The State of Air Quality in Canada: National Patterns

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Abstract

This chapter provides a description of what air pollutants are routinely monitored in Canada and then focuses on ambient concentrations, geographic differences, temporal patterns and recent trends. In the context of the past 30–40 years, Canadian air quality has been an environmental management success story. Levels throughout the populated regions of the country are considerably lower than in the 1970's and 1980's. The 1990's also brought reductions and this chapter shows that even up through the 2000's improvements in air quality have been realized. However, these recent measurements indicate that there are areas where the current Canadian standards or provincial guidelines are still exceeded for ozone, $PM_{2.5}$ and toxics. Ozone has been the most problematic although there has been improvement in recent years. Similarly, on a national scale $PM_{2.5}$ has decreased. However, pollutants such as $PM_{2.5}$ can have effects on the population at low levels. Thus, air quality remains an important public health issue to track in Canada through routine monitoring; nationally, regionally and at the local scale, where areas of concentrated emissions due to industrial activity or population behaviour (e.g., traffic, wood burning) can lead to higher population exposures. Monitoring is critical in regions of projected industrial or population growth in order to maintain current levels, to identify options for improvement and to inform adaptive management approaches.

Keywords

Particulate matter · Ground-level ozone · Nitrogen dioxide · Volatile organic compounds · Air pollutants · Canadian trends · Air quality monitoring · Hazardous air pollutants

3.1 Introduction

Ambient air quality measurements provide crucial information to the public, policymakers and scientists. This chapter provides a description of what air pollutants are routinely monitored in Canada and then focuses on ambient concentrations, geographic differences, temporal patterns and recent trends.

Ground-level ozone (O_3) and fine particulate matter ($PM_{2.5}$—particulate matter below 2.5 μm in diameter) have traditionally been of greatest interest for monitoring and policy because they are considered to be the main contributors to photochemical smog events. One of the reasons for this is that they tend to build-up in the atmosphere and levels can increase over relatively large areas, from hundreds to thousands of square kilometers. Their slow rate of removal

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from the atmosphere and their formation processes, which involve conversion of other air pollutants, such as sulphur dioxide, nitrogen oxides and volatile organic compounds, in the presence of sunshine, are the main reasons for this build-up.

Another important reason for focus on O_3 and $PM_{2.5}$ is their human health effects. Numerous studies have shown that O_3 has significant respiratory effects, contributing to increases in emergency department visits (e.g., Stieb et al. 1996) and hospital admissions (e.g. Burnett et al. 1997). Ozone has also been linked to an increased rate of mortality for respiratory illnesses (e.g., Jerrett et al. 2009). Long-term exposure to $PM_{2.5}$ is considered to present the greatest mortality risk among air pollutants due to its significant cardiovascular effects (e.g., Brook et al. 2010; Pope et al. 2004). Recently, Crouse and co-workers (2012) found that long term outdoor $PM_{2.5}$ exposures across Canada during the 1991–2001 period (i.e., outdoor concentrations), which, compared with many countries in the world, were relatively low, are having a detectable impact on life expectancy. Furthermore, the magnitude of this effect is similar to what has been found in other countries. These results, as well as the large body of recent health effects research (Pope and Dockery 2006), suggest that public health benefits can be achieved by further reducing average $PM_{2.5}$ in Canada.

Several other air pollutants, known collectively as air toxics or hazardous air pollutants (HAPs) have also been identified as having the potential to induce adverse health effects. This category of pollutants generally excludes criteria air contaminants (i.e., those mentioned above) whose concentrations tend to be higher than those of most HAPs.

National scale patterns of air pollutant concentrations, temporal patterns and $PM_{2.5}$ and volatile organic compound (VOC) composition during the decade of the 2000's are presented in this chapter. First, the main national monitoring networks are described. Ozone and $PM_{2.5}$ are then the main focus starting with a discussion of background levels and finishing with information on recent trends in annual concentration. The latter sections present data and discuss spatial and temporal patterns for nitrogen dioxide (NO_2), nitric oxide (NO), VOCs, sulphur dioxide (SO_2) and hazardous air pollutants.

Policy Context Canada-wide Standards (CWS) In June 2000, the Canadian Council of Ministers of the Environment (CCME), except Québec*, signed the Canada-wide Standards for Particulate Matter and Ozone, which comprise ambient targets to be achieved by 2010. Each jurisdiction is committed to actions aimed at meeting the Standards for PM and Ozone by reporting on compliance once the target date is reached in 2010. This includes comprehensive reports every five years on all provisions of the CWS beginning in 2006 and annual reports on compliance and maintenance of the CWS beginning in 2011.

The CWS for $PM_{2.5}$ of 30 μg m^{-3} is based on a three year average of the 98th percentile of 24-hr measurements and requires daily sampling. The CWS for O_3 of 65 ppbv is based on a three-year average of the 4th highest daily 8 h average.

Management of HAPS The compounds considered to be HAPs differ by jurisdiction. The United States (U.S.) has developed a formal list of HAPs through its Clean Air Act (US EPA 2010) and the U.S. Environmental Protection Agency has released several National Air Toxics Assessments in recent years (US EPA 2013). In Canada, no such formality exists, though many atmospheric contaminants found toxic under the Canadian Environmental Protection Act (CEPA) fit the common definition of HAPs. Compounds declared CEPA-toxic have been managed individually under the Toxic Substances Management Policy (TSMP) (Canada 1995). CEPA-toxic compounds that are persistent, bioaccumulative, and predominantly anthropogenic are assigned to TSMP's Track 1 requiring virtual elimination from the environment. Compounds not meeting all TSMP Track 1 criteria are assigned to Track 2 with a goal of preventing or minimizing releases to the environment.

3.2 Routine Monitoring in Canada

Ambient air quality monitoring across Canada is conducted by government, industry, and in some provinces, non-government organizations (NGOs). Many of the industrial monitoring sites are established in close proximity to their facilities and are usually required as part of a provincial permit to operate. In some jurisdictions industry also collaborates with government (and NGOs) to operate regional monitoring networks. Most of the air quality measurement data presented in this chapter are from the National Air Pollution Surveillance (NAPS) network. However, another important national network is the Canadian Air and Precipitation Monitoring Network (CAPMoN) and some data from this program are also used in this chapter.

The National Air Pollution Surveillance (NAPS) Program is a joint federal, provincial, territorial and municipal initiative. The purpose of this Program is to coordinate the collection of air quality data from existing provincial, territorial and municipal air quality monitoring networks and provide a unified Canada-wide air quality data base. Sites designated as NAPS sites are selected by Environment Canada and the Province or Territory in which the site is located, primarily to support national air quality programs like the CWS and the national ambient air quality objectives NAAQOs. Several provinces, territories and regional governments operate additional monitoring stations to meet the requirements of their own ambient monitoring programs. Ambient air pollution data from most of these sites are also submitted to the NAPS Canada-wide database at the discretion of each monitoring agency.

Table 3.1 provides 2009 information on each of the main pollutants monitored, such as number of operating sites and measurement frequency. The associated provincial/territorial/regional monitoring networks reporting data to the NAPS

Table 3.1 Overview of the main NAPS and CAPMoN ambient air pollutant measurements across Canada for 2009. A complete listing of observations by NAPS site code can be found at http://www.etc-cte.ec.gc.ca/NapsData/Web-Information

Pollutant	Time resolution (h)	Measurement method	No. of Stns	Start year	Comments
O_3	1	UV light absorption	212		Reporting to CWAQD[f]
O_3	1	UV light absorption	16		CAPMoN
NO_2	1	chemiluminescence	147		
NO	1	chemiluminescence	147		Reported nationally since 1986
SO_2	1	pulse-fluorescence ultraviolet (UV) adsorption	124		Reporting to CWAQD[f]
SO_2	24	integrated open-face filterpack	13		CAPMoN multiple PM constituents
$PM_{2.5}$	1	Tapered element oscillating microbalance (TEOM)[a]	134		Urban
$PM_{2.5}$	1	TEOM with filter dynamic measurement system (FDMS)	11		Urban
$PM_{2.5}$	1	Beta attenuation (BAM)	51		Urban
$PM_{2.5}$[c]	24	dichotomous sampler or FRM sampler[b]	41		Every 3rd or 6th day 0-0 LST
PM_{coarse}[c]	24	dichotomous sampler	30		Every 3rd or 6th day 0-0 LST
$PM_{2.5}$	24	Speciation sampler[d]	13	2002	Every 3rd day 0-0 LST
VOC[e]	24	Summa™ polished canisters	37		Urban Every 6th day 0-0 LST
VOC[e]	4	Summa™ polished canisters	14		Rural Every 6th day 12–16 LST

The Province of Québec, while not a signatory to the CWS, has undertaken analogous efforts as those covered by the CWS and has also developed working inter-jurisdictional arrangements for many provisions of the CWS.

[a] Beginning in 2002 many TEOM instruments in the NAPS $PM_{2.5}$ network were fitted with a sample equilibration system (SES). The SES incorporates a low-particle-loss Nafion dryer allowing for conditioning of the PM sample stream to a lower humidity and temperature level. Unless indicated otherwise, all $PM_{2.5}$ mass data in this chapter are from TEOM-SES instruments operated at 30 °C or TEOMs operating at 40 °C

[b] FRM = EPA Federal Reference Method

[c] The pre and post weighed Teflon filters from the dichotomous and FRM samplers are analyzed for elements using energy dispersive X-ray fluorescence (EDXRF) and for anions and cations using ion chromatography (IC)

[d] This particle speciation program was designed for more accurate and complete measurement of all the important components of $PM_{2.5}$. Along with the major ions (e.g., sulphate, nitrate, ammonium) and mass and trace elements from the co-located dichotomous sampler, this includes organic carbon (OC), elemental carbon (EC), gaseous ammonia and nitric acid. By 2006 this program included 14 (4 rural and 10 urban) sites, which are a subset of the $PM_{2.5}$ dichotomous sampler locations. A complementary short-term program was initiated at 5 CAPMoN sites following the same methods

[e] Content of canister analyzed for more than 100 different C_2 to C_{12} volatile organic carbon (VOC) species; Beginning in 2003 a number of polar species including α-pinene, β-pinene, δ-limonene and camphene began to be quantified

[f] Stations reporting to the Canada-wide Air Quality Database (CWAQD) include NAPS designated sites, CAPMoN stations and some additional provincial stations not designated as "NAPS"

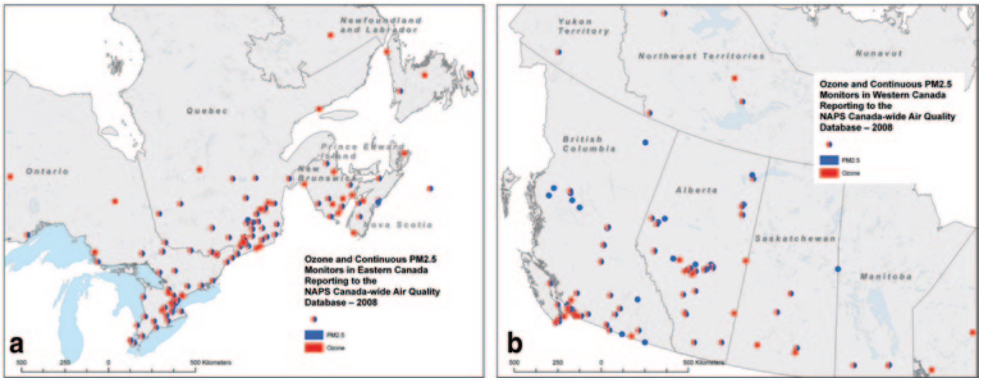
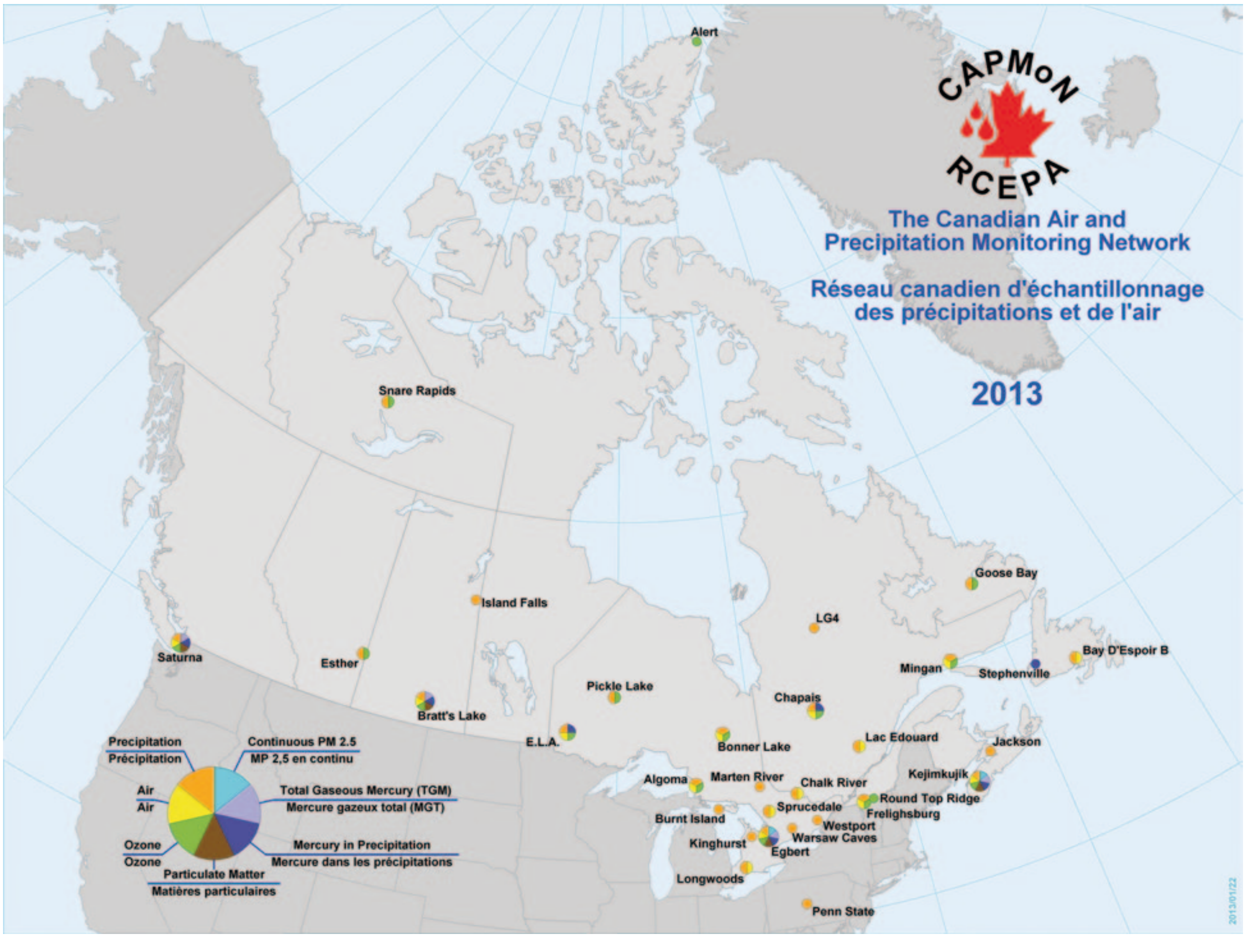


Fig. 3.1 Location of NAPS ozone and $PM_{2.5}$ monitoring sites in **a** eastern Canada in 2008, **b** western Canada in 2008

database in 2009 consisted of approximately 325 monitoring stations in over 215 communities. Although NAPS is primarily an urban monitoring network, 82 of the monitoring stations are located in rural areas. On a pollutant-by-pollutant basis there are approximately 831 continuous monitors for sulphur dioxide (SO_2), carbon monoxide (CO), nitrogen dioxide (NO_2), ozone (O_3), and particulate matter (PM), and over 113 air samplers measuring components of PM and volatile organic compounds (VOCs). Maps showing NAPS $PM_{2.5}$ and ozone monitoring sites are provided in Fig. 3.1a, b.

