Canada – United States

Transboundary Particulate Matter

Science Assessment 2013

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LIST OF ACRONYMS AND ABBREVIATIONS

APEI – Air Pollutant Emissions Inventory

AURAMS – A Unified Regional Air Quality Modelling System

BAQS-Met – Border Air Quality and Meteorological Study

Bc:Al – base cation to aluminum ratio

BLIERS – Base Level Industrial Emission Requirements

CAC – Criteria Air Contaminants

CAAQS – Canadian Ambient Air Quality Standards

CAP – criteria air pollutant

CAPMoN – Canadian Air and Precipitation Monitoring Network

CASTNET – Clean Air Status and Trends Network

CO – carbon monoxide

CWS – Canada-wide Standards

FCCE – Future Climate Current Emissions

FCFE – Future Climate Future Emissions

FRM – Federal Reference Method

GEM-MACH – Global Environmental Multi-scale–Modelling Air quality and Chemistry (model)

HAP – hazardous air pollutant

HNO_3 – nitric acid

H_2SO_4 – sulphuric acid

IMPROVE – Interagency Monitoring of Protected Visual Environments (network)

NAPS – National Air Pollution Surveillance

NAAQS – U.S. National Ambient Air Quality Standard

NAESI – National Agri-Environmental Standards Initiatives

NEI – National Emissions Inventory

NH_3 – ammonia

NH_4^+ – ammonium

NH_4NO_3 – ammonium nitrate

$(NH_4)_2SO_4$ – ammonium sulphate

NO_x – nitrogen oxides

NO_3^- – nitrate

NPRI – National Pollutant Release Inventory

O_3 – ozone

PM – particulate matter

PM$_{2.5}$ – in general terms, particulate matter with an aerodynamic diameter of less than or equal to a nominal 2.5 microns; a measurement of fine particles

PM$_{10}$ – in general terms, particulate matter with an aerodynamic diameter of less than or equal to a nominal 10 microns; a measurement of thoracic particles (i.e., that subset of inhalable particles thought small enough to penetrate beyond the larynx into the thoracic region of the respiratory tract)

RCP6 – Representative Concentration Pathways; stabilization without overshoot pathway to 6 W/m^2 at stabilization after 2100

SOA – secondary organic aerosols

SO$_x$ – sulphur oxides

SO$_2$ – sulphur dioxide

SO$_4^{-2}$ – sulphate

SRES – Special Report on Emissions Scenarios

U.S. EPA or EPA – United States Environmental Protection Agency

VOCs – volatile organic compounds

MEASUREMENT UNITS

km – kilometre

km^2 – square kilometre

L – litre

mol/ha/year – moles per hectare per year

Mm^{-1} – inverse megametres

µg/m^3 – micrograms per cubic metre

µmol of charge/L – micromoles of charge per litre

nm – nanometre

µm – micron; micrometre

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EXECUTIVE SUMMARY

This document provides an update of the findings from the 2004 *Canada–United States Transboundary Particulate Matter Science Assessment* (the 2004 Assessment). Its goal is to present a scientific and technical basis for discussions of adding a particulate matter (PM) annex to the *1991 Canada–United States Air Quality Agreement* (the Agreement), to assess the potential impacts of a PM annex and, ultimately, to help determine whether such an annex is currently warranted.

This assessment focuses on the fine particle fraction of PM, or $PM_{2.5}$, because this size fraction can remain suspended in the air for several days to weeks, can be transported by winds over large distances, and thus is subject to atmospheric transboundary transport in North America.

This document is organized around five key science questions:

1) What are the impacts of $PM_{2.5}$ on human/ecosystem health and public welfare, and what are the current air quality standards to protect human and ecosystem health in Canada and the U.S.?
2) What are the recent levels of $PM_{2.5}$ in Canada and the U.S.?
3) What are the emissions and emission trends of the pollutants that contribute to ambient $PM_{2.5}$ concentrations in Canada and the U.S.?
4) What is the evidence that transboundary flow of $PM_{2.5}$ occurs across the Canada – U.S. border, and what changes are projected, given future emission rates in both countries?
5) Are there emerging science issues that could affect the understanding of $PM_{2.5}$ formation, $PM_{2.5}$ levels, and its impacts on human and ecosystem health?

Key Findings

- **$PM_{2.5}$ and its precursors have significant effects on the health of humans and ecosystems.**

The already extensive body of studies providing evidence on the effects of fine particles on health has grown significantly since the 2004 Assessment. These studies provide evidence of consistent increases in premature mortality and morbidity related to ambient $PM_{2.5}$ concentrations, with the strongest evidence being reported for cardiovascular-related effects. Furthermore, the ubiquity of $PM_{2.5}$ implies that exposure to ambient $PM_{2.5}$ concentrations can have a substantial public health impact, even with recent reductions. In addition, although deposition (wet and dry) of acidifying sulphur and nitrogen compounds related to $PM_{2.5}$ in Canada and the U.S. has been reduced since 2004, recent deposition in both countries continues to exceed thresholds (termed critical loads) in some geographic areas, thus posing a risk of harmful effects to terrestrial and aquatic ecosystems. Finally, although significantly reduced in most border areas, $PM_{2.5}$ continues to contribute to visibility impairment in both Canada and the U.S., particularly in highly populated regions of southern Ontario and Quebec in Canada and in the Midwest and Montana in the U.S. In response, both Canada and the U.S. recently lowered ambient air quality standards to protect human and ecosystem health from the harmful impacts of $PM_{2.5}$.

- **Recent levels of ambient $PM_{2.5}$ have been declining in both Canada and the U.S.**

In Canada and the U.S., ambient concentrations of $PM_{2.5}$ have diminished significantly from the levels reported in the 2004 Assessment. More specifically, between 2000 and 2012 the U.S. national average annual and 24-hour (h) concentrations of $PM_{2.5}$ decreased 33% and 37%, respectively. Data from Canadian $PM_{2.5}$ speciation sites indicate that, between 2003 and 2010, average annual concentrations of $PM_{2.5}$ declined about 4 $\mu g/m^3$ in eastern Canada while average levels across western Canada remained fairly constant. In 2012, ambient concentrations reported at most monitoring sites in the U.S. along the Canadian border would meet the annual and 24-h National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ set in 2012. In both eastern and western Canada, data from the filter-based monitoring network indicate that average annual concentrations (2008–2010) would meet the Canadian Ambient Air Quality Standard (CAAQS) set for 2015.

- **The decline of most $PM_{2.5}$ precursors is expected to continue, while direct emissions of $PM_{2.5}$ and ammonia (NH_3) have and are expected to remain relatively stable.**

National emission inventories in both Canada and the U.S. show that emissions of the $PM_{2.5}$ precursors sulphur dioxide, nitrogen oxides and volatile organic compounds (SO_2, NO_x and VOCs) declined between 2002 and 2010. However, total direct emissions of anthropogenic $PM_{2.5}$ have remained fairly stable in both Canada and the U.S. during this period, as have emissions of ammonia (NH_3).

