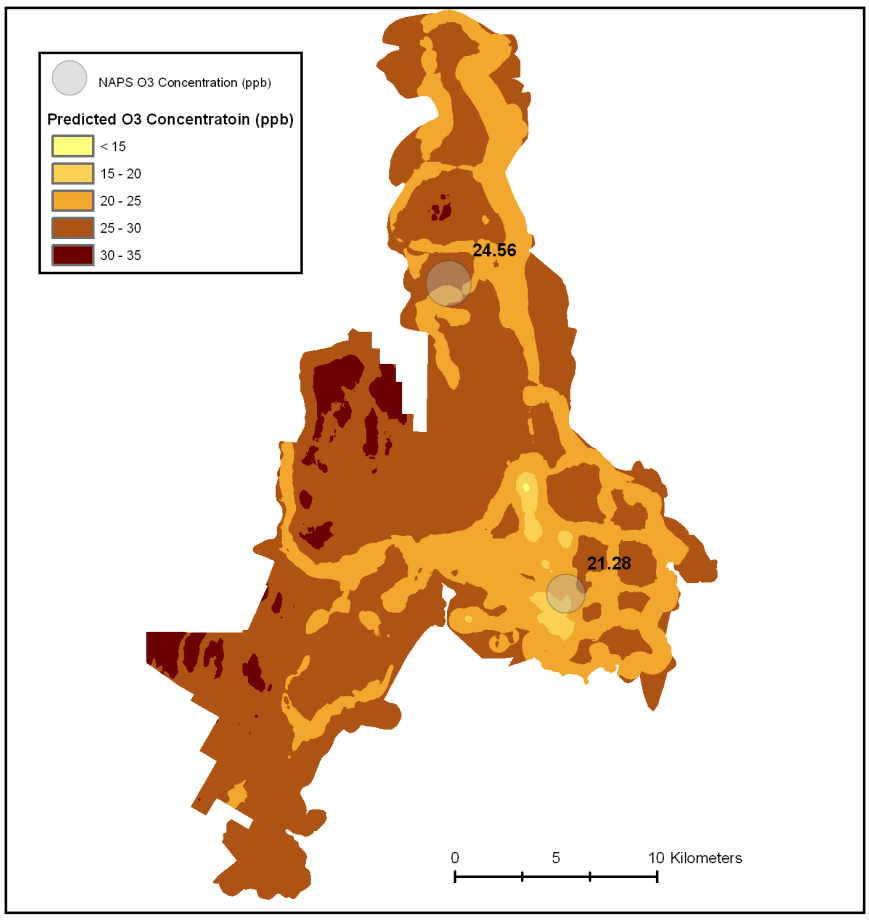
Appendix 2d. Predicted 2008 spatial concentrations of O_3 in Edmonton.



Appendix 2e. Predicted 2008 spatial concentrations of O_3 in Winnipeg.



Appendix 2f. Predicted 2003 spatial concentrations of O_3 in Vancouver.



Appendix 2g. Predicted 2006 spatial concentrations of O_3 in Victoria.