CAPMoN



The Canadian Air and Precipitation Monitoring Network (CAPMoN) was initially motivated by the Acid Rain issue, but has grown to include a range of other measurements. Four types of measurements are made, namely: the chemical composition of precipitation (major ions and cations), the chemical composition of atmospheric particles (both acidic and basic), particle mass ($PM_{2.5}$ and PM_{10}), and the concentration of selected gases including O_3, SO_2, nitric acid (HNO_3), $NO/NO_2/NO_y$[1], peroxyacetyl nitrate (PAN), mercury (Hg) and ammonia (NH_3). CAPMoN measurement methods include both size-selective and non-size-selective filter methods for particle composition and mass, specialized denuders, impregnated filters and continuous monitors for gases, and wet-only deposition collectors for precipitation chemistry.

The objectives of CAPMoN differ from those of NAPS in that CAPMoN measurements provide data for research into: (1) regional-scale spatial and temporal variations of air pollutants and deposition, (2) long range transport of air pollutants (including transboundary transport), (3) atmospheric processes, and chemical transport model evaluation. To meet these objectives and to best complement NAPS, the CAPMoN sites are located in rural and remote areas.

3.2.1 Measurement of $PM_{2.5}$

NAPS has been measuring the fine (<2.5 µm—$PM_{2.5}$) and coarse (2.5–10 µm—$PM_{10-2.5}$) fractions of particle mass since 1984. As of 2009 there were 27 dichotomous samplers and an additional 13 FRM $PM_{2.5}$ samplers (Table 3.1) operating in the network. Continuous or real-time particle monitoring began in the NAPS network in 1995, and the number of instruments grew rapidly with over 185 instruments reporting to the network in 2009. The use of these instruments has greatly increased the spatial and temporal coverage of the network. It is well-known that measurements obtained using the Tapered Element Oscillating Microbalance (TEOM) are biased low under certain ambient conditions due to the loss of semi-volatile chemical constituents (Allen et al. 1997; Brook and Dann 1998; Dann et al. 2006). Unless otherwise stated all $PM_{2.5}$ observations from TEOMs that are reported in this chapter have been corrected for this bias.

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[1] $NO_y = NO_z + NO_x$ and $NO_z = HNO_3 + HONO + 2N_2O_5 + HO_2NO_2 + PAN + NO_3$ + organic nitrates (not NH_3)

Table 3.2 Air toxics (Hazardous Air Pollutants) exceeding or approaching health-based guidelines in Canada

Air toxic/HAP	Volatility Class and Sampling Details
PAHs[a]	Semivolatile organic compounds (SVOCs) (gas phase and particles combined) *Samples collected for 24 h every 6, 12 or 24 days in southern Canada or for 7 days every week in the Arctic*[c] *using high-volume filter-sorbent samplers* *Samples analysed by gas chromatography/mass spectrometry or high performance liquid chromatography with fluorescence detection*
PCDD/Fs[b]	
Acrolein	Volatile organic compounds (VOCs) (gas phase only) *see VOCs in Table 3.1*
Acrylonitrile	
Benzene	
1,3-Butadiene	
Carbon tetrachloride	
Chloroform	
1,4-Dichlorobenzene	
1,2-Dichloroethane	
Dichloromethane	
Ethylene oxide	
Formaldehyde	
Naphthalene[d]	
Perchloroethylene	
Styrene	
Trichloroethylene	
Vinyl chloride	
Xylenes	
Arsenic	Metals and trace elements (particles only) *Fine and coarse particulate matter samples collected for 24 h every 3 or 6 days using dichotomous samplers* *Samples analysed by energy dispersive x-ray fluorescence and/or inductively coupled plasma/mass spectrometry*
Cadmium	
Lead	
Manganese	
Nickel	

[a] polycyclic aromatic hydrocarbons represented by benzo[a]pyrene
[b] polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans
[c] PAHs only; one sample per month analysed and remainder of samples archived
[d] naphthalene is commonly listed as a PAH but is sampled with methods used to collect VOCs due to its high volatility

Adjustment of TEOM measurements In Canada, the manual, 24-hour, filter-based, gravimetric method (Method No.: 8.06/1.0/M) has been adopted as the NAPS Reference Method (RM) for $PM_{2.5}$ measurements. Data quality objectives (DQOs) have also been established for comparison of continuous $PM_{2.5}$ instruments with the RM monitors. At a number of locations in Canada, TEOMs, Beta-Attenuation Monitors (BAMs) and TEOM-FDMS (filter dynamic measurement system) units have been co-located with RM filter-based samplers. Consequently, numerous comparisons of continuous and manual $PM_{2.5}$ measurement methods have been conducted. These show that TEOM mass measurements are lower than mass measured by the NAPS reference method due to the volatilization of semi-volatile compounds from the TEOMs. This is caused by heating of the sampled air to 30 or 40 °C in the TEOM to remove unwanted water. This loss is typically larger in the winter than in the summer because of the larger differences between the TEOM filter and ambient temperatures in winter (Environment Canada 2004). Losses also appear to occur after very high concentration events are observed, such as during biomass burning. In this case, TEOMs often report negative concentrations for a short time (i.e., a few hours) after the event while semi-volatile material that rapidly accumulated on the filter during the event evaporates. TEOMs operated at 40 °C can over-report summer $PM_{2.5}$ concentrations when the ambient dew point is higher than approximately 20 °C, because of elevated relative humidity conditions by the filter, which can enhance water uptake on the particles.

The relationship between the TEOM and the RM in the cold season is quite consistent across Canada (Environment Canada 2004). Thus, correction or adjustment equations have been developed and perform reasonably well at reducing the bias in reported average $PM_{2.5}$ concentrations. The effects of such adjustments on the observed levels are discussed in Sect. 3.5.

3.2.2 Measurements of Hazardous Air Pollutants (Air Toxics)

Table 3.2 lists air toxics that exceeded or approached health-based guidelines at Canadian ambient air concentrations in recent years (*adapted from* Galarneau and Dann 2011). These

HAPs are measured routinely in Canadian air though NAPS and some are also measured in the Great Lakes region by the Integrated Atmospheric Deposition Network (IADN) and in the Canadian Arctic through the National Contaminants Program (NCP). It is important to note that federal ambient air quality standards are not available for the vast majority of HAPs, though some provinces have devised standards applicable in their jurisdictions. Standards from the province of Ontario have been used in this chapter.

3.3 The Unperturbed Airmass Over Canada: Background O_3 and $PM_{2.5}$

The existence of a background level of O_3 in the atmosphere is well established. Background O_3 is defined as the ambient level resulting from anthropogenic and natural emissions outside North America and natural sources within North America (CCME 2006). Background O_3 concentrations are variable, with variability due to geographic location, elevation, time of year, meteorology and long-range transport influences. Geographically, background O_3 levels are higher in the northern hemisphere, with notably large enhancements observed during the spring season. Elevation is particularly important, especially at sites with exposure to the free troposphere.

Unlike most air pollutants, background O_3 can exert a significant influence on ambient O_3 levels. There are four sources which contribute to background O_3: (1) downward mixing from the stratosphere to the troposphere, (2) transport of O_3 from outside North America, (3) in-situ photochemical production from natural precursors (biogenic methane, NO_X, VOC) and (4) intercontinental transport of O_3 precursors. Much of the background O_3 affecting surface levels resides in the free troposphere. Downward transfer from the free troposphere to the surface occurs relatively frequently and is facilitated by vertical motion caused by convective and frontal activity. A combination of processes appears to contribute to this dynamic repository, including in situ O_3 production, downward fluxes from the stratosphere and intercontinental transport. There is currently some controversy around the magnitude of the influence of stratospheric O_3 on surface background levels (Lefohn et al. 2001; Fiore et al. 2003).

The spring O_3 maximum is a northern hemisphere phenomenon and is a defining feature of the annual cycle at remote and rural sites (Monks 2000). There appear to be several mechanisms responsible for the spring maximum, including UV-enhanced photochemistry in the free troposphere (Dibb et al. 2003), stratospheric-tropospheric exchange (Diem 2004; Tarasick et al. 2005) and enhanced hemispheric transport (Jaffe et al. 2010). This observation of springtime O_3 maxima is contrary to the notion that O_3 peaks during the summer, when local photochemistry is usually greatest. As shown in Fig. 3.2, Alberta sites experience March to May peaks in mean daily maximum O_3 whereas the Ontario sites experienced the highest mean daily maximum O_3 in the months of June, July or August. Assuming that the spring peak is a background feature affecting all areas, the excess ozone in Ontario for the May-September period reflects the photochemical creation of ozone in the region related to emissions from human activities. Figure 3.2 also shows that during the months of January-April and October-December mean ozone concentrations are essentially the same in the two regions and hence are related to continental background conditions.

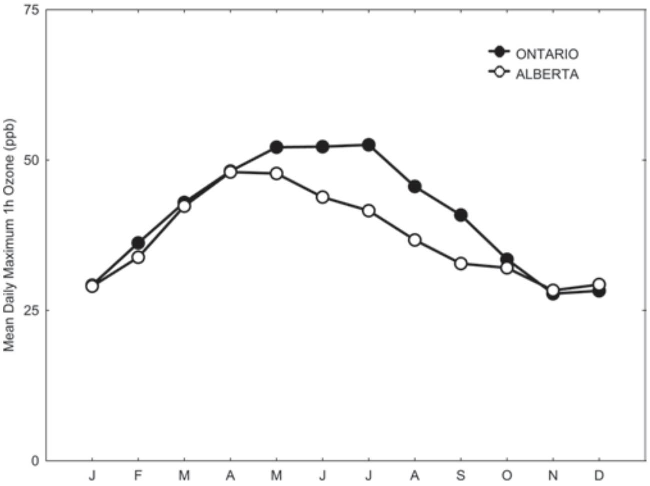


Fig. 3.2 Mean daily maximum ozone (ppb) by month (2006–2008) for southern Ontario and Alberta sites

"Background" particulate matter (PM) is defined here as PM arising from local natural (non-anthropogenic) emissions of PM and PM precursors, as well as PM and precursors transported into an airshed from afar. This long range transport PM may be both natural and anthropogenic in origin (McKendry 2006), but comes from outside North America. Examples of natural emissions of PM and its precursors include wind-blown dust, sea spray, volcanic eruptions, lightning, forest fires, wild animals and plants.

Few values of background $PM_{2.5}$ are available in the scientific literature. Background annual median $PM_{2.5}$ concentrations in Canada are both location- and time-dependent and have been estimated to range from 1.2 μg m^{-3} at a continental site in northern Alberta to 4.2 μg m^{-3} at an agricultural site near Montréal, QC (unpublished analysis of NAPS data). Of considerable relevance to Canada are estimates from McKendry (2006) who reported a mean background concentration in air masses arriving in British Columbia from north Pacific trajectories to be 1.5–2 μg m^{-3}. Lower background $PM_{2.5}$ values, generally less than 1.0 μg m^{-3} for northern portions of the U.S., were used to estimate the overall public health burden due to $PM_{2.5}$ in the U.S. (Fann et al. 2011). These were derived from air quality model simulations that excluded anthropogenic emissions as opposed to from ambient observations.

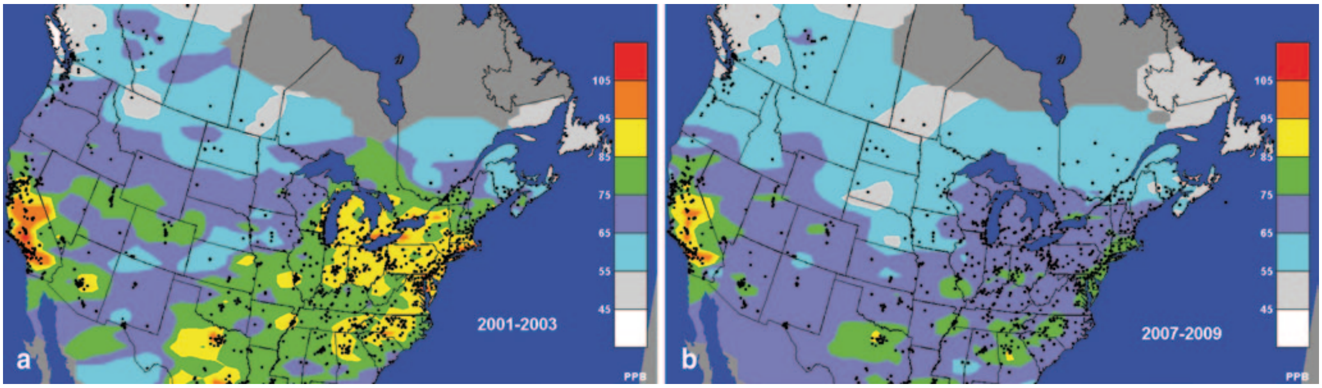


Fig. 3.3 **a**, **b** Comparison of the spatial distribution of the 3-year-average 4th highest daily maximum 8-hr ozone concentration in ppb for 2001–2003 vs. 2007–2009. The figure includes data from the United States Environmental Protection Agency (U.S. EPA) Air Quality System (AQS) database. Annual statistics for the measurement sites for these two time periods were spatially interpolated using inverse distance weighting

3.4 Observations of Ozone Concentrations Across Canada

Ozone data have been analyzed extensively throughout the past to illustrate seasonal, diurnal and day-of-the-week patterns and episodic O_3 concentrations in different regions of Canada (Fuentes and Dann 1993; Pryor et al. 1995; Environment Canada 1997). A complete analysis of ozone in terms of the Canada-wide Standard can be found in the Government of Canada Five-year Progress Report on Canada-wide Standards for Particulate Matter and Ozone (Government of Canada 2007).

For the period 2003–2005, at least 40 % of the Canadian population (approximately 13 million) lived in communities with levels above the CWS. Most of these were located in Ontario and Québec. Outside these two provinces, only one community in British Columbia and one non-urban area in Atlantic Canada had levels above the CWS. With the exception of Saskatchewan, Manitoba and the territories, all other regions had at least one location with levels within 10 % of the CWS. Provinces and territories reported data for a total of 70 communities (CMA, CA and RSA) representing more than 70 % of the Canadian population.