Projections based upon known policies established in Canada and the U.S. for governing future emissions indicate that emissions of $PM_{2.5}$ and its precursors will follow recent trends. In Canada, primary emissions of $PM_{2.5}$ are expected to remain stable through 2020, while emissions of sulphur oxides (SO_x) and NO_x are projected to decline by 33% and 13%, respectively, between 2006 and 2020. By contrast, Canadian VOC and NH_3 emissions are not projected to change significantly in this period. In the U.S., emissions of SO_2, NO_x and VOCs are forecast to decrease 65%, 42% and 21% from 2008 to 2020, while emissions of $PM_{2.5}$ are projected to decrease modestly (8%). NH_3 emissions in the U.S. are expected to be 2% higher in 2020 than in 2008.

- **Projections are that the influence of transboundary transport between Canada and the U.S. will be reduced and that current and planned $PM_{2.5}$ ambient air standards will likely not be exceeded.**

Modelling analyses of the impact of future emission projections show notable anticipated reductions in ambient $PM_{2.5}$ concentrations between 2006 and 2020 in both Canada and the U.S. Significant declines in ambient $PM_{2.5}$ concentrations are expected to occur in most border region cities, with percentage reductions ranging up to 35% in major U.S. cities near the border and up to 25% in their Canadian counterparts.

There is ongoing evidence that $PM_{2.5}$ is transported across the Canada – U.S. border. However, for most cities in both countries, the dominant sources of $PM_{2.5}$ in 2020 continue to be domestic emissions; overall, transboundary influence is projected to be less in 2020 than in 2006. The influence of U.S. emissions on $PM_{2.5}$ concentrations in Canadian cities near the border is projected to decrease by about 2–10%, with the largest reductions occurring in eastern Ontario and southwestern Quebec. The exception is Abbotsford, B.C., where there is a small projected increase in U.S. influence. The influence of Canadian emissions on select U.S. cities is also projected to decrease, but by less, in the range of 1–3%, with the exceptions of Seattle, Wash., Buffalo, N.Y., and Rochester, N.Y., where the Canadian influence is projected to increase slightly.

In the U.S., no areas in the border region are predicted to exceed the current annual or 24-h $PM_{2.5}$ NAAQS (12 $\mu g/m^3$) in 2020, including areas with projected increases in Canadian influence. In Canada, the predicted 2006–2020 decreases in $PM_{2.5}$ are expected to result in rural/regional background $PM_{2.5}$ concentrations over the region near the southern Ontario and southern Quebec border that will be below the 2015 and 2020 annual and 24-h CAAQS. However, these levels are close enough to the CAAQS that some populated areas with relatively large local emissions may experience $PM_{2.5}$ above the CAAQS. In the border regions of western and Atlantic Canada, 2015 and 2020 CAAQS levels are not projected to be exceeded.

- **Emerging air quality issues could influence future concentrations of $PM_{2.5}$ in both countries; thus there is a continued need to improve our scientific understanding of health and ecological effects, the impacts of air quality management activities, and the magnitude of transboundary transport.**

The following emerging science issues will probably affect future ambient $PM_{2.5}$ concentrations and/or how air quality management activities are developed to address $PM_{2.5}$:

- the need for improved understanding of the health effects of $PM_{2.5}$ and its components in the context of exposure to other pollutants and how these combined effects might affect air quality standards and management strategies
- the need for increased understanding of the impacts of climate change on $PM_{2.5}$ concentrations and of the effects of $PM_{2.5}$ and its components on climate change
- the effects of changes in the mix of energy generation and end-use technologies on the concentrations of $PM_{2.5}$ and the impacts of growing domestic fossil fuel extraction activities, such as the oil sands, and unconventional oil and gas development, such as the use of hydraulic fracturing (fracking)
- changes in the relative importance of natural sources and intercontinental transport that could affect the management of ambient $PM_{2.5}$ concentrations in Canada and the U.S.

As the science continues to evolve on these issues, air quality management activities in both Canada and the U.S. may need to adjust in order to continue to effectively protect public health and the environment.

Conclusions

This updated Transboundary PM Science Assessment cites a number of studies (including those on which the U.S. NAAQS and Canadian CAAQS are based) documenting that exposure to $PM_{2.5}$ leads to significant impacts on human health, ecosystem health and public welfare; and that additional reductions of $PM_{2.5}$ below the NAAQS and CAAQS would have additional public health benefits. To address $PM_{2.5}$ standards as well as other important air quality issues, regulatory and non-regulatory actions are being implemented in both countries to reduce emissions of $PM_{2.5}$ and its precursor pollutants. Air quality modelling analyses indicate that future concentrations of ambient $PM_{2.5}$ are not expected to exceed the current $PM_{2.5}$ NAAQS or CAAQS along the Canada–U.S. border, with a few possible exceptions limited to cities near the border with significant local contributions to $PM_{2.5}$.

Because of the important health and environmental effects associated with $PM_{2.5}$, it would be beneficial for both countries to track progress and exchange information relevant to achieving $PM_{2.5}$-related emissions reductions, air quality improvement and program implementation over time. Doing so would provide confidence that future $PM_{2.5}$ concentrations in the border region will be below the NAAQS and CAAQS; it would also afford an ongoing opportunity to determine how emerging issues, such as climate change and an evolving energy landscape, may be impacting $PM_{2.5}$ concentrations and transboundary transport over time.

Given the science reported in this assessment and the important role of $PM_{2.5}$ in air quality management activities in both Canada and the U.S., there would be value in addressing $PM_{2.5}$ in some manner under the Agreement.

CHAPTER 1: INTRODUCTION

1.1 Historical Context

On March 13, 1991, the President of the United States and the Prime Minister of Canada signed the Canada–United States Air Quality Agreement (hereafter referred to as the Agreement). Its purpose was to establish “a practical and effective instrument to address shared concerns regarding transboundary air pollution.” Although at its inception, the Agreement was intended to address the primary pollutants responsible for acid rain, it also confirmed the commitment of the signatories to consult on, and develop, the means to address other transboundary air pollution issues.

In 1997, in response to shared concerns over the transboundary transport of ozone and fine particulate matter (PM $_{2.5}$), Canada and the U.S. signed a “Commitment to Develop a Joint Plan of Action for Addressing Transboundary Air Pollution.” The commitment articulated the intent of the parties to jointly address the shared problems of ground-level ozone (O_3) and PM within the framework of the Agreement. Stemming from this commitment, the parties signed a Joint Work Plan for Transboundary Fine Inhalable Particles in June 1998. As part of this work plan, the joint *Canada–United States Transboundary PM Science Assessment* was completed in 2004 (the 2004 Assessment). To provide the scientific support required to determine the need for a PM annex pursuant to the Agreement, the 2004 Assessment articulated seven key objectives:

- to identify whether or not there is a fine PM problem in the border regions (ambient observations vs. standards), focussing on health, visibility and environmental endpoints;
- to identify the extent of the problem (if standards are exceeded, by how much, where and when are they exceeded);
- to describe the PM issue in terms of geographic regions (i.e. west, central, east);
- to identify PM precursors of concern on a regional or sub-regional basis;
- to describe sources (or source regions) of PM and PM precursors in the context of geographic regions (i.e. west, central, east);
- to describe emissions of PM and PM precursors, the spatial distribution of emissions and the transport characteristics of these emissions; and,
- to identify the impact of current and proposed emission reduction scenarios on fine PM levels in North America.

The following is a brief summary of the key findings from this 2004 document.

- Transboundary transport of $PM_{2.5}$ can contribute to above-average PM levels in both Canada and the U.S.
    - Ambient levels of $PM_{2.5}$ near the border exceed the standards set for $PM_{2.5}$ in several regions of both Canada and the U.S.
    - $PM_{2.5}$ is transported across the border region between Canada and the U.S., leading to elevated concentrations of $PM_{2.5}$ in both countries.
- PM levels vary significantly over geographic regions.
    - Elevated concentrations of $PM_{2.5}$ are found more often in the following regions: the northeastern and northwestern U.S., the U.S. industrial Midwest, and southwestern Ontario.

- There are many sources of PM and PM precursors.
    - Local mobile and industrial sources make a constant contribution to PM levels in Toronto and dominate on relatively clean days with air flow coming from the north.
    - Coal-related sources in the U.S. make a substantial contribution to PM in Toronto and elsewhere in Canada.
    - High agricultural NH_3 emissions influence ammonium nitrate (NH_4NO_3) levels.
    - Sulphur dioxide (SO_2) and nitrogen oxides (NO_x) are primarily regional contributors to PM, while organic or black carbon and other PM constituents tend to originate from local sources.
- Air quality modelling of emission reduction scenarios for PM and PM precursors found:
    - Projected reductions vary spatially and by season.
    - Additional reductions of SO_2 and NO_x emissions should produce concomitant effects on ambient $PM_{2.5}$ levels as well as concurrent reductions in particle ammonium (NH_4^+).
    - There are linkages between PM and other air quality issues, including ecosystem deposition leading to acidification, O_3 production, and visibility reductions.

Negotiations toward a PM annex to the Agreement were initiated in 2007, but an annex was never completed due to a variety of factors in both countries, and negotiations were suspended in 2008. The timeframe for resuming negotiations was discussed at the annual Canada–U.S. Air Quality Committee meetings in 2010 and 2011, and Sub-Committee 2 was tasked with updating the 2004 Assessment.

In February 2011, Canada and the U.S. created the Regulatory Cooperation Council (RCC) to provide smarter, more effective approaches to regulation that enhance the economic competitiveness and well-being of the U.S. and Canada, while maintaining high standards of public health and safety and environmental protection. In December 2011, the RCC released an initial Joint Action Plan to foster regulatory alignment. It listed several initiatives with separate work plans, including an approach to better synchronizing the implementation of environmental regulations and leveraging existing expertise in each party's regulatory systems. The environmental initiative included plans "to consider the expansion of the Canada–U.S. Air Quality Agreement to address transboundary particulate matter, the air pollutant most commonly associated with premature mortality, based on comparable regulatory regimes in the two countries."

1.2 Scope and Objective of this Assessment

The aerosol particles suspended in the air referred to as PM are pollutants that adversely affect human and ecosystem health. PM includes both solid particles and liquid droplets that vary in size and chemical composition. Two size fractions of ambient PM are commonly measured for regulatory purposes: particles with a diameter of 10 microns or less (PM_{10}), which is considered a limit for inhalation into the upper regions of the human respiratory tract, and smaller particles with a diameter of 2.5 microns or less (fine PM or $PM_{2.5}$). This latter size fraction has received considerable attention for the past 10–20 years because of concern for human health, due to the findings of a large body of health effects research and considerations of size, exposure, and relative potency. The fine fraction is also an important contributor to the degradation of visibility (haze).

As PM and its precursor pollutants originate from multiple sources, its chemical composition is complex and variable. The dominant chemical constituents, particularly for $PM_{2.5}$, are organic compounds, elemental or black carbon, sulphate (SO_4^{-2}), ammonium (NH_4^+), NO_3^- and other inorganic constituents, such as crustal material and trace metals. PM_{10} consists of these fine particles and also the coarse fraction (between 2.5 and 10 µm), which generally contains more re-suspended dust. This includes particles from abrasive action such as that associated with vehicle traffic (e.g. tire wear, brake wear) and also wind-blown soil, but also a considerable amount of biological material (e.g. pollen and spore fragments).

This assessment focuses on $PM_{2.5}$ because it can remain suspended in the air for several days to weeks and can be transported by winds over large distances. As a result, $PM_{2.5}$ is subject to atmospheric transboundary transport over the Canada–U.S. border through complex atmospheric processes, which involve a range of meteorological patterns occurring on multiple spatial scales and emissions from numerous sources as a result of both human and natural activities. In cities, for example, $PM_{2.5}$ in the air can originate from both local sources and from others that may be hundreds to thousands of kilometres away. Therefore, binational cooperation is needed to manage and reduce $PM_{2.5}$ air pollution in both countries.

The objective of this document is to review and update, as appropriate, the findings of the 2004 Assessment to provide the scientific and technical basis for discussions of a potential PM annex to the 1991 Agreement. The document is organized around the following five questions.

- What are the impacts of $PM_{2.5}$ on human/ecosystem health and public welfare, and what are the current air quality standards to protect human and ecosystem health in Canada and the U.S.? (Chapter 2)
- What are the recent levels of $PM_{2.5}$ in Canada and the U.S.? (Chapter 3)
- What are the emissions and emission trends of the pollutants that contribute to ambient $PM_{2.5}$ concentrations in Canada and the U.S.? (Chapter 4)
- What is the evidence that transboundary flow of $PM_{2.5}$ occurs across the Canada – U.S. border and what changes are projected, given future emission rates in both countries? (Chapter 5)
- Are there emerging science issues that could affect the understanding of $PM_{2.5}$ formation, $PM_{2.5}$ levels, and its impacts on human and ecosystem health? (Chapter 6)

Due to timing and resource constraints, this updated assessment relies on existing monitoring and modelling data and analyses to address these questions: no new targeted data collection or modelling was done. However, the document does present new insights into, and interpretation of, data and modelling studies that relate specifically to transboundary transport of PM along the Canada – U.S. border.

Like the 2004 Assessment, this document is an example of the important bilateral scientific cooperation that occurs under the Agreement. Such cooperation not only supports and enhances the implementation of the Agreement, but also adds to the body of knowledge required for achieving broader air quality management and public health objectives in both nations. Toward these ends, this document will clarify the potential effects of an annex to the Agreement that addresses PM and, ultimately, will help to determine whether such an annex is warranted at this time.