The spatial pattern of peak O_3 concentrations, as defined by the CWS metric, is shown for 2001–2003 and for 2007–2009 in Fig. 3.3a, b. From the earlier part of the last decade to the latter part there was a dramatic drop in O_3, including throughout the United States (U.S.). However, through 2009 southern Ontario was still experiencing levels above the CWS. The maps in Fig. 3.3a, b show that these high levels are part of a large area that encompasses the eastern U.S. These high levels are due, in part, to Ontario emissions and in part to transboundary transport of O_3 and ozone precursors from the U. S. Evidence of the impact of local Canadian sources is best exemplified by noting the small area of higher levels downwind of Toronto (i.e., to the east) in both time periods (Fig. 3.3a, b).

Compared to southern Ontario, western and eastern Canada have had lower levels of O_3, with limited evidence of CWS exceedances. Slightly elevated levels can be seen around Edmonton, Alberta and there are parts of the Lower Fraser Valley (LFV) of British Columbia and southern Atlantic Canada with locally elevated levels. Due to their location downwind of the populated areas of southwestern British Columbia and northwestern Washington, parts of the eastern LFV (e.g., Hope) may continue to be near CWS exceedance levels. However, the scale of the maps do not allow for such local maxima to be visible.

3.4.1 Temporal Variations in Ozone Concentrations

Ozone is well known to be present in higher concentrations during the warmer months and during the afternoon. In terms of seasonal behaviour, however, much of Canada is not as strongly influenced by local to regional scale sources and strong summertime photochemical O_3 production events as expected. Out of 125 sites examined for the 2001–2005 period only 46 actually experienced the highest mean daily maximum O_3 in the months of June, July or August. At the remaining sites, which are classified as being more remote rural and background sites, the maxima in O_3 concentrations occur in the springtime. All of the sites where O_3 peaks in June-August, except Hope, British Columbia, were located in southern Ontario and southern Québec. In these areas, regional-scale photochemical O_3 production and long-range transport of pollutants are relatively more important and thus, summer-time maximum O_3 concentrations are significantly higher than those in other areas of the country.

The relative frequency of days per month during 2006–2010 with eight hour average O_3 greater than 65 ppb is shown for selected geographic regions in Fig. 3.4. Only 8 sites outside of Ontario and Québec met the criteria of inclu-

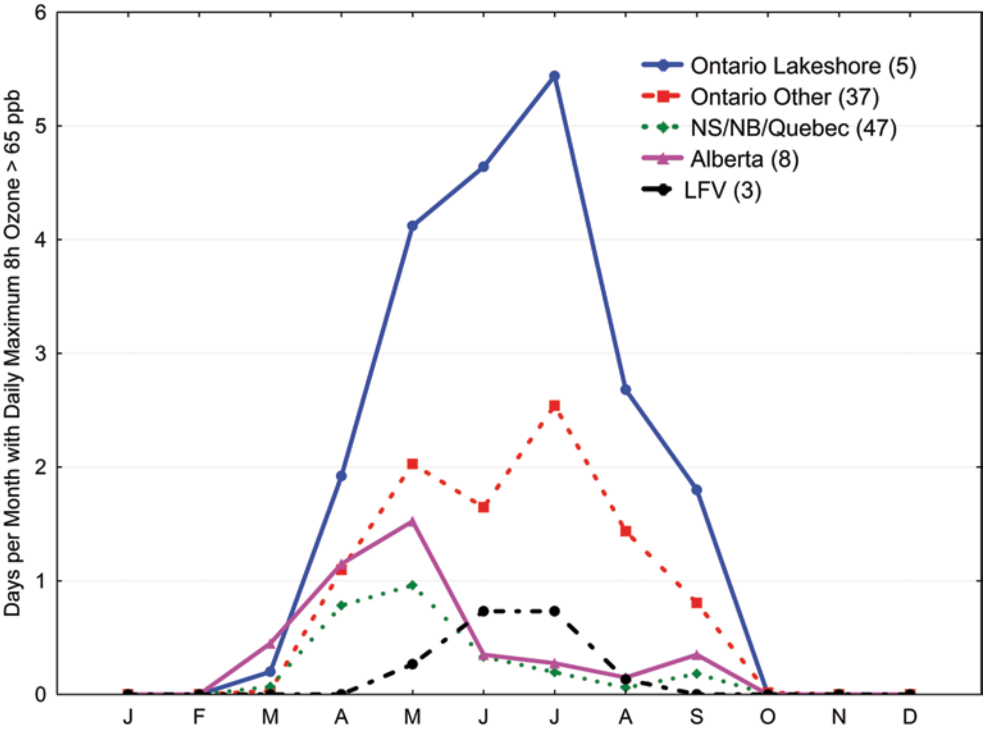


Fig. 3.4 Average days per month with ozone concentrations greater than 65 ppb for the years 2006–2010. (Only sites averaging at least one day per year > 65 ppb are included in the plot)

sion in the figure of having at least one day > 65 ppb. Sites in Ontario near the Great Lakes shorelines experienced the highest frequency of days and thus these are shown separately on the figure. It can be seen that for all areas O_3 mixing ratios greater than 65 ppb can be experienced in any of the months from March through October, but the greatest frequencies occur for June through September. The most noticeable spring peak was in the eastern part of the country (NS/NB/Québec).

Through the course of the day, greater sun intensity in the afternoon typically leads to higher O_3 levels at this time. In addition, the O_3 precursor emissions have strong temporal variations on several time scales, which may influence the diurnal pattern in O_3. For example, the main source of NO in urban areas—traffic—shows a strong diurnal cycle with a maximum generally in the daylight hours. Superimposed on this is a further modulation, especially in urban areas, related to the maximum traffic flow in the morning and evening rush hours. In addition, the diurnal variation of emissions is different on weekends to that occurring on weekdays. The number of heavy duty diesel vehicles is also substantially reduced on Saturdays and even further on Sundays. These changes in precursor emission patterns, especially during the daylight hours have led to substantial differences between weekdays and weekends in urban diurnal O_3 profiles (Environment Canada 1997; Fuentes and Dann 1993; Pryor et al. 1995). For the 2001–2005 time period 65 out of 72 urban sites in Canada experienced a "weekend effect" while 32 out of 44 rural sites showed lower O_3 or no difference on weekends—this included all rural sites in southern Ontario.

3.4.2 Trends in Ozone

During the past several decades attempts have been made to reduce O_3 by addressing precursor emissions in the U.S. and Canada from a variety of mobile and stationary sources. NARSTO (2011) recently examined North America changes in O_3 in response to these controls to assess the extent to which management actions have resulted in the intended benefits. In some places in Canada the results have been favourable, but not in all situations, due to the complexity of the atmospheric chemistry and growth in some regions. In addition, trends are difficult to detect due to the large influence of meteorology. Trends in O_3 vary markedly depending on site location (urban, rural, background) and on which statistic is being examined (Jenkin 2008; Ainslie and Steyn 2007).

Year to year changes in daily maximum 8h O_3 (April to September only) at different percentile levels (10th, 25th, 50th, 75th, 95th and maximum) are shown for a group of urban and rural sites for the period 1990–2010 in Figs. 3.5 and 3.6, respectively. The urban sites shown are distributed across Canada but the rural sites are primarily located in Ontario and Québec with 3 sites in Atlantic Canada and 2 sites in British Columbia. Trends in the higher end of the O_3 distribution tends to reflect the impact from precursor emissions at closer distances to the measurement sites, while the average or lower portions of the distribution generally represent the background O_3 (Lelieveld et al. 2004).

Figure 3.5 shows trend results for the urban sites with a statistically significant (at the 95th confidence level) positive trend for the 10th and 25th percentiles indicating a rise in urban background levels. These results illustrate the effect

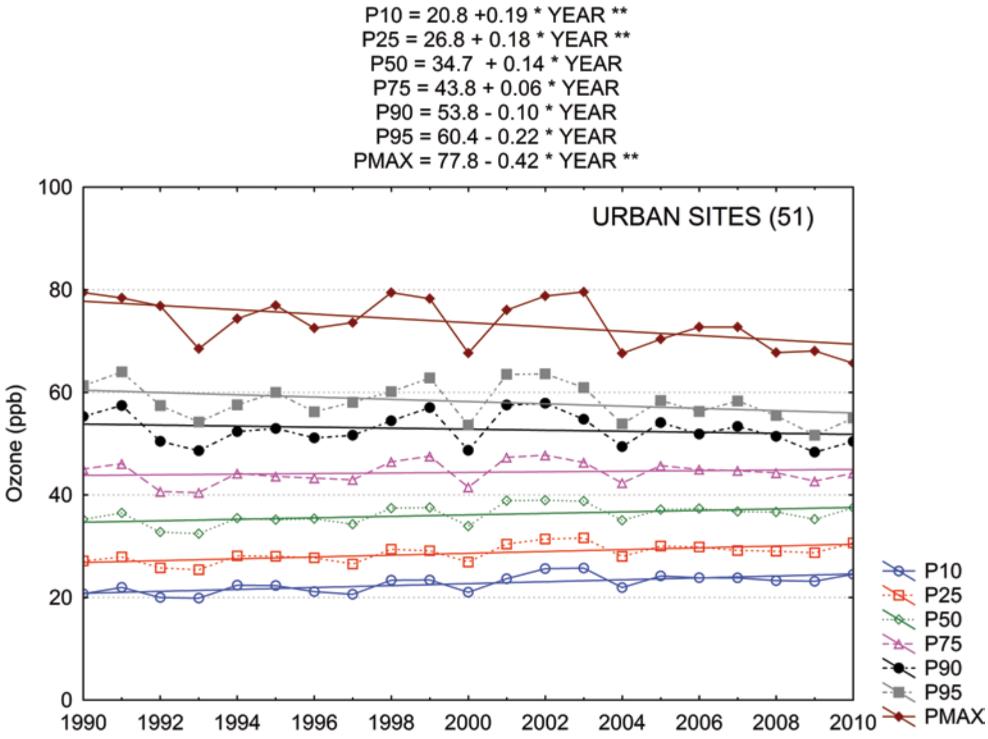


Fig. 3.5 Trend in daily maximum 8 h ozone 10th, 25th, 50th, 75th, 90th, 95th percentiles and maximum for Canadian urban sites for 1990–2010. Regression lines marked with ** are statistically significant at the 95th percentile confidence level

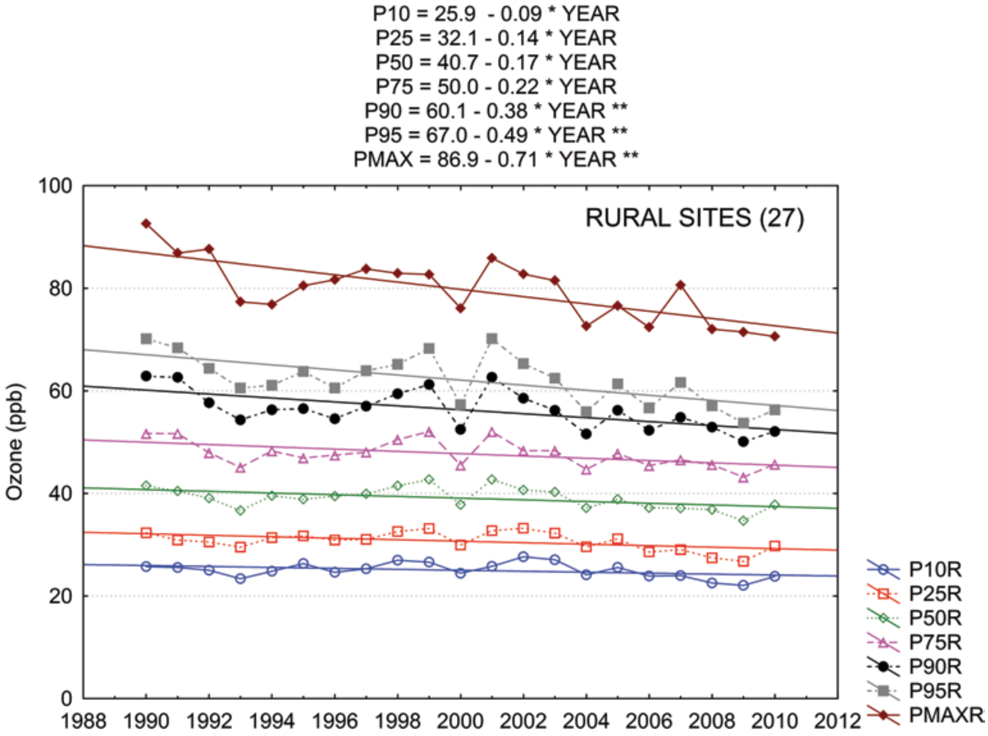


Fig. 3.6 Trend in daily maximum 8 h ozone 10th, 25th, 50th, 75th, 90th, 95th percentiles and maximum for Canadian rural sites for 1990–2010. Regression lines marked with ** are statistically significant at the 95th percentile confidence level

of local NO reductions in reducing O_3 scavenging leading to a net upward trend in lower percentile and average ozone concentrations. In contrast, a significant negative slope was detected for the maximum urban O_3 values, while for the 90th and 95th percentiles there were non-significant decreases. This behaviour suggests that O_3 arising from emission sources that are more local to Canadian cities has decreased during the past 20 years.

For the rural sites, Fig. 3.6 shows that there have been decreases (i.e., negative slopes) throughout the O_3 distribution, but only the maximum, 95th and 90th percentiles of 8 h O_3 had statistically significant decreases. Thus, region-

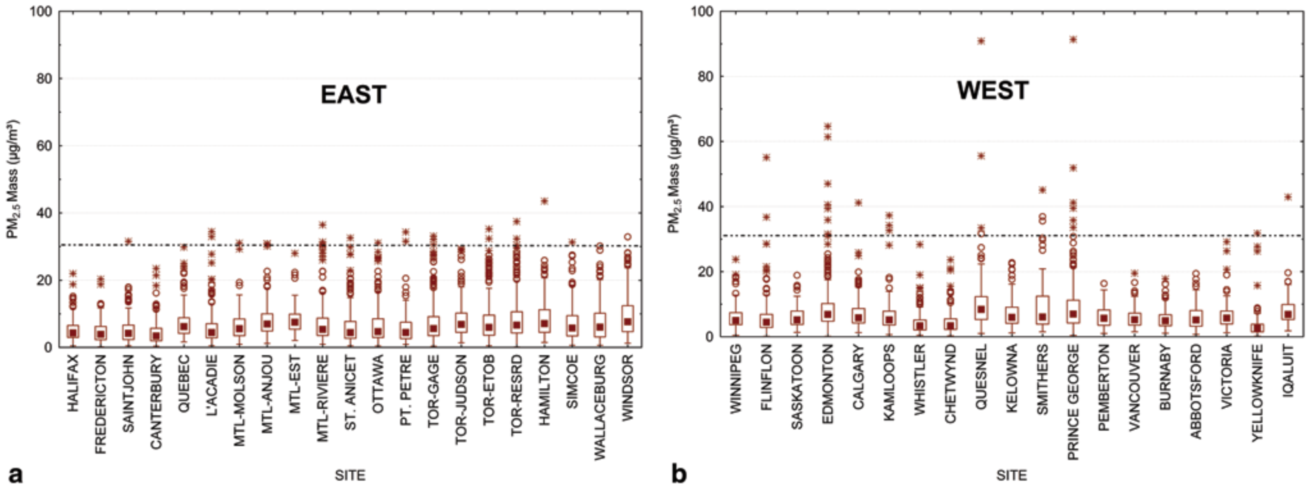


Fig. 3.7 Comparison of 24 h $PM_{2.5}$ mass concentrations from filter-based samplers: 2008–2010. The statistics provided are the median, 2nd, 25th, 75th and 98th percentiles and outliers and extremes. Only sites with a minimum of 100 samples collected over this period are included and the sites are arranged from eastern Canada to the west

al scale O_3 has responded to the past emission reductions implemented in Canada and the U.S. Importantly, there is no evidence from this group of rural sites (predominantly in Ontario and Québec) that there has been an increase in background O_3.