CHAPTER 2: IMPACTS OF PARTICULATE MATTER

Chapter Summary

- There is an extensive body of scientific studies providing evidence that $PM_{2.5}$ adversely affects human health. These studies provide evidence of consistent increases in premature mortality and morbidity related to ambient $PM_{2.5}$ concentrations, with the strongest evidence being reported for cardiovascular-related effects.
- As $PM_{2.5}$ is a ubiquitous pollutant, exposure to ambient $PM_{2.5}$ concentrations can have a substantial public health impact.
- The available scientific evidence does not identify a population-level threshold below which $PM_{2.5}$-related health effects do not occur; recent evidence indicates that incremental benefits are observable even at relatively low concentrations. Therefore further reductions in ambient $PM_{2.5}$ concentrations in both Canada and the U.S. can be expected to yield population health benefits.
- Modelled depositions of acidifying sulphur and nitrogen in Canada and the U.S. indicate exceedances of thresholds (termed critical loads) in some geographic areas that pose a risk of harmful effects to terrestrial and aquatic ecosystems.
- $PM_{2.5}$ contributes to visibility impairment in both Canada and the U.S. In Canada, an updated assessment of visibility impairment due to atmospheric $PM_{2.5}$ indicated that the poorest visibility occurs primarily in highly populated regions of southern Ontario and Quebec. Between 2000–2004 and 2005–2009, visibility in the U.S. on the 20% haziest days improved at Class I areas in the Northeast and Pacific Northwest, but not in the Midwest and Montana.
- Existing Canada-Wide Standards (CWS) for fine PM to protect human and ecosystem health have been replaced by new and more stringent Canadian Ambient Air Quality Standards (CAAQS), which also include an annual $PM_{2.5}$ standard.
- In December 2012, the U.S. Environmental Protection Agency (EPA) completed a review of the National Ambient Air Quality Standards (NAAQS) for PM and revised the suite of primary (health-based) $PM_{2.5}$ standards to provide increased protection against health effects associated with long- and short-term fine particle exposures, including premature mortality, increased hospital admissions and emergency department visits, and development of chronic respiratory disease.

2.1 Effects of $PM_{2.5}$ on Human Health

Since the late 1990s, a significant research investment has been made in understanding the human health impacts associated with exposures to ambient PM. As a result, evidence has continued to accumulate about adverse health effects associated with PM exposures. The main focus of research and monitoring activities has been on improving our understanding of the impacts of fine particles (largely indexed by $PM_{2.5}$) on human health. This evidence has been reviewed, summarized and evaluated by national and international agencies (EPA, 2009b; GoC, 2012b). Overall, the evidence indicates that $PM_{2.5}$ contributes measurably to both morbidity and premature mortality. Epidemiological studies have produced evidence of effects associated with both long- and short-term $PM_{2.5}$ exposures at the population level. Toxicological and controlled human exposure studies have contributed to a better understanding of the mechanistic basis for many of these outcomes, thereby providing coherence and biological plausibility for the effects observed in epidemiological studies.

Since the 2004 Assessment, a number of large multi-city epidemiological studies have been conducted, including extended analyses of the seminal studies of long-term $PM_{2.5}$ exposures (i.e. the American Cancer Society and Harvard Six Cities studies), new long-term cohort studies (Crouse et al., 2012), and several new multi-city time-series studies that used uniform methodologies to investigate the effects of short-term $PM_{2.5}$ exposures (Dominici, 2007; Franklin, 2008; Krewski, 2009). Collectively, these studies provide strong evidence of consistent increases in morbidity and/or premature mortality related to ambient $PM_{2.5}$ exposures. The strongest evidence comes from recent studies investigating long-term exposure to $PM_{2.5}$ and cardiovascular-related effects, including heart attacks, congestive heart failure, stroke, and premature mortality. This evidence is coherent with studies of short-term exposure to $PM_{2.5}$ that have observed associations with a continuum of effects. These ranged from subtle changes in indicators of cardiovascular health (heart-rate variability perturbations, vasodilation disturbance, blood pressure alteration) to serious clinical events, such as increased emergency department visits, hospitalizations and mortality (Brook et al., 2010).

In addition, recent research also provides stronger evidence of respiratory-related morbidity effects associated with long- and short-term $PM_{2.5}$ exposures. Such effects include decrements in lung function growth, increases in respiratory symptoms, development of asthma, and increased respiratory-related hospital admissions and emergency department visits (Rückerl et al., 2011). More limited scientific evidence is available for a broader range of health effects, including adverse birth outcomes, cancer, and neurological effects (Rückerl et al., 2011).

Extensive efforts have been made over the last decade to examine the effects that different sources of $PM_{2.5}$ and its chemical composition have on human health. The currently available scientific information continues to provide indications that many different components of the fine particle mixture, as well as the groups of components associated with specific source categories of fine particles, are linked to adverse health effects. However, the scientific evidence is not yet sufficient to allow differentiation of those components or sources that are more closely related to specific health outcomes, or to exclude any component or group of components from the mix of fine particles included in the $PM_{2.5}$ indicator (Lippmann et al., 2013).

With regard to the quantitative relationship linking $PM_{2.5}$ concentrations to adverse health effects (the concentration–response function), the currently available scientific information supports the use of a no-threshold, log-linear model . In other words, the effects of $PM_{2.5}$ are generally proportional to its concentration. Health effects may occur over the full range of concentrations observed in epidemiological studies of long- and short-term $PM_{2.5}$ exposures, since no population-level thresholds (below which it can be concluded with confidence that $PM_{2.5}$ - related effects do not occur) can be discerned from the available scientific evidence (Correia et al., 2013; Crouse et al., 2012). This relationship is of considerable significance for risk management because of its prediction of health benefits associated with air quality improvements focusing on $PM_{2.5}$.

As $PM_{2.5}$ is a ubiquitous pollutant, all Canadians and Americans are at risk of a $PM_{2.5}$ - related health effect. In addition, specific groups within the general population are at increased risk: children, older adults, individuals with pre-existing heart and lung disease, and people who live in poverty. There is emerging, though still limited, evidence for additional subpopulations being potentially at increased risk, such as diabetics, the obese, pregnant women, and the developing fetus. Taken together, this suggests that exposure to ambient $PM_{2.5}$ concentrations can have substantial public health impacts.

With regard to understanding the nature and magnitude of $PM_{2.5}$-related risks, epidemiological studies evaluating health effects associated with long- and short-term $PM_{2.5}$ exposures have reported variations in the concentration–response function both within and between cities and in geographic regions across the U.S. (Franklin et al., 2006; EPA, 2009b; Zanobetti and Schwartz, 2009). This heterogeneity may be attributed, in part, to differences in the fine particle composition as well as to exposure measurement error (Jerrett et al. 2005; EPA, 2009b). Furthermore, while there is clear evidence that $PM_{2.5}$ affects human health, there is growing awareness of the importance of improving our understanding of the impacts of $PM_{2.5}$ and its components within the broader ambient mixture, which includes other particle fractions and gaseous co-pollutants.

Public health research studies and assessments have found substantial health benefits from lower ambient $PM_{2.5}$ concentrations that have been forecast, modelled, or observed as a result of emission reduction programs. For example, reductions in ambient $PM_{2.5}$ concentrations have been correlated with increased life expectancy (Pope et al., 2009; Correia et al., 2013). Recent analyses indicate that despite significant improvements in air quality, recent $PM_{2.5}$ concentrations measured in North America still pose a risk to public health (Schindler et al., 2009; Fann et al., 2012; Lepeule et al., 2012; Correia et al., 2013). Furthermore, in the recent Global Burden of Disease study (Lim et al., 2012), urban air pollution (primarily particulate air pollution) was characterized as one of the top public health risk factors in both the developed and developing worlds.

Overall, there is substantial evidence to indicate that $PM_{2.5}$ poses a significant human health risk. Despite recent reductions in $PM_{2.5}$ due to regulatory measures, a health burden remains associated with recent ambient concentrations of this pollutant, and public health would benefit from further $PM_{2.5}$ reductions. Furthermore, future health-related research, model development, and data collection activities will improve our understanding of $PM_{2.5}$-related health impacts.

2.2 Effects of $PM_{2.5}$ and Its Precursors on Ecosystem Health

2.2.1 Critical loads of acidity for forest and aquatic ecosystems in Canada

The impact of anthropogenic sulphur and nitrogen emissions on terrestrial and aquatic ecosystems is widely assessed using a critical load approach; moreover, critical loads underpin the Canada-Wide Acid Rain Strategy for Post-2000. Acidic deposition and fine PM are closely linked; both are strongly affected by the emissions of gaseous SO_2 and NO_x and their secondary products (particle-bound SO_4^{2-} and NO_3^-), and these atmospheric pollutants have been linked to ecosystem damage in Canada (Jeffries et al., 1999).

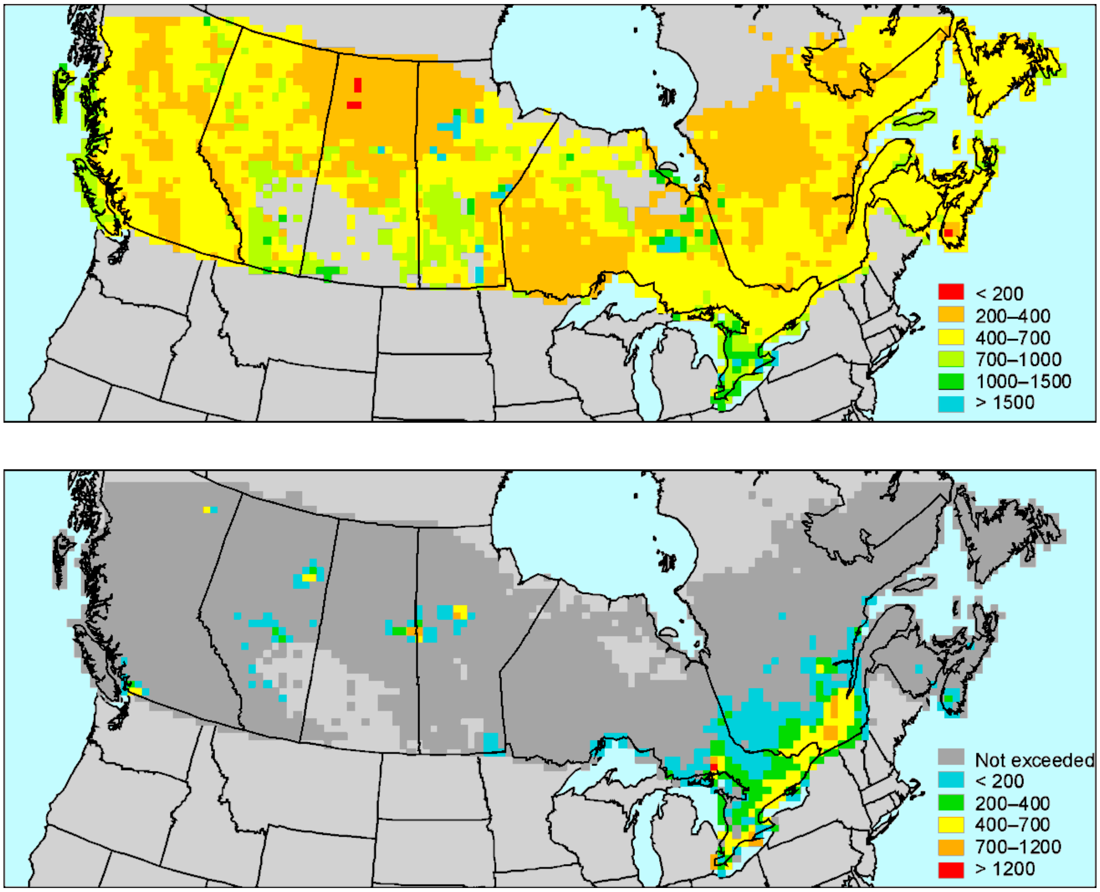
The critical load approach is based on setting a critical chemical limit to protect a specific biological indicator for a chosen receptor ecosystem (e.g. fish species for surface waters), and via inverse modelling, estimating a deposition load (the critical load) to ensure that the limit is not violated, thus avoiding “harmful effects.” If deposition of sulphur or nitrogen is above its critical load, it is said to be “exceeded” and there is a high risk of harmful effects to ecosystem health. The methods are well established and widely used. For example, the Steady-State Mass Balance model has been used to estimate critical loads of acidifying sulphur and nitrogen deposition for forest soils from plot to regional scales (Mongeon et al., 2010; Whitfield et al., 2010).

A base cation to aluminum ratio (Bc:Al) of 10 in soil solution has been widely used in Canada as a critical limit to protect long-term nutrient pools in forested mineral soils (Ouimet et al., 2006). In concert, an acid neutralizing capacity of 10 µmol of charge/L in excess of natural organic acidity (following Lydersen et al., 2004) has been widely used in Canada to protect aquatic species (Jeffries et al., 2010; Strang et al., 2010).

The critical loads of sulphur and nitrogen acidity for forested mineral soils (mapped for an area of 395,361 km^2) and lakes (n = 4694) are summarized as the area-weighted 5th percentile of critical loads on a deposition grid (42 km × 42 km) developed by AURAMS (A Unified Regional Air Quality Modeling System) (Figure 2.2.1). These critical load results show a range in ecosystem sensitivity, with lower critical loads centred on northern and southwestern Quebec, northwestern Ontario, northern Saskatchewan and the coastal mountain range of British Columbia. The most sensitive areas (< 100 mol of charge/ha/year; red grid squares in the upper panel of Figure 2.2.1) are driven by low critical loads of acidity for surface waters.

Critical loads are exceeded across Canada, based on 2006 AURAMS-modelled sulphur and nitrogen deposition (“hot” coloured, positive exceedance squares in the lower panel of Figure 2.2.1). The highest levels of exceedance are observed in southern Ontario and Quebec in eastern Canada, and in regions with large point sources (e.g. oil and gas, and smelter industries) or large urban centres (e.g. Vancouver, B.C.) in western Canada. Approximately 18% of the mapped grid area receives acidic deposition in excess of the critical load under 2006 deposition estimates; however, the extent of exceedance is strongly dependent on the critical chemical limit (the specified ecosystem protection level). The extent of exceedance is reduced to approximately 5% of the mapped grids (not shown) when a Bc:Al ratio of one is used. This lower Bc:Al ratio is the most widely used chemical limit protecting tree roots from damage (UBA, 2004).