3.5 Observations of $PM_{2.5}$ Concentrations Across Canada

3.5.1 $PM_{2.5}$ Mass

An analysis of $PM_{2.5}$ in terms of the Canada-wide standard can be found in the Government of Canada Five-year Progress Report on Canada-wide Standards for Particulate Matter and Ozone (Government of Canada 2007). For the period 2003–2005, at least 30% of the Canadian population (approximately 10 million) lived in communities with levels above the CWS. Most of these were located in Ontario and Québec. Outside these two provinces, only two communities in the interior of British Columbia reported levels above the CWS. Communities within 10% of the standard were also primarily located in Ontario and Québec.

Figure 3.7 provides a box-plot comparison of 24 h $PM_{2.5}$ concentrations across the country for more recent years: 2008 through 2010. The sites shown represent all the available, routinely operating filter-based samplers. As seen for 2008–2010, there were more sites with 24 h observations greater than 30 µg m $^{-3}$ in eastern Canada compared to the west. However, there continue to be locations in the interior of BC (e.g., Prince George) with relatively high means and peak 24 h concentrations. Edmonton is also seen to have experienced a number of days above the 30 µg m^{-3} threshold.

Similar to O_3, the conditions over southern Ontario and southern Québec are part of a high concentration Canada-U.S. airshed. Throughout this large area, annual mean $PM_{2.5}$ exceeded 8 µg m^{-3} in 2004–2006. In central British Columbia, where there are also high concentrations, it is likely that these levels were confined to the more populated valleys where a range of local industries, residential wood smoke and traffic make more significant contributions. The winter months are generally when these problems occur due to stronger valley inversions, which trap the local emissions, and due to a greater need for supplemental heating by wood-burning appliances.

As Table 3.1 indicates there are many more locations in Canada reporting hourly $PM_{2.5}$ from continuous samplers such as the TEOM. These data are valuable for enhancing the density of the network such that it is more comparable to O_3 monitoring and thus, provides greater population coverage. However, as indicated above, there are potential problems with some of the available instruments. To demonstrate the potential importance of the TEOM instrument's loss of volatile mass, Fig. 3.8 provides the annual 98th percentile $PM_{2.5}$ values for a selection of TEOM trend (at least 8 out of 10 years of data) sites based upon adjusted (transformed) and unadjusted $PM_{2.5}$ concentrations. Values are 2–5 µg m^{-3} higher after adjustment, although both measures show that during the 2000–2009 period there was a net decrease in concentration. As another example, adjusting $PM_{2.5}$ values among all the available TEOM sites across the country among the measurements taken during 2004–2006 increased the number of sites surpassing the CWS by 27. Clearly, in order to obtain a proper understanding of the current state of $PM_{2.5}$ levels over as much of Canada as possible it critical that the continuous monitors, which are necessary to provide daily sampling frequency, produce measurements that are consistent with the reference method.

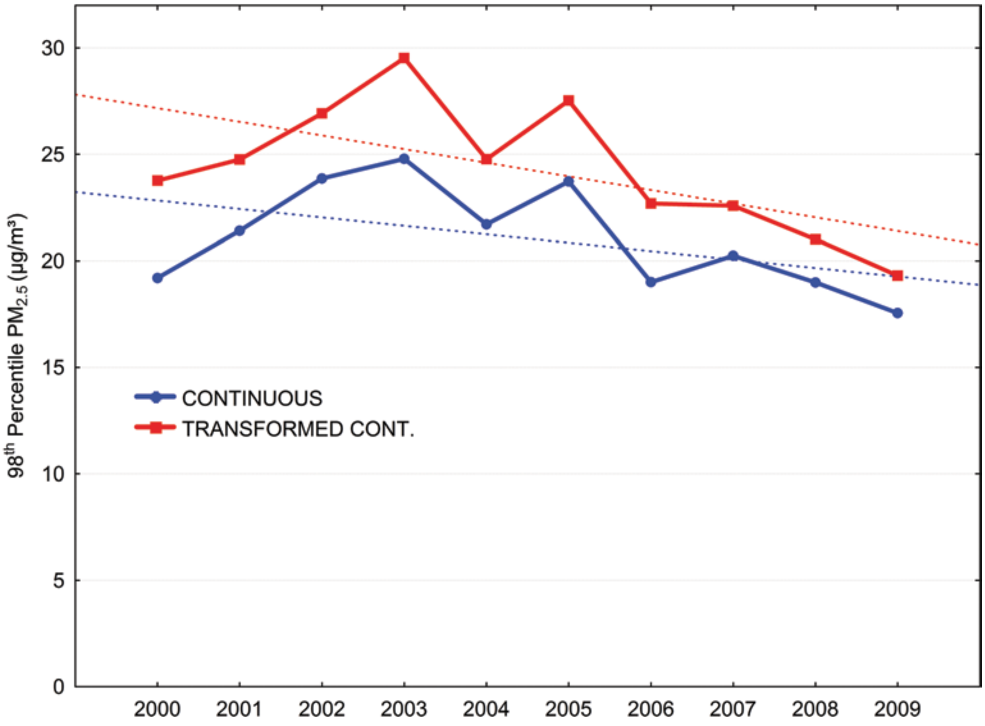


Fig. 3.8 Yearly variation of 98th percentile $PM_{2.5}$ for national TEOM trend sites 2000–2009. Adjusted and unadjusted concentrations are shown to provide an indication of the amount of mass that is estimated to be lost due to the sample heating inside the TEOM instrument

Current Status of Continuous $PM_{2.5}$ Measurement Across Canada Due to reasons described above the TEOM continuous $PM_{2.5}$ instruments deployed across the Canadian monitoring networks since the mid-1990's do not meet the data quality objectives (DQOs) that have been established for comparison with NAPS reference method samplers (Allen 2010). However, NAPS managers have determined that instruments approved as U.S. EPA Class III federal equivalency methods (FEM) do meet the NAPS DQOs. Thus, monitoring agencies across Canada have either transitioned to or are in the process of transitioning to $PM_{2.5}$ FEM instruments in their monitoring networks (Allen 2010).

There are primarily three makes of Class III FEM instruments currently being deployed across Canada: the Met One BAM1020; the Thermo TEOM 1400ab-FDMS (to be discontinued); and the Thermo 5030 Sharp. The U.S. EPA conducted an assessment of $PM_{2.5}$ FEMs compared to collocated federal reference method (FRM) samplers. For this analysis, 61 Met One BAM stations and 17 Thermo 8500 FDMS stations were examined. Based on these comparisons it was found that both FEMs predicted annual means similar to the FRM or higher by 2–5 µg m^{-3}. The bias for both instruments appeared to be highest for the spring/summer seasons (U.S. EPA 2011). The Canadian Council of Ministers of the Environment (CCME) has recommended that all jurisdictions strive to deploy new Class III FEM instruments of their choice by December 31, 2012.

Ultimately with greater geographic coverage of $PM_{2.5}$ monitoring using consistent daily measurement techniques (i.e., RM or FEM) it may be possible to generate $PM_{2.5}$ maps such as shown in Fig. 3.3a, b for O_3. However, the current network does not have the density of monitors for this purpose and differences in measurement techniques among the available sites add to the problem. In addition, $PM_{2.5}$ differs from O_3 in that there can be significant primary emissions of $PM_{2.5}$. This can lead to greater uncertainty in the interpolation necessary to produce maps, particularly in areas of significant local sources and limited monitoring.

Satellite-based estimates of surface $PM_{2.5}$ concentrations are proving to be useful in filing the surface monitoring gap (van Donkelaar et al. 2010), but limitations remain (Hoff and Christopher 2009). This includes the limited temporal coverage, lost observations due to clouds and the columnar nature of satellite observations. Nonetheless, satellite-derived data represent an important emerging technology for air pollutant monitoring. The national level exposure assessment used by Crouse et al. (2012) to successfully quantify the effects on mortality rates of long term $PM_{2.5}$ exposure across Canada relied on observations derived from satellite. Lee et al. (2011) showed what level of information on surface NO_2 patterns over southern Ontario could be derived by combining satellite and routine monitoring data and recently McLinden et al. (2012) used satellite data to study NO_2 and SO_2 patterns over the oil sands region of Alberta.

3.5.2 Temporal Variations in $PM_{2.5}$ Concentrations

Unlike O_3, which has a strong seasonality, $PM_{2.5}$ varies less through the year. Winter and summer levels of $PM_{2.5}$ mass

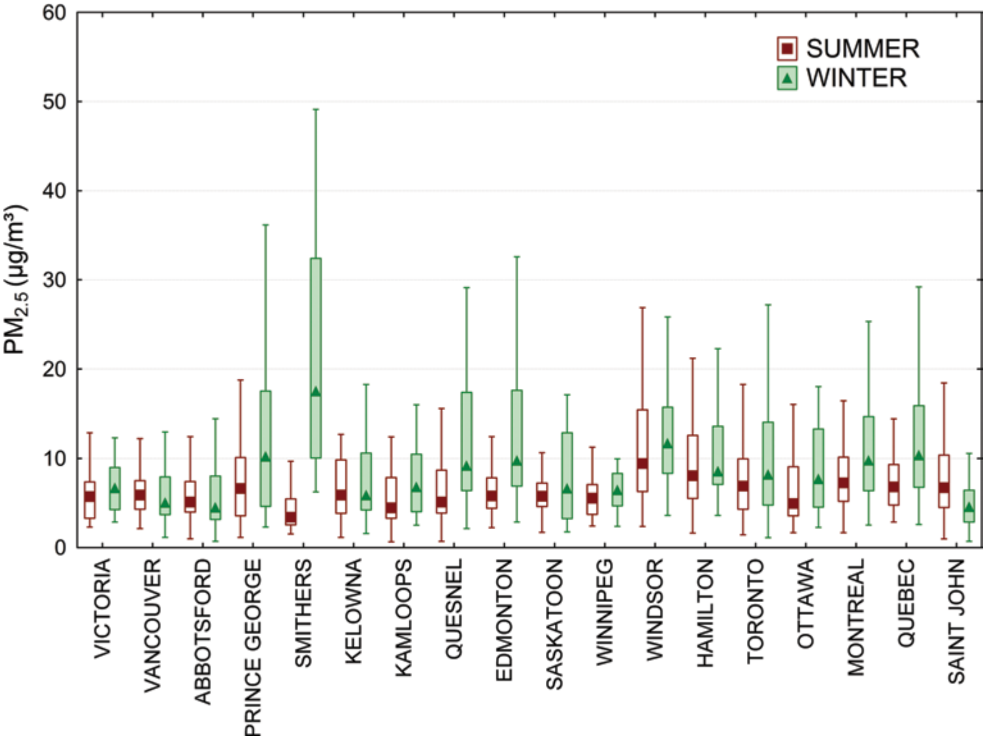


Fig. 3.9 Winter (December, January, February) vs. summer (June, July, August) concentrations of $PM_{2.5}$ mass (2007–2009)—filter-based samplers for selected urban areas (median, 25th and 75th percentile, non-outlier maximum and minimum). The time periods correspond to those in Fig. 3.12

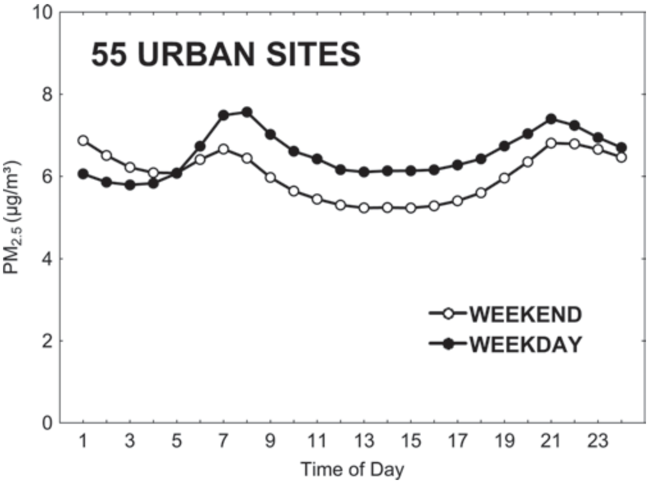


Fig. 3.10 Diurnal variation in $PM_{2.5}$ mass—weekday vs. weekend—TEOM samplers (2007–2009)

are compared for 2008–2010 in Fig. 3.9. Overall there were more sites with higher winter means and peaks.

Using composite hourly data from the TEOM network the diurnal variability of $PM_{2.5}$ mass for weekdays versus weekends is illustrated in Fig. 3.10. This figure clearly shows the impact of transportation sources on $PM_{2.5}$ concentrations with an approximate 25 % decrease in the morning peak and a 17 % decrease in the daily mean on weekends as compared to weekdays.

3.5.3 Recent Trends in $PM_{2.5}$

The lack of long-term $PM_{2.5}$ mass data at rural/remote monitoring sites in Canada precludes any analyses of long-term trends outside of cities. Figure 3.11 shows how the annual mean $PM_{2.5}$ mass composited across multiple urban dichotomous sampler sites changed during the last decade. In eastern Canada the peak year was 2002 with a level of 13 µg m^{-3} and since then $PM_{2.5}$ decreased to below 9 µg m^{-3}, on average. During this same ten year period the $PM_{2.5}$ also decreased among the western Canadian sites, but only by 2 µg m^{-3}. As a result, in 2008 the mean was 6 µg m^{-3} and thus, the difference between the east and west urban areas has diminished.

Also shown in Fig. 3.11 is the annual mean sulphate concentration (SO_4^{2-}). This $PM_{2.5}$ constituent is one of the main contributors to mass (see next section) and during the past 20 years there have been considerable North American reductions in the emissions of SO_2 in order to lower acid deposition rates and vehicular emissions of $PM_{2.5}$. In eastern Canada sulphate was variable from 1999 to 2005 at around 11 µg m^{-3}. However, decreases in the last three years have led to a composite average concentration of 2.2 µg m^{-3} in 2008. The rate of reduction of $PM_{2.5}$ during this time period was very similar to the rate observed for sulphate suggesting that the SO_2 controls had a beneficial impact on $PM_{2.5}$ in the east. Sulphate was a smaller fraction of the $PM_{2.5}$ in the west and its concentration has remained steady at about 1 µg m^{-3} from 2001 to 2008.

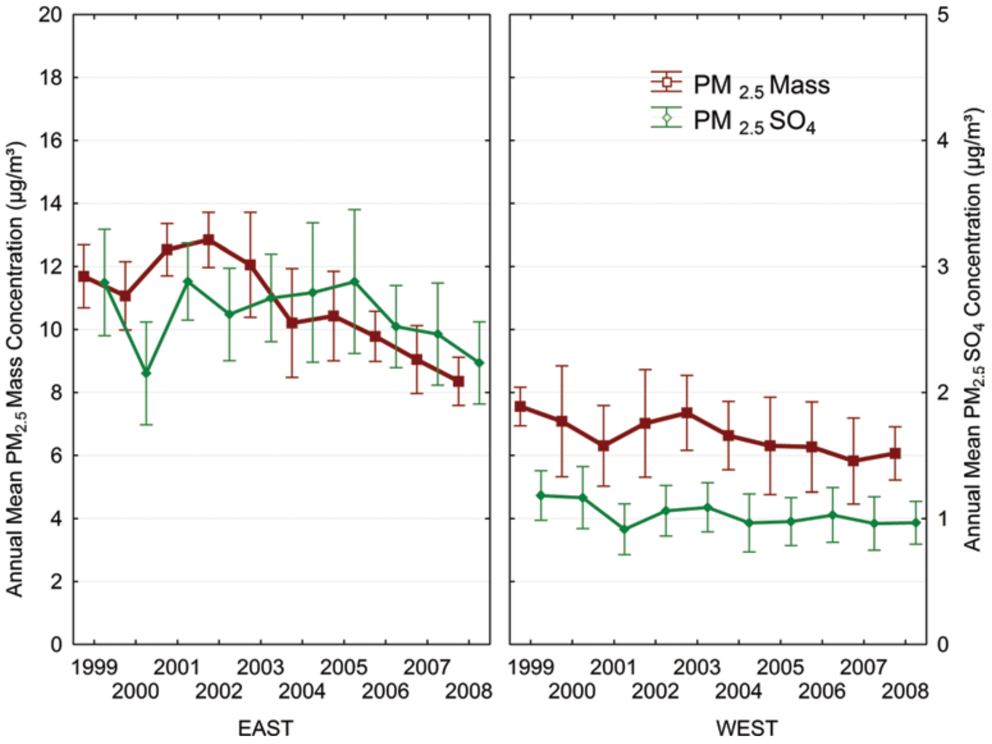


Fig. 3.11 Yearly variation in mean $PM_{2.5}$ mass and sulphate (SO_4) from filter based network urban trend sites east and west of the Ontario/Manitoba border (1999–2008). Composite means and the 90th percent confidence interval around the mean are plotted

3.5.4 Chemical Composition of $PM_{2.5}$

$PM_{2.5}$ is a complex mixture of inorganic and organic chemical compounds. The main contributors to the mass are the ions of sulphate, nitrate and ammonium and organic carbon (OC), which consists of thousands of different species and not all have been identified. Another important component of the $PM_{2.5}$ is elemental carbon (EC), which is often referred to as black carbon (BC). Here we use EC because of the approach to determine the concentration reported, which is based upon the thermal-optical approach (Chow et al. 1993). A small, but consistent contribution to $PM_{2.5}$ is also made by a large number of trace elements or metals such as iron, aluminum, silicon, calcium, zinc, manganese, titanium, nickel and arsenic.

Sulphate and major elements of $PM_{2.5}$ mass from the dichotomous sampler network have been determined since 1986. These data have been previously described (Brook et al. 1997, 1999; Brook and Dann 1999). Monitoring of complete fine particle speciation began at Canadian network sites in 2000 (Lee et al. 2003). The number of sites started to grow beyond Toronto and Burnaby South (greater Vancouver area) in 2003. In this speciation network, which currently consists of 11 sites (Table 3.1), the major components of $PM_{2.5}$ are reported. These measurements were recently discussed by Dabek-Zlotorzynska et al. (2011), including the seasonal variation in each of the major chemical species making up the $PM_{2.5}$.

Complete fine particle speciation allows the mass to be reconstructed from the chemical species. Dabek-Zlotorzynska et al. (2011) described the reconstruction approach used for the Canadian $PM_{2.5}$ network. This involves grouping the chemical composition measurements according to ammonium sulphate ($(NH_4)_2SO_4$ or ASO_4), ammonium nitrate (NH_4NO_3 or ANO_3)), organic matter (OM), which is taken as 1.6 × OC at urban sites and 1.8 × OC at rural sites, elemental carbon (EC), crustal material and other oxidized metals (SOIL) and sodium chloride (NaCl). Particle-bound water (PBW) can also be estimated from the measurements. Figure 3.12a, b shows the reconstructed mass for the warm (April to September) and cold (October to March) seasons of 2007–2009.

On average the reconstructed mass was in agreement with the measured mass. Combined $(NH_4)_2SO_4$ and NH_4NO_3 account for 32–43 % of total $PM_{2.5}$ mass at the eastern sites during the summer and 36–59 % in the winter. Thus, secondary compounds (i.e., those that form in the atmosphere from emissions of gaseous precursors) are a major contributor. For the western sites $(NH_4)_2SO_4$ and NH_4NO_3 account for 18–30 % of mass in summer and 22–45 % in winter, also indicating that gaseous precursor emissions are quite important to $PM_{2.5}$. Figure 3.12a, b also shows that the relative amounts of $(NH_4)_2SO_4$ and NH_4NO_3 change from summer to winter. Nitrate species increase dramatically at most locations with the highest concentrations in Edmonton and Windsor.

At the eastern sites OM is the next most important contributor to mass after the two inorganic secondary species. In contrast, in Edmonton and at the British Columbia sites, OM contributes the most. OM contributions to $PM_{2.5}$ mass for urban and rural sites range from 31 to 52 % in summer and 23 to 58 % in winter. Interior BC locations, such as Quesnel

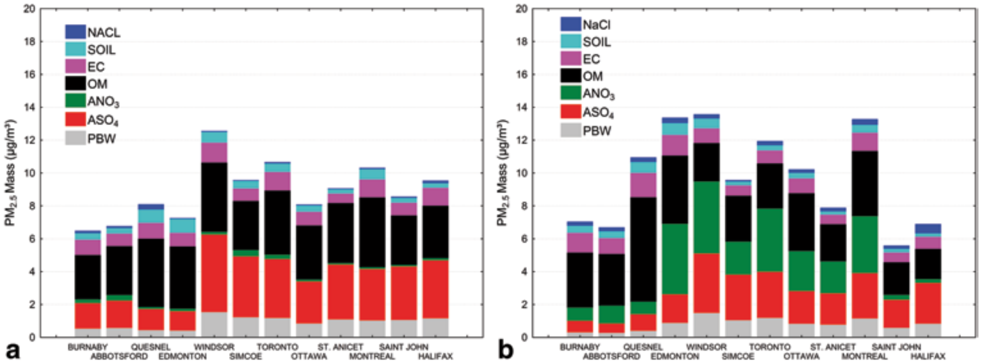


Fig. 3.12 **a**, **b** Reconstructed $PM_{2.5}$ mass by major component and site for (a) June, July, August (2007–2009) and (b) December, January, February, March (2007–2009)

and Golden (Dabek-Zlotorzynska et al. 2011), have the largest OM percentages, particularly in the winter. Wood combustion, either from industry and residential heating, as well as from wildfires, is the key contributor in these areas. The fraction of OM that is due to secondary formation, originating from gas phase VOC emissions, has not been accurately quantified. On average, it is suspected to be a significant fraction and also increases away from populated areas and/or as the aerosol age in the atmosphere. Both anthropogenic and biogenic emissions of VOCs are important contributors to the secondary OM (e.g., de Gouw et al. 2008; Slowik et al. 2010, 2011; Liggio et al. 2010).

The average $PM_{2.5}$ chemical composition in Fig. 3.12a, b changes during high mass concentration episodes due to greater contributions from certain sources or atmospheric formation processes. For the ten highest days in summer, $PM_{2.5}$ mass at eastern sites is primarily composed of $(NH_4)2SO_4$ and OM (70–80 %), while at the western sites other than Golden, NH_4NO_3 is also an important contributor. For the highest concentration days in winter NH_4NO_3 and OM are the primary contributors to mass at almost all the sites, but $(NH_4)2SO_4$ is also an important contributor at the eastern sites and in particular in Halifax.

Among major cities, trace metals (four selected toxic elements) have the highest concentrations in Halifax. However, Fig. 3.13 shows that among the locations monitored these metals were by far the highest in the small, base metal smelting community of Flin Flon, Manitoba, where arsenic was very high during the monitoring period (2007–2009). In terms of cities, these four metals are also relatively high in Montreal, followed by Windsor, Saint John and Burnaby. Both the east and west coast and Montreal have high nickel and vanadium levels. These are due to use of oil for power and heating (Halifax), heating (Montreal) and due to ship traffic (all four locations).

3.6 Elevated $PM_{2.5}$ and O_3 (SMOG) Events

Smog episodes are an important feature of Canadian air quality because the response by the government is to issue an air quality advisory. This leads to greater public awareness, affects their perception of the problem and focuses attention on the actions needed to avoid such events. High O_3 and $PM_{2.5}$ events often occur during multi-day, regional scale episodes, particularly in eastern Canada. The meteorological conditions contributing to pollutant build up may become established over Ontario and then propagate eastward. However, such episodes are relatively uncommon over Atlantic Canada unless the air flow is such that pollutants that have built up over Ontario, the U.S. Midwest and particularly the eastern seaboard of the U.S. are transported to the region to combine with local emissions.

Defining an episode is subjective in that to generate frequency statistics a threshold concentration needs to be defined. The appropriate threshold is a matter of perception and ideally should be set relative to the prevailing conditions in each region of the country. For example, the threshold level for high $PM_{2.5}$ in Ontario is well above the level that noticeably impacts visibility in the Lower Fraser Valley. In this chapter the CWS metrics for $PM_{2.5}$ and O_3 are used to identify episodes, but again, this may misrepresent how local populations perceive the occurrence of episodes. Thus, there is a need for local authorities and stakeholders to determine their own threshold to best meet their needs of public awareness and health and welfare protection.

Historically the greatest frequency of regional-scale episodes $PM_{2.5}$ have occurred in Ontario followed by Québec and for both these regions high $PM_{2.5}$ values often persisted for several days. Regional scale episodes of $PM_{2.5}$ were infrequent in the Prairies with the most notable regional scale $PM_{2.5}$ event associated with the August 2003 forest fires in the southern British Columbia interior. The relative frequency of days per month with $PM_{2.5}$ concentrations greater than 30 µg m^{-3} is shown in Fig. 3.14 for the five year period 2005–2009. $PM_{2.5}$ concentrations greater than 30 µg m^{-3} occurred in all months of the year. However, in British Columbia these events were largely a cold season phenomenon and were most often due to local emissions being trapped under inversions in the interior valleys. A similar seasonal behaviour occurred in Alberta with strong inversions limiting vertical mixing over large emission areas (e. g., cities) being responsible. The summertime occurrences of $PM_{2.5}$ episodes in western Canada were usually associated with forest fires

Fig. 3.13 Mean concentrations (ng m^{-3}) of vanadium, nickel, arsenic and selenium at NAPS sites (2007–2009). Measurements are of water soluble $PM_{2.5}$ by ICP-MS

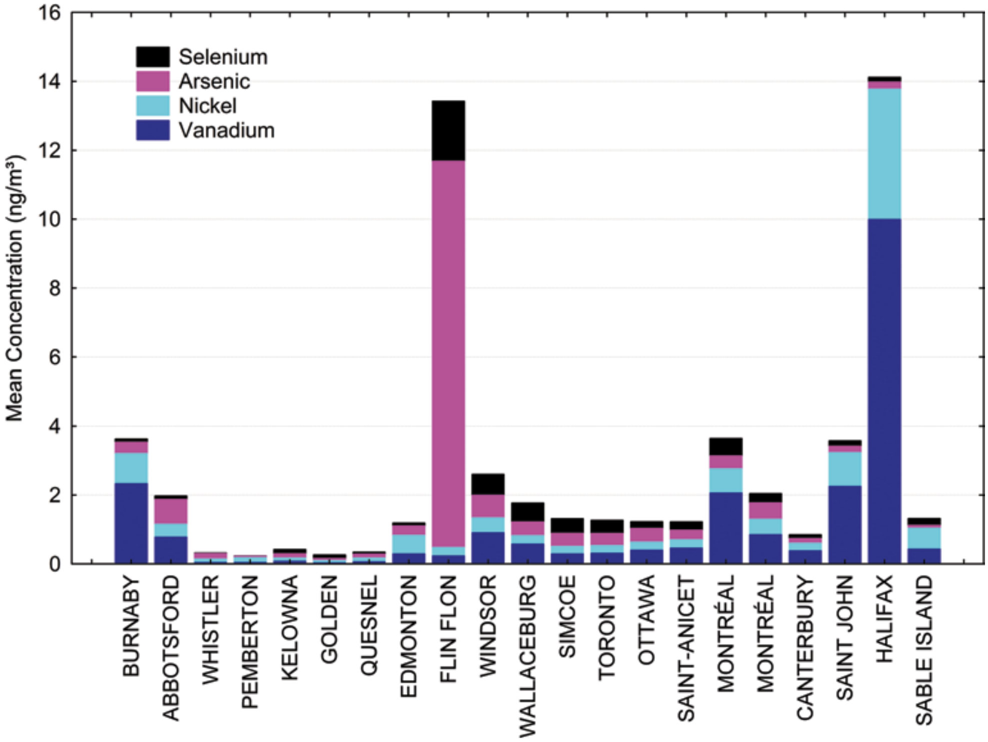
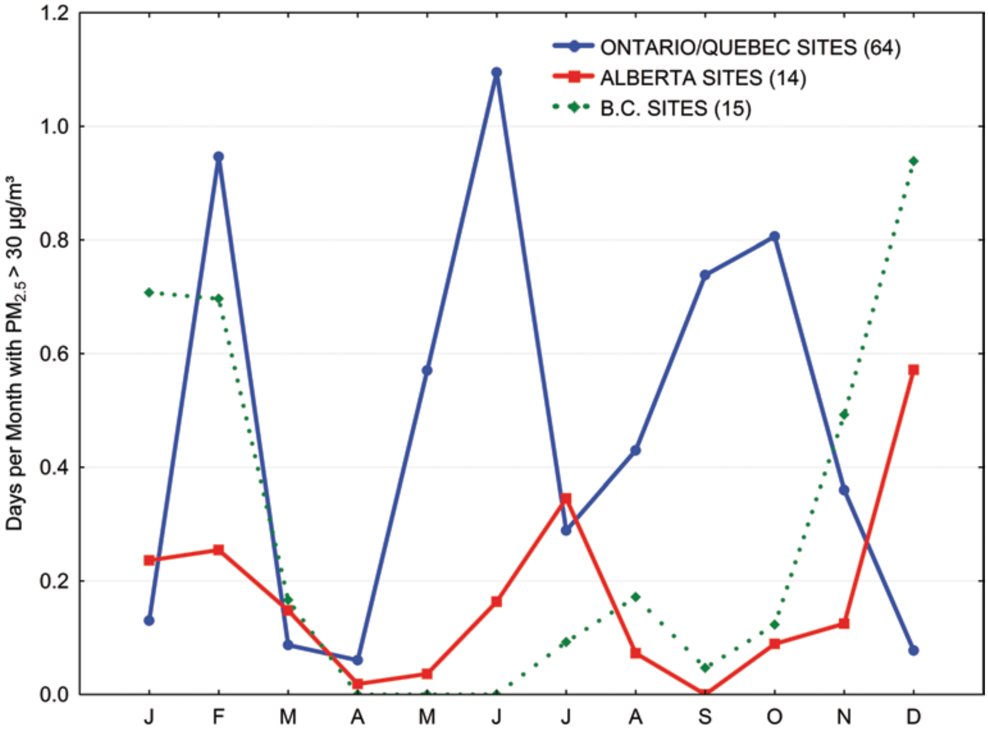


Fig. 3.14 Average days per month with $PM_{2.5}$ concentrations greater than 30 µg m^{-3} for the years 2005–2009. Results have been adjusted for cold-season TEOM mass loss. Only sites averaging at least one day per year >30 µg m^{-3} were used to prepare this graph, which resulted in no days in Atlantic Canada, Manitoba and Saskatchewan. The numbers of sites included are shown in brackets in the legend



(e.g., August 2003). As expected, Ontario/Québec sites experienced the greatest overall frequency of days >30 µg m^{-3} with the peak month being June followed by February and October. Thus, a $PM_{2.5}$ episode can occur in any season.