Figure 2.2.1: Upper panel: the 5th percentile of critical loads of acidity (molc ha–1 a–1) for forest (mineral) soils (395,361 km2) and lakes (n = 2868) displayed on the AURAMS grid (42 km × 42 km); lower panel: exceedance of critical loads of acidity (molc ha–1 a–1) under 2006 AURAMS sulphur and nitrogen deposition



Note: During 2010, Hudson Bay Mining and Smelting closed their smelter in Flin Flon, on the border of Manitoba and Saskatchewan, the large-point source of sulphur emissions drives exceedance under the 2006 deposition.

2.2.2 The impact of PM on ecosystem health in the United States

Acid deposition (acid rain) resulting from SO_2 and NO_x emissions negatively affects the health of U.S. lakes and streams. The 2011 National Acid Precipitation Assessment Program Report to Congress noted that the Acid Rain Program (ARP) has been successful in reducing emissions of SO_2 and NO_x from power generation; as a result, acidic deposition has decreased to the extent that some acid-sensitive lakes and streams are beginning to recover from acidification. However, the report also indicated that current emission-reduction levels are not sufficient to allow full recovery of acid-sensitive ecosystems.

Like Canada, the EPA uses critical loads as a useful metric to help quantify the potential aquatic ecological benefits that have resulted from emission reduction programs such as the ARP. In a recent analysis (EPA, 2013b), critical loads were estimated for more than 6,400 lakes and streams using the Steady-State Water Chemistry model. This analysis incorporates lakes and streams where surface water samples have been collected through programs such as the National Surface Water Survey, the Environmental Monitoring and Assessment Program, the Wadeable Stream Assessment, the National Lake Assessment, the Temporally Integrated Monitoring of Ecosystems and Long-term Monitoring programs, and other water quality programs. The lakes and streams associated with these programs consist of a subset of those that are located in areas affected by acid deposition, but are not intended to represent all water bodies in the eastern half of the U.S.

For this analysis, the critical load represented the combined deposition loads of sulphur and nitrogen to which a lake or stream could be subjected and still maintain an acid-neutralizing capacity of 50 µmol of charge/L or greater, an indicator that the lake or stream is healthy enough to protect most fish and other aquatic organisms. Critical loads of combined total sulphur and nitrogen deposition are expressed in terms of ionic charge balance as milliequivalents per square metre per year (meq/m^2/yr).

To assess the extent to which regional lake and stream ecosystems are protected, this analysis compared the critical loads to the measured deposition for the period before implementation of the 1990 *Clean Air Act* amendments (1989–1991) and for a recent post-implementation period (2008–2011). The percentage of water bodies examined that were receiving levels of combined sulphur and nitrogen deposition exceeding the critical load decreased from 42% in 1989–1991 to 23% in 2009–2011 (Figure 2.2.2).

Many areas still have numerous lakes and streams affected by acid rain. The largest concentration of water bodies where acidic deposition currently exceeds estimated critical loads includes the southern Adirondack mountain region in New York, southern New Hampshire, Cape Cod in Massachusetts, and along the Appalachian Mountain spine from Pennsylvania to North Carolina. The Upper Midwest also contains lakes that receive acidic deposition above their critical loads, but many may be naturally acidic and not responsive to changes in acidic deposition.

Figure 2.2.2: Lake and stream exceedances of estimated critical loads for total nitrogen and sulphur deposition in the eastern U.S. between 1989–1991 and 2009–2011

(Source: EPA 2011 Progress Report: Clean Air Interstate Rule, Acid Rain Program, and Former NO_x Budget Trading Program)

2.2.3 Visibility

Visibility is a measure of how clearly scenic vistas and landscape features can be perceived at great distances, a factor particularly important for visitors to national parks and wilderness areas. Visibility is degraded when sunlight is scattered or absorbed by fine particles or gases in the atmosphere. Light scattering reduces the clarity, colour, and contrast that can be discerned by the human eye. Visibility impairment, or haze, is caused primarily by fine particles of $(NH_4)_2SO_4$, NH_4NO_3, organic carbon (OC), elemental carbon (EC), soil, and sea salt. Many of these pollutants are not emitted directly into the atmosphere, but are formed by secondary reactions with other pollutants. SO_4^{-2} and NO_3^- particles are secondary pollutants formed from SO_2 and NO_X emissions, primarily from fossil fuel combustion. Oil and gas development, agriculture, vegetation, and fire also are sources of NO_X emissions. OC and EC are emitted by natural sources such as wildfire and vegetation and by anthropogenic sources such as fossil fuel combustion, prescribed and agricultural fires, and residential wood burning. Dust from agriculture, construction, roadways, and land surfaces contributes to fine and coarse soil particles. Sea salt contributes to haze in coastal areas.

In the U.S., the term regional haze describes the cumulative visibility impact from numerous air emission sources across a broad geographic region. The 1977 amendments to the *Clean Air Act* set a national visibility goal to prevent "any future, and remedy any existing, impairment of visibility in mandatory Class I Federal areas" that results from human-made air pollution. The Regional Haze Rule requires states to demonstrate progress in returning visibility in Class I areas on the 20% haziest days of the year to natural conditions by 2064 and to ensure no degradation in visibility for the 20% clearest days of the year. States are required to establish baseline visibility for the 2000–2004 period, develop long-term strategies to improve visibility, and review progress every five years. States worked through one of five regional planning organizations to develop the technical analyses necessary to evaluate source contributions to haze and emissions management strategies.

The Interagency Monitoring of Protected Visual Environments (IMPROVE) visibility monitoring network measures fine-particle concentrations at U.S. Class I national parks and wilderness areas. Currently there are 110 monitoring sites at Class I areas, plus additional monitors that follow the IMPROVE protocols. Filter samples of fine particles are collected for 24 hours every third day. Methods, sample analyses, and data are available on the IMPROVE website (http://vista.cira.colostate.edu/improve/). Light extinction due to each pollutant is calculated separately. The visibility metric used to track progress is the deciview, which is a logarithmic scale calculated from total light extinction. A one-deciview change is perceptible by the human eye. A decrease in the deciview metric indicates an improvement in visibility.

To evaluate visibility effects due to transboundary transport of pollutants along the Canada – U.S. border, IMPROVE data for 2000–2010 were reviewed for Class I areas near the border to assess pollutant contributions to haze and visibility trends for the 20% haziest and 20% clearest days in each year. In 2012, the most recent year for which data are available, $(NH_4)_2SO_4$ was the major contributor on the 20% haziest days in the Northeast, the Upper Midwest, and North Dakota, as illustrated in Figure 2.2.3. In Montana, Idaho, and Washington, OC mass, primarily due to wildfire, was the dominant contributor on the 20% haziest days in 2012 (Figure 2.2.3), and to a lesser extent in several other years. NH_4NO_3 is an important contributor to the 20% haziest days in 2012 in the Upper Midwest, the central states, and North Dakota, eastern Montana, and California. SO_2 emissions from coal-fired electric utilities and industries are the major contributors to $(NH_4)_2SO_4$. NO_x emissions that form NH_4NO_3 are primarily attributable to fossil fuel combustion (e.g. highway vehicles, off-road equipment, electric utilities, industry) and oil and gas production. Agriculture, fire, and biogenic sources also emit NO_x, while agricultural sources are primarily responsible for NH_3 emissions.