The greatest frequency of regional-scale episodes of O_3 also has occurred in Ontario followed by Québec. For both these regions high ozone values often persisted for several days and were highly likely to be associated with $PM_{2.5}$ values greater than the CWS metric of 30 µg m^{-3}. This situation has changed over the past ten years, however, and to illustrate this Table 3.3 provides a comparison of a regional scale episode that occurred in Ontario in 2003 contrasted with one that occurred in 2010.

Table 3.3 Comparison of a Regional Scale Ozone and $PM_{2.5}$ Episode in Ontario for two heat-wave periods in 2003 and 2010. (A regional scale episode is defined as days where 33 % of monitoring sites in a region record maximum daily 8 h ozone greater than 65 ppb)

Episode date	Total reporting sites (O_3)	Sites with 8 h $O_3 > 65$ ppb	Max. 8 h O_3 (ppb)	Sites with $PM_{2.5} > 30$ µg/m³	Max 24 h $PM_{2.5}$ (µg/m³)	Max. temp Toronto (°C)	Max. temp Ottawa (°C)
24-Jun-2003	56	36	112	8	33	32.6	32.5
25-Jun-2003	56	36	123	28	46	34.2	33.0
26-Jun-2003	56	35	108	28	55	33.3	32.3
05-Jul-2010	50	29	82	1	32	33.8	33.7
06-Jul-2010	50	32	99	1	31	33.1	33.6
07-Jul-2010	50	26	89	3	33	32.0	33.8
08-Jul-2010	50	26	97	2	38	33.3	34.5

Environment Canada defines a heat wave as three consecutive days when the maximum temperature is 32 °C or higher. For Ottawa there was a seven year time gap between heat waves that occurred in June 2003 and July 2010. It is these two heat wave periods that are compared in Table 3.3. As shown in the table, a large number of Ontario sites exceeded the 65 ppb threshold for 8 h O_3 during both episodes. However the maximum O_3 values were 20–30 ppb lower during the 2010 episode. More striking is the reduction in sites that exceeded 30 µg m^{-3} of $PM_{2.5}$ during the two episodes (despite the same number of reporting sites) with over 25 sites exceeding this value in 2003 and only 1–3 in 2010. Maximum $PM_{2.5}$ concentrations were 15–20 µg m^{-3} lower in 2010.

3.7 Observations of Other Routinely Monitored Air Pollutants

Levels of sulphur dioxide (SO_2), nitrogen dioxide (NO_2), nitric oxide (NO), carbon monoxide (CO) and volatile organic compounds (VOCs) have been monitored at several locations across Canada for many years (>20 years). At high levels each of these pollutants can have human health and/or environmental impacts. Consequently ambient air quality standards or objectives have been in place for many years to encourage reductions in concentrations and in recent years their levels rarely approach the objective values. However, these pollutants are reactive, contributing to the formation of O_3 and/or $PM_{2.5}$, and thus tracking and minimizing their concentrations continues to be important to ensure good air quality.

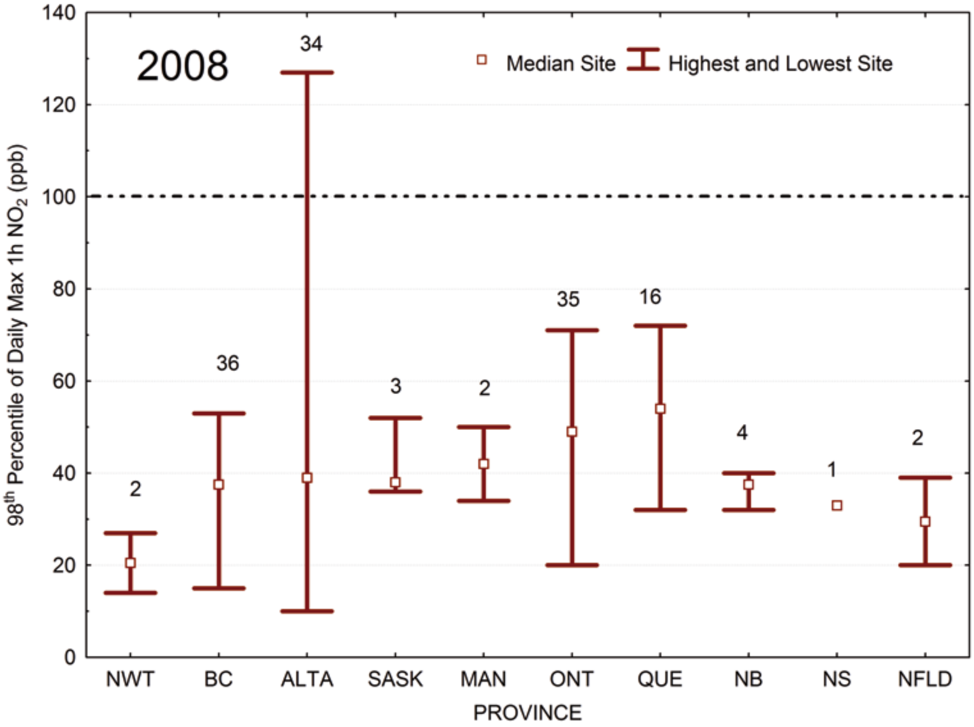
Measurement of NO_2 Often NO and NO_2 are added together and reported as NO_x. Due to the measurement technique used to determine NO_2 at NAPS sites the concentration reported can experience a small interference from other oxidized nitrogen compounds, namely nitric acid, peroxyacetyl nitrate, organic nitrates and particle nitrate. When these species make it through the inlet of the measurement system and are converted to NO before being detected they cause an overestimation of NO_2 (and NO_X). The magnitude of the overestimation is dependent on the extent of photochemical processing of these other nitrogen species during the course of atmospheric transport to the measurement site. In general, the overestimate of NO_2 increases as the distance from emission sources increases.

The magnitude of this positive interference has been estimated at a few rural locations in Canada as part of CAPMoN research (NARSTO 2011) and have also been discussed by Lee et al. (2011) for southwestern Ontario. At a rural site ~75 km northwest of Toronto (Egbert) the interference ranges from approximately 4 % in the winter to 39 % during summer periods of elevated photochemical activity. It should be noted that most Canadian NO_X measurement sites are located in urban environments which are impacted mostly by local emissions. Thus the positive interference seen at the rural Egbert location is expected to be closer to the worst case scenario for most NAPS sites.

Volatile organic compounds, or VOCs, are generally defined as compounds containing at least one carbon atom (excluding carbon dioxide and carbon monoxide) and with a vapour pressure of 0.01 kPa or greater at 25 °C. Although there are many thousands of organic compounds in the natural and polluted troposphere that meet the definition of a VOC, most measurement programs have concentrated on the 50 to 150 most abundant C2 to C12 hydrocarbons consisting of the general formula CxHy and on C2 to C6 carbonyls (compounds that contain the structural element R2C = O). In this chapter, total non-methane hydrocarbons (total NMHC) are defined as the sum of all identified C2 to C12 hydrocarbons. Total VOCs are defined as total NMHC plus carbonyls and other polar species. NMHC also includes a number of species emitted from biogenic sources including isoprene, α-pinene, β-pinene, δ-limonene and camphene.

VOC Reactivity Ozone formation includes a complex array of reactions involving the atmospheric oxidation of VOCs. In this process, individual VOCs differ in their efficiency towards O_3 formation. Therefore, a scale in which each compound is ranked according to its potential to form O_3 can help guide certain emission regulations. The use of the rate coefficient for the reaction of OH + VOC (kOH) as a measure of the reactivity of a VOC is one common ranking approach. While this approach does not take into account the complex chemical reactions that follow the initial reaction between

Fig. 3.15 Provincial comparison of 98th percentile of daily maximum 1 h NO_2 concentrations (ppb) for 2008. The plotting point represents the median of all sites and the whiskers represent highest and lowest site. The number of reporting sites in each province is also provided



the VOC and OH, it is useful to consider the *k*OH reactivity scale under conditions where the production of O_3 is largely limited by the supply of NO_X rather than VOCs. These sort of conditions prevail at locations removed from major source areas of VOCs, e.g., in rural and remote areas. To help with this limitation Chameides et al. (1992) introduced a procedure for scaling VOC-based *k*OH that involves scaling a VOC by multiplying its concentration (in ppbC) by the ratio of its rate coefficient with OH and the rate coefficient of the reaction between propylene and OH. This adjusted VOC concentration is called its Propy-Equivalent concentration and also helps rank VOCs according to their impact on O_3.

3.7.1 Urban NO and NO_2 Concentrations

Figure 3.15 summarizes the NO_2 concentrations by province for 2008. This figure shows the median and range among urban sites of the 98th percentile values of the daily hourly maximum. The median was highest among the Ontario and Québec sites at about 55 ppb. Western Canadian cities from Manitoba westward and New Brunswick had similar median 98th percentiles at near 40 ppb. However, the location with the highest 98th percentile was in Alberta as was the site with the lowest value. Nova Scotia, Newfoundland and the Northwest Territories all had lower values. Annual mean NO concentrations ranging from 22 to 32 ppb are found at centre city or roadway sites in the larger metropolitan areas. At centre city or roadway sites in the larger metropolitan areas NO can reach high levels surpassing 150 ppb and explaining 50–70 % of measured NO_X. At suburban sites and commercial sites in smaller urban centres, annual mean NO concentrations are typically in the range of 10 to 20 ppb while NO_2-to-NO ratios are in the range of 1 to 4.

Monthly mean NO and NO_2 concentrations for selected sites are presented in Fig. 3.16 averaged over the years 2003–2005. Both NO and NO_2 concentrations are lower in the summer months and NO_2/NO ratios are higher. The majority of urban sites show a strong seasonal cycle in NO, with maximum concentrations experienced in the winter months. The winter maximum is a result of three factors: increased emissions in the winter (primarily from fuel combustion); reduced atmospheric dispersion and a shallower mixed layer in winter; and less photochemical activity, resulting in slower destruction of NO_x. Figure 3.16 shows that these factors contribute to the largest seasonal variability and highest wintertime NO concentrations in the country in Calgary. The NO_2 seasonal cycle had a smaller amplitude compared to NO. The minimum was generally recorded in midsummer, due to faster conversion of NO_2 to other more oxidized nitrogen species (e.g., nitric acid) and lower NO levels.

3.7.2 Levels of Volatile Organic Compounds (VOCs)

VOC samples are routinely collected at 37 urban and 13 rural sites across Canada with five sites located near refineries. In Fig. 3.17, these sites are categorized according to these three settings and are compared using mean, 10th and 90th percen-

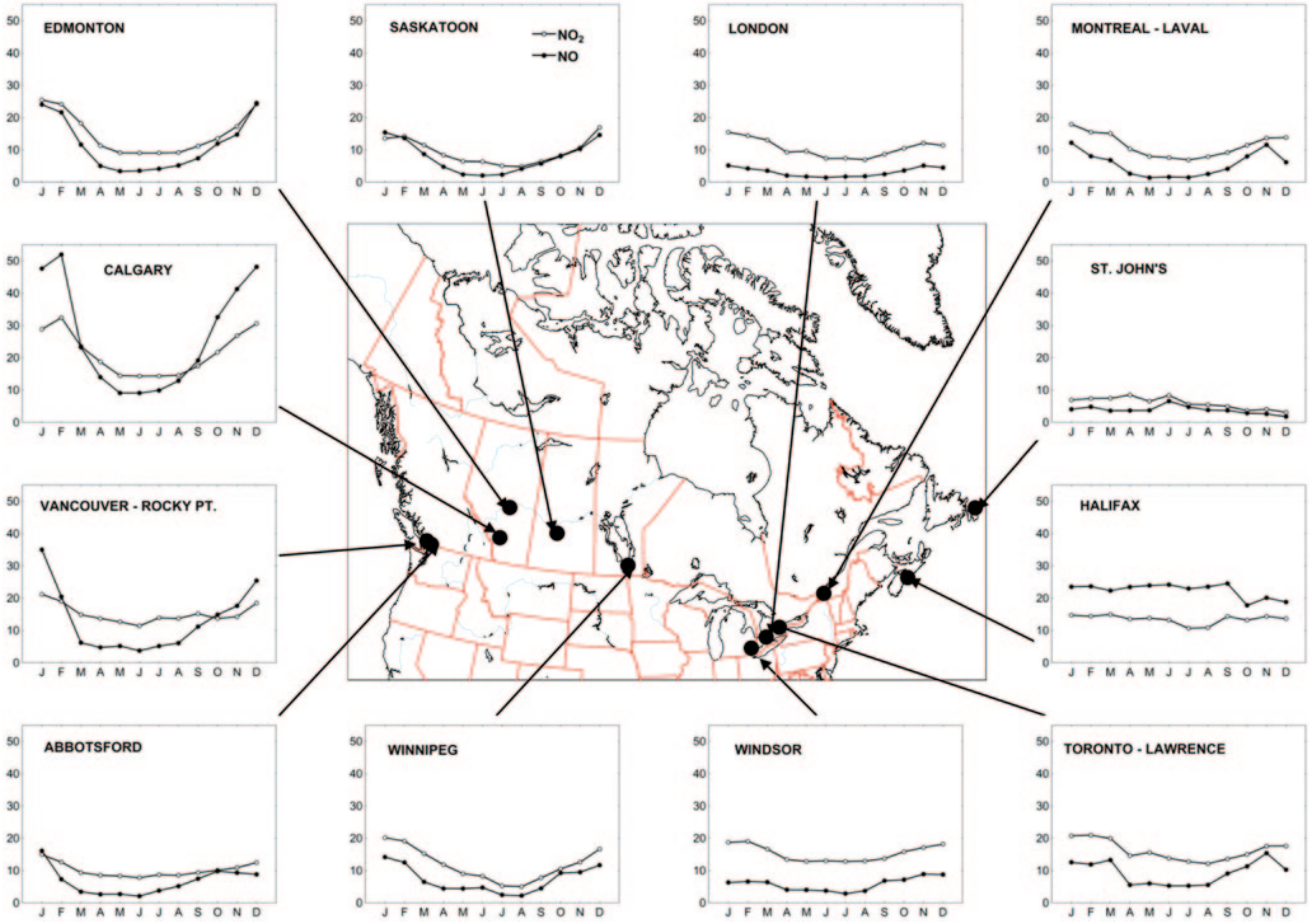


Fig. 3.16 Mean NO (*open circles*) and NO_2 (*solid circles*) by month averaged over the years 2007–2009. (Mean NO and NO_2 concentration (ppb) shown on y-axis)

Fig. 3.17 Mean, 10th and 90th percentile non-methane hydrocarbon concentrations (ppbC) for May to September in 2007–2009

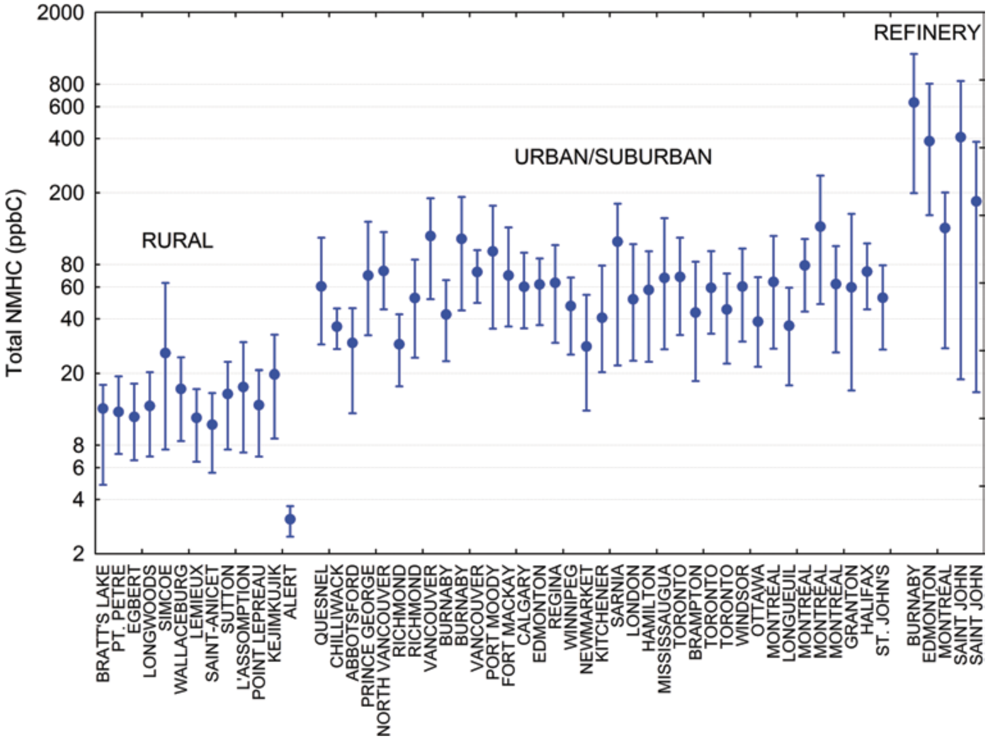


Table 3.4 Ten most abundant NMHC Species as a percentage of total carbon and as a percent of NMHC$_{prop}$—Urban and Rural Sites (Summer Only) 2007–2009

Rank	Compound	Mean (ppbC)	% of total	Compound	Mean (ppbC)	% of total
Urban—sorted by concentration				*Urban—sorted by NMHC$_{prop}$*		
1	Toluene	5.6	9.5	*m* and *p*-Xylene	2.7	15.3
2	Isopentane	4.9	8.4	Toluene	1.9	10.9
3	Propane	4.7	8.0	Ethylene	1.5	8.6
4	Ethane	3.7	6.3	Isoprene	1.0	5.7
5	Butane	3.5	5.9	Propylene	0.8	4.5
6	*m* and *p*-Xylene	2.9	5.0	Isopentane	0.7	4.0
7	Pentane	2.2	3.8	*o*-Xylene	0.6	3.4
8	Isobutane	2.2	3.7	1,2,4-Trimethylbenzene	0.5	3.1
9	Ethylene	1.9	3.3	3-Ethyltoluene	0.4	2.3
10	2-Methylpentane	1.3	2.2	Butane	0.4	2.3
All species		*59.0*			*17.6*	
Rural—Sorted by Concentration				*Rural—sorted by NMHC$_{prop}$*		
1	Ethane	2.3	16.3	Isoprene	1.3	30.0
2	Isoprene	1.4	10.0	Ethylene	0.3	6.9
3	Propane	1.2	8.9	Toluene	0.3	6.4
4	Isopentane	0.8	5.9	α-Pinene	0.3	6.2
5	Toluene	0.8	5.8	m and p-Xylene	0.2	5.2
6	α-Pinene	0.7	5.1	Propylene	0.2	4.3
7	Butane	0.7	4.7	Isopentane	0.1	2.8
8	Pentane	0.5	3.2	β-Pinene	0.1	2.4
9	Ethylene	0.4	2.7	1-Butene/Isobutene	0.1	2.1
10	β-Pinene	0.4	2.6	δ-Limonene	0.1	1.7
All species		*14.0*			*4.3*	

tile NMHC concentrations. Not surprisingly concentrations were much lower at the regionally representative and remote sites compared to the rural-urban impact sites. Higher levels are apparent at locations situated in the downtown core (e.g., Vancouver-Robson) and (Montréal-Maisoneuve) as well as in cities near petrochemical industry (Sarnia).

Mean concentrations varied by almost three orders of magnitude from the lowest site (Alert—3.1 ppbC) to the highest site (Burnaby-Eton/Madison—633 ppbC). The Burnaby site is located approximately 1 km from a Chevron refinery. The differences among sites are not the same for all individual compounds. Some of the NMHC species with relatively long atmospheric life times such as ethane are found in similar concentrations at urban and rural sites. Biogenic species are usually more abundant at forested, rural sites (e.g., Kejimkujik). For the rural sites the proximity of a large source regions plays a role. For example, NMHC concentrations are relatively large at such sites as Simcoe, Wallaceburg and l'Assomption which are relatively close to large urban areas.

Table 3.4 provides a list of the 10 most abundant NMHC species as a percent of total carbon and as a percent of reactivity weighted total carbon NMHC$_{prop}$ for the urban and rural site categories. The most abundant species on a percent of carbon basis in the urban area are toluene, isopentane, propane, butane and m and *p*-xylene. In contrast, the species *m* and *p*-xylene, toluene, ethylene, propylene and isopentane are the most important contributors to NMHC$_{prop}$ (i.e., important for O_3 formation). The ranking of major species measured in NMHC mixtures has remained stable during the past 20 years of measurement (Environment Canada 1997), particularly at the urban sites, which reflects the large contribution from transportation sources. Species profiles for the refinery impact sites (not shown) are similar to urban sites except for a greater abundance of C3-C5 alkanes (propane, butanes and pentanes). There also systematic differences between the profiles among refinery sites due to differences in industrial activities. For example, the Saint John, NB, site records high levels of MTBE and propylene while the Burnaby site shows high levels of 2,2-dimethylbutane. At the rural sites the importance of biogenic NMHC species, such as isoprene and α-pinene, is relatively large. In terms of NMHC$_{prop}$ isoprene is the dominant species.

3.7.3 Sulphur Dioxide Concentrations

Sulphur dioxide is a major precursor of $PM_{2.5}$ and has been routinely monitored across Canada for decades. Ambient SO_2 concentrations across Canada exhibit a seasonal cycle with higher urban SO_2 concentrations in the winter than in the summer. As shown below in Fig. 3.22, concentrations have continued to decrease in recent years, but sites with

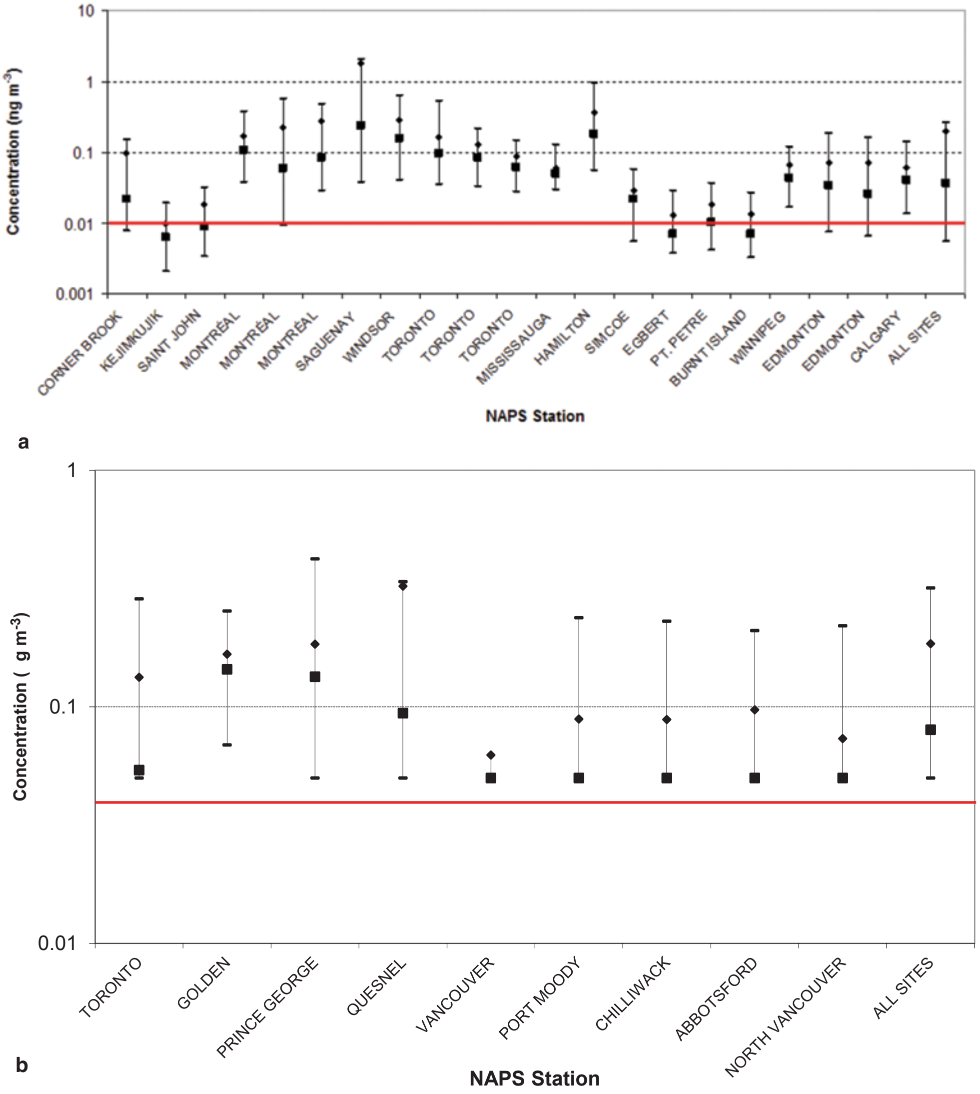


Fig. 3.18 Concentrations of polycyclic aromatic hydrocarbons (PAHs), as represented by benzo[a]pyrene (**a**), and ethylene oxide (**b**) at NAPS sites over 2003–2009. ■ Median concentration. — 10th and 90th percentile concentrations. ♦ Mean concentration. — Annual average ambient air quality criterion (AAQC) from Ontario

nearby point sources (usually non-ferrous smelters) continue to experience the highest SO_2 concentrations. However, the observed mean annual concentrations across the country are still below 10 ppb at all but a few sites, such as the industrially-impacted Temiscaming, Québec, and Trail, BC, locations. The lowest concentrations are found at western and northern sites with no nearby industrial emissions. Some rural sites in Ontario, such as Egbert and Longwoods, continue to experience higher SO_2 levels than a number of the western urban sites. This is because of higher emissions of SO_2 within the region, including the U.S. Midwest.

3.7.4 Levels of Hazardous Air Pollutants

Typical concentrations of the HAPS listed in Table 3.2 vary over a wide range depending upon compound (e.g., from femtograms (10^{-15} g) to micrograms (10^{-6} g) per cubic meter). Similarly, the concentrations at which no adverse effects are expected vary by compound.

Two of the HAPS listed in Table 3.2 exceeded their respective annual Ontario ambient air quality criteria (AAQC) by substantial margins in recent years: polycyclic aromatic hydrocarbons (PAHs, as represented by benzo[a]pyrene) and ethylene oxide. Their concentration distributions among NAPS sites are shown in Figs. 3.18a, b and compared to their AAQCs. Figure 3.18a shows that PAH concentrations varied widely by location. Sparsely populated areas, such as Kejimkujik National Park and Burnt Island (Lake Huron), had median concentrations below Ontario's annual AAQC. Conversely, most urban areas had median concentrations that exceeded the AAQC due to the impact of local mobile and industrial sources. All NAPS sites that measured ethylene oxide had median concentrations that were above the

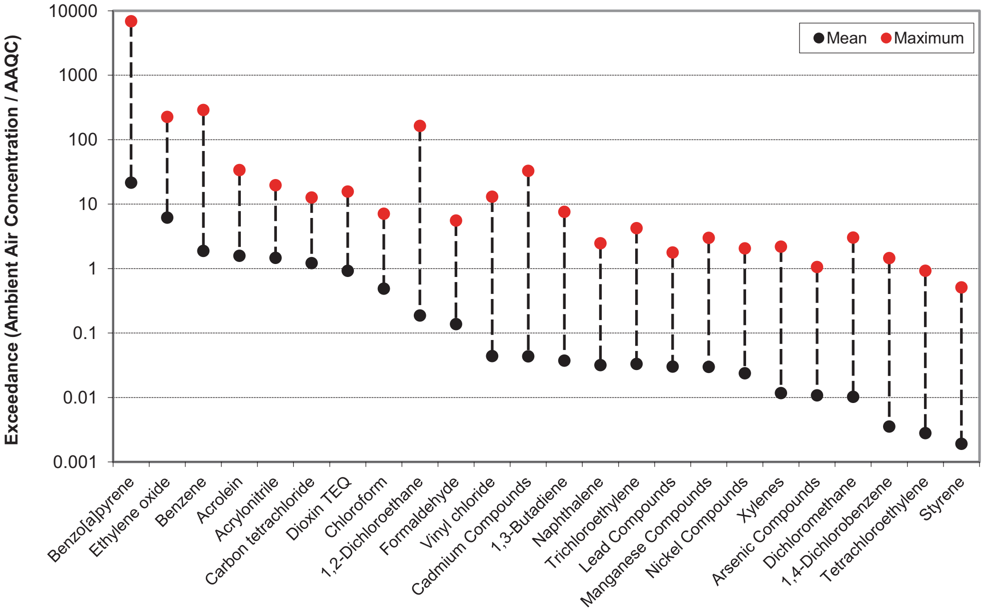


Fig. 3.19 Air toxics in Canada with maximum 24-Hour exceedance greater than 0.1 over 2003–2008. Note that not all pollutants are measured at all stations and/or in all years. Benzo[a]pyrene is a surrogate compound representing the PAHs. 24-hour AAQCs used to calculate exceedances

Ontario AAQC (Fig. 3.18b). However, this HAP was only measured in British Columbia and at a single site in Ontario. It is possible that improved spatial coverage in ethylene oxide measurements would reveal areas with concentrations below the AAQC and others further above it (i.e., hotspots). More measurements are needed to determine the spatial variability of ethylene oxide in Canada.