Visibility on the 20% haziest days improved across the eastern U.S. between 2000 and 2012 (Figure 2.2.4), largely due to reductions in SO_2 emissions from electric utilities under federal and state regulatory requirements and the shift from coal to natural gas. Visibility improvement on the 20% haziest days was much smaller in the Upper Midwest and Northwest, and degradation was observed at sites in North Dakota and Montana. NH_4NO_3 is increasing in the central states, while OC mass is increasing at several western Class I locations. In contrast, visibility on the 20% clearest days is improving across the U.S. (Figure 2.2.5).

In the western states, episodic wildfire events are highly variable spatially and temporally, which complicates interpretation of trends for the 20% haziest days. The annual mean contributions of $(NH_4)_2SO_4$ and NH_4NO_3 to light extinction better reflect trends that are directly attributable to changes in anthropogenic emissions. National maps of annual mean light extinction for $(NH_4)_2SO_4$ (Figure 2.2.6) and NH_4NO_3 (Figure 2.2.7) further demonstrate significant reductions in light extinction in the eastern U.S. that correspond to emission reductions from utilities and mobile sources.

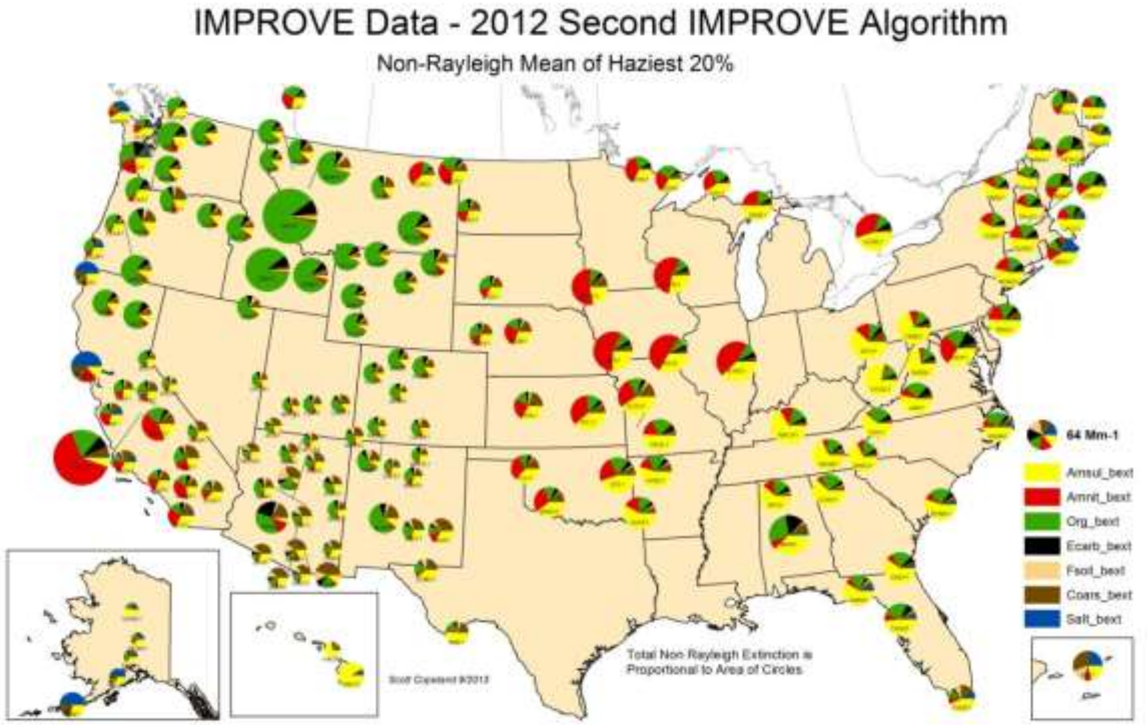
Slight increases from 2000 to 2012 in annual mean light extinction in western North Dakota and eastern Montana likely reflect the contributions of increasing NH_3 emissions from agricultural sources, as well as SO_2 and NO_x emissions from oil and gas production and associated activities in North Dakota's Bakken fields. Hand and colleagues (2012) found that wintertime concentrations of NH_4NO_3 and $(NH_4)_2SO_4$ are increasing in North Dakota, Montana, and the central states; they suggested that long-distance transport from the oil sands operations in Alberta could also be contributing to these trends.

PM also impairs visibility in some parts of Canada and is a concern due to the loss of scenic vistas and the contribution of transboundary transport to this loss. As part of Environment and Climate Change Canada's National Visibility Pilot Study, visibility is being monitored at Barrier Lake, AB, Wolfville, NS, and Abbotsford, B.C. The Barrier Lake site is operated by Environment and Climate Change Canada using the U.S. IMPROVE protocol and includes both aerosol and optical measurements. Data collected in 2011 from Barrier Lake (Figures 2.2.7(a) and (b)) show that the location has very good visibility, typical of a remote site.

Visibility conditions in Canada have been updated since the 2004 Assessment. The data primarily available for urban sites across Canada are for the 2003–2010 period and are part of the National Air Pollution Surveillance (NAPS) PM speciation network (Figures 2.2.8 and 2.2.9). Reconstructed visibility from these measurements indicates that visibility is reduced more in highly populated regions of southern Ontario and Quebec than in sites closer to the east and west coasts (Figure 2.2.10).

One exception is Golden, B.C., a small community in the Canadian Rocky Mountains that has relatively poor visibility. This location is heavily influenced by winter heating emissions and valley inversions. Overall, the spatial visibility pattern may be attributed to the dominance of $(NH_4)_2SO_4$ at all eastern Canadian sites, compared with much lower levels of $(NH_4)_2SO_4$ at sites in the west. Visibility impairment at western sites is generally dominated by organic matter, with the exception of Abbotsford, B.C., and Edmonton, AB, where NH_4NO_3 is the main species responsible for reducing visibility.