The range in concentrations among the HAPS in Table 3.2 is large, and a useful approach to summarizing their levels and potential impact is to express their concentrations relative to their respective Ontario AAQCs. Compounds that exceeded or approached their relevant AAQCs in Canada in recent years are shown in Fig. 3.19. A plotted value > 1 indicates that the concentration (mean or maximum Canadian value) was above the Ontario AAQC. The mean Canadian concentrations of six air toxics (PAHs as represented by benzo[a]pyrene, ethylene oxide, benzene, acrolein, acrylonitrile, and carbon tetrachloride) exceeded their respective annual Ontario AAQCs. A further sixteen air toxics exceeded their AAQCs at their maximum recorded concentrations. In other words, there was at least one observation among the limited number of measurement sites in Canada that was above the AAQC for those compounds. Continued investigations of HAPs across Canada will be useful to understanding the risks posed to Canadians by air toxics. However, careful attention must be paid to setting science priorities given the large list of HAPs that are potentially relevant in the Canadian context (Galarneau and Dann 2011).

3.7.5 Trends in NO_2, NO, VOCs and SO_2

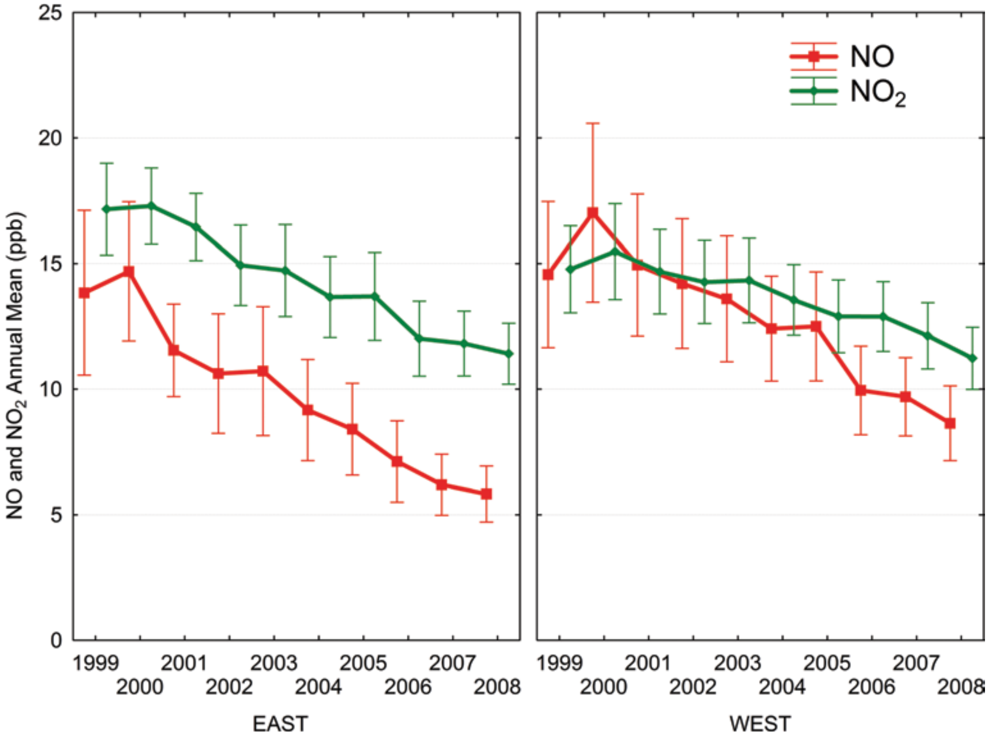
Recent trends in annual mean NO and NO_2 are shown in Fig. 3.20 and trends in May-September VOCs at urban, rural and refinery impacted sites are provided in Fig. 3.21. Between 1999 and 2008 annual mean NO decreased by 48 %, NO_2 by 26 % and summer mean VOCs by 46 % at these Canadian urban sites. The trends were consistent, on a site by site basis, with essentially all urban sites in Canada, recording similar decreases in ambient levels. Interestingly, given the reductions in urban NO_2 during the period, the levels in eastern and western Canadian cities are now comparable. In contrast, mean NO is higher in the cities in western Canada. As a result, the ratio of composite annual mean NO_2 to NO increased from 1.1 to 1.5 during the time period. Rural sites experienced a decline in anthropogenic VOCs similar to the urban sites with a 49 % decrease in mean summer concentration between 1999 and 2008. The reductions in ambient air NO_X and NMHC also matches the reduction in on-road transportation sector emissions (Government of Canada 2007). In contrast, Fig. 3.21 shows that mean and 90th percent confidence limits for the VOCs at the six sites located near refineries have increased during the past decade. This upward trend was largely driven by higher concentrations measured at the Edmonton and Vancouver monitoring sites.

In Fig. 3.22 annual mean SO_2 trends are shown separately for 12 urban and 9 industrial influence sites for the 1999–2008 time period. Composite annual mean SO_2 concentrations decreased by approximately 52 % at both groups of sites. Mean levels in locations impacted by industrial emissions now range from 3 to 9 ppb, while at most other urban locations they average less than 2 ppb.

3.8 Summary

There are over 300 air quality monitoring locations across Canada from coast to coast to coast. Ozone and fine particulate matter ($PM_{2.5}$) are the most frequently measured pollut-

Fig. 3.20 Yearly variation in annual mean NO and NO_2 from Canadian urban trend sites (1999–2008). Composite means and the 90th percent confidence interval around the mean are plotted



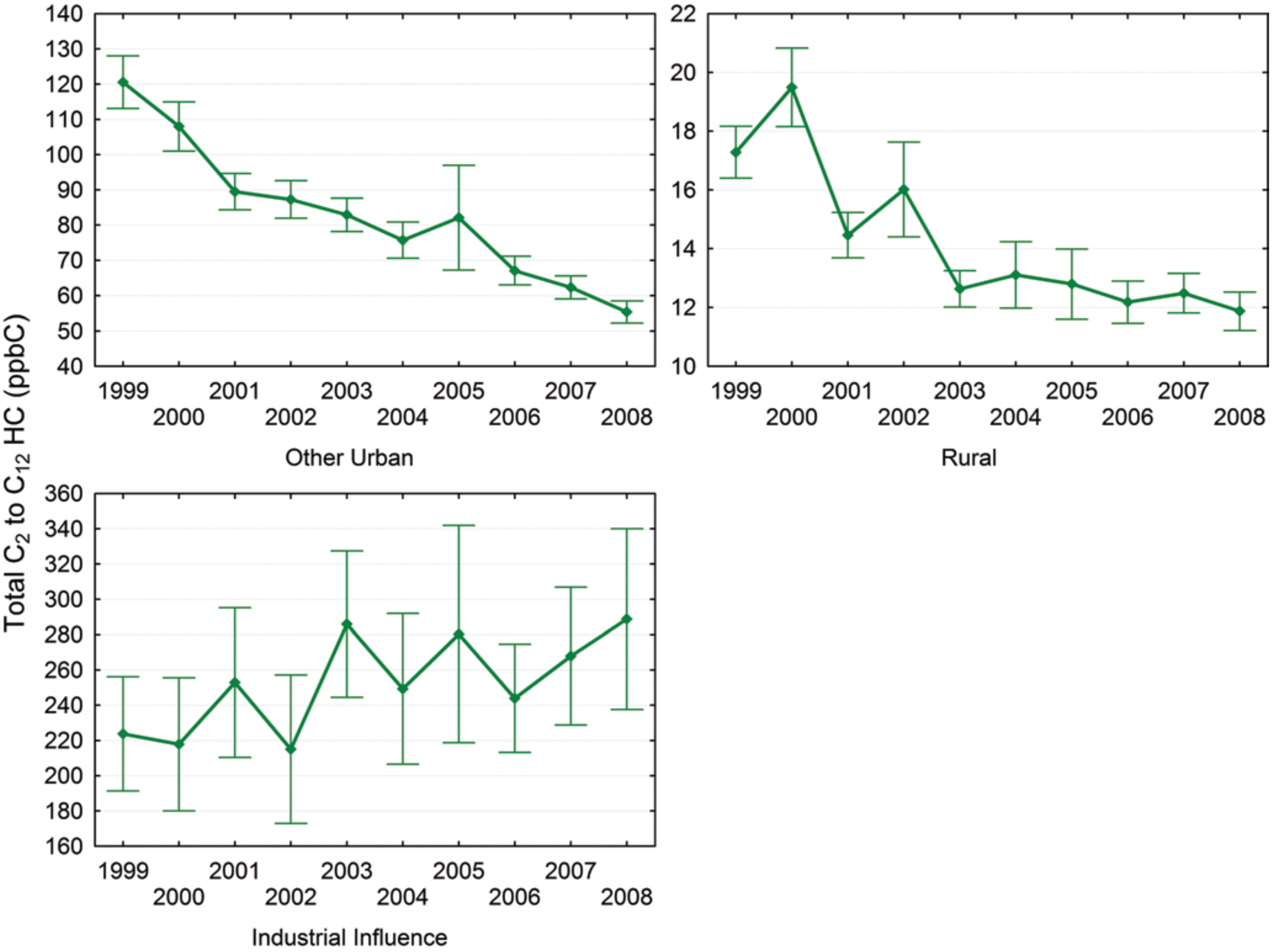
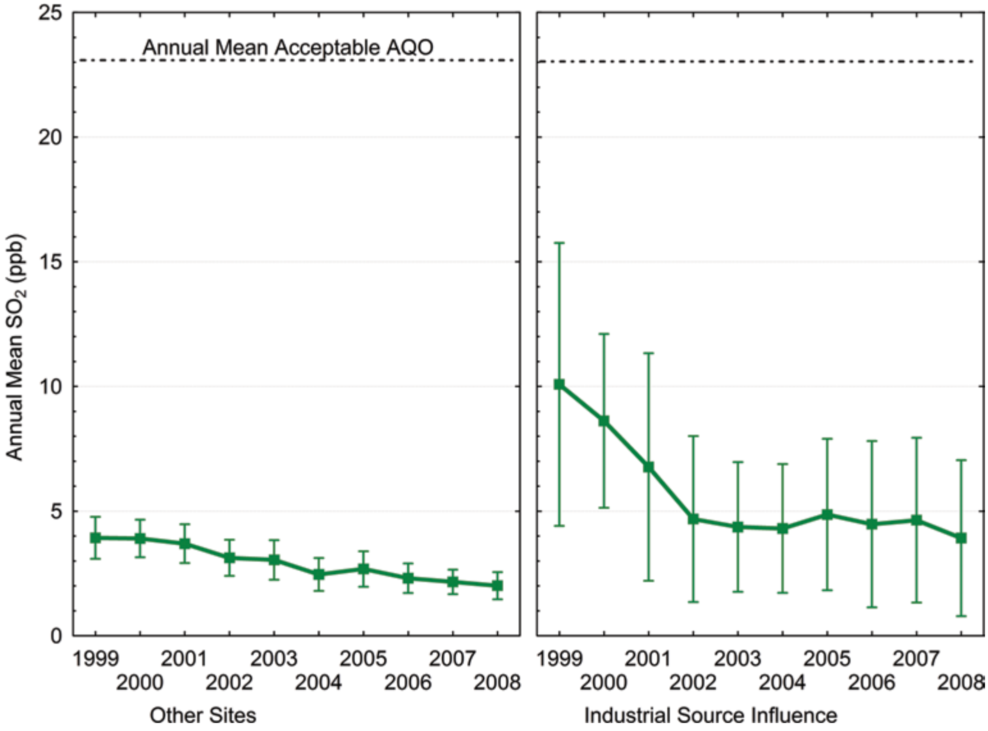


Fig. 3.21 Yearly variation in non-biogenic C_2 to C_{12} hydrocarbons at industrial influence, other urban and rural sites (May to September only). Composite means and the 90th percent confidence interval around the mean are plotted. Independent scales are used for each plot

Fig. 3.22 Yearly variation in annual mean SO_2 from Canadian industrial influence and other trend sites (1999–2008). Composite means and the 90th percent confidence interval around the mean are plotted



ants with the majority of locations situated where population density is high or where there is considerable industrial activity. Nitrogen oxides and sulphur dioxide are also measured at a considerable number of locations in Canada while volatile organic compounds (VOC) and other air toxics are measured at a relatively small number of places. In this chapter, national scale patterns for these air pollutant concentrations have been presented along with information on their temporal variations, such as diurnal, seasonal and year to year changes. The composition of fine particulate matter ($PM_{2.5}$) and volatile organic compounds (VOC) has also been determined and described in this chapter.

The concentration measurements indicate that there are areas where the current Canadian standards or provincial guidelines are exceeded for ozone, $PM_{2.5}$ and toxics. Ozone in southern Ontario and southern Québec has been the most problematic although there has been some improvement in recent years. Similarly, on a national scale $PM_{2.5}$ has decreased indicating that, due to the range of emission reduction strategies implemented in the past decade, air quality is improving. There are exceptions, such as VOCs in areas of industrial influence and, although not a focus in this chapter, local areas of industrial growth such as NO_2 over the oil sands region (McLinden et al. 2012). Thus, there remains a need to monitor air quality, especially in regions of projected industrial or population growth in order to maintain current levels and to identify options for improvement and to inform adaptive management approaches. Furthermore, current health research indicates that pollutants such as $PM_{2.5}$ have effects on the population at low levels. Thus, air quality remains an important public health issue to track; nationally, regionally and in particular at the local scale where areas of concentrated emissions due to industrial activity or population behaviour (e.g., traffic, wood burning) can lead to higher population exposures.

In the context of the past 30–40 years, Canadian air quality has been an environmental management success story. Levels throughout the populated regions of the country are considerably lower than in the 1970's and 1980's. The 1990's also brought reductions and this chapter shows that even up through the 2000's improvements in air quality have been realized.

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