Figure 2.2.3: Contributions to light extinction on the 20% haziest days in 2012



Note: Sources are ammonium sulphate (Amsul_bext), ammonium nitrate (Amnit_bext), organic carbon mass (Org_bext), elemental carbon (Ecarb_bext), fine soil (Fsoil_bext), coarse particles (Coars_bext), and sea salt (Salt_bext)

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Figure 2.2.4: Visibility trends for the 20% haziest days between 2000 and 2012 (percentage change in deciview (dv))

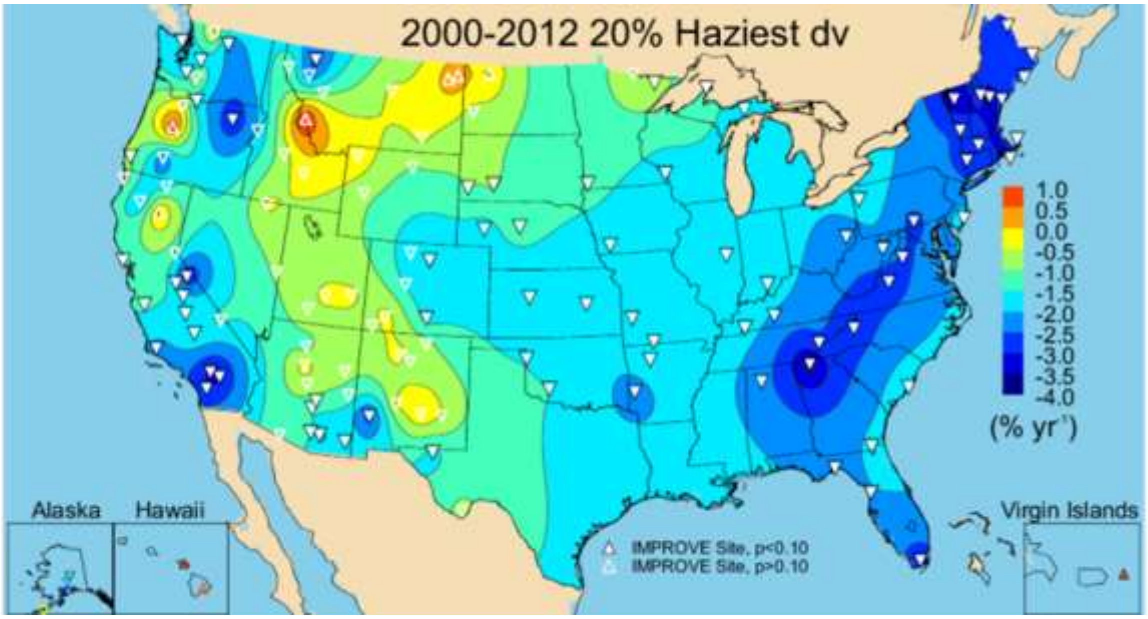

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Figure 2.2.5: Visibility trends for the 20% clearest days between 2000 and 2012 (percentage change in deciview (dv))

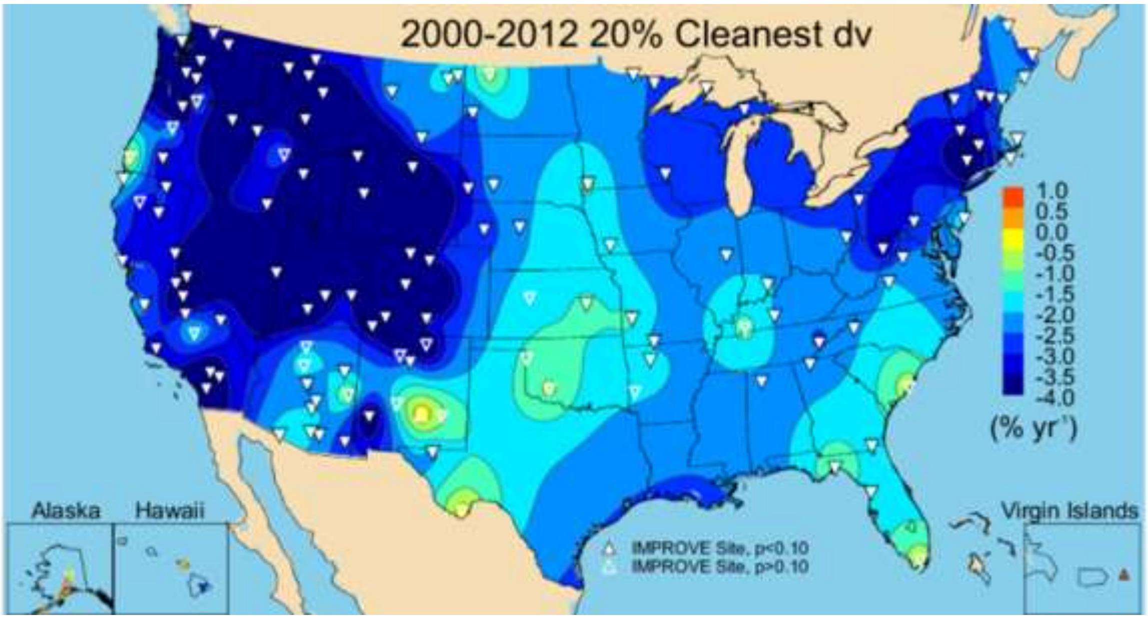

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Figure 2.2.6: Trends in annual mean light extinction due to ammonium sulphate between 2000 and 2012

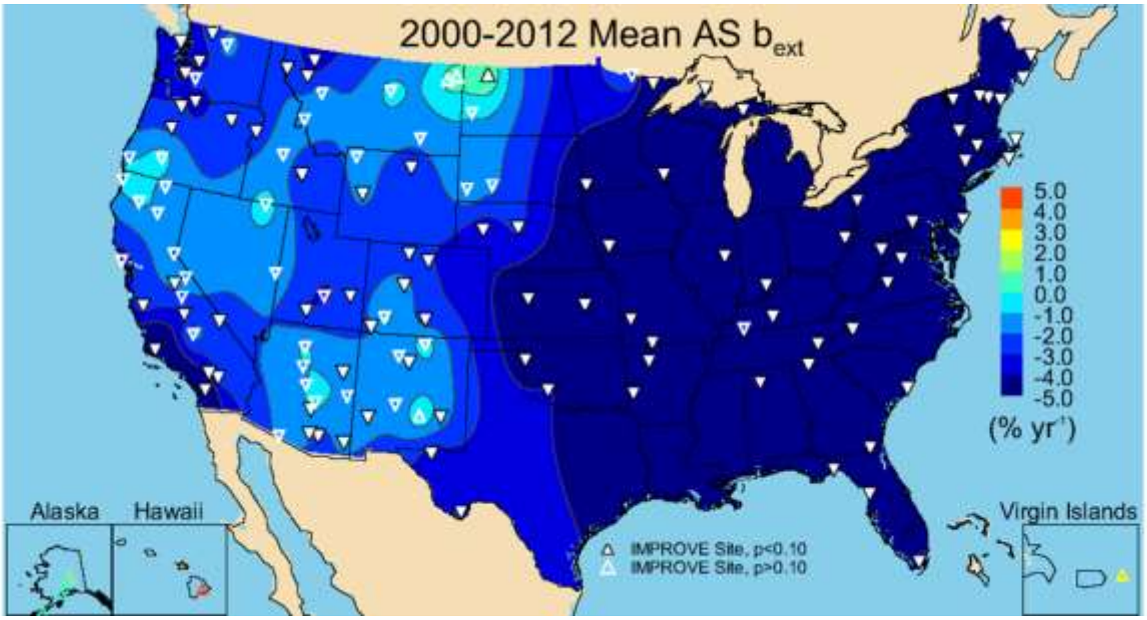

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Figure 2.2.7: Trends in annual mean light extinction due to ammonium nitrate between 2000 and 2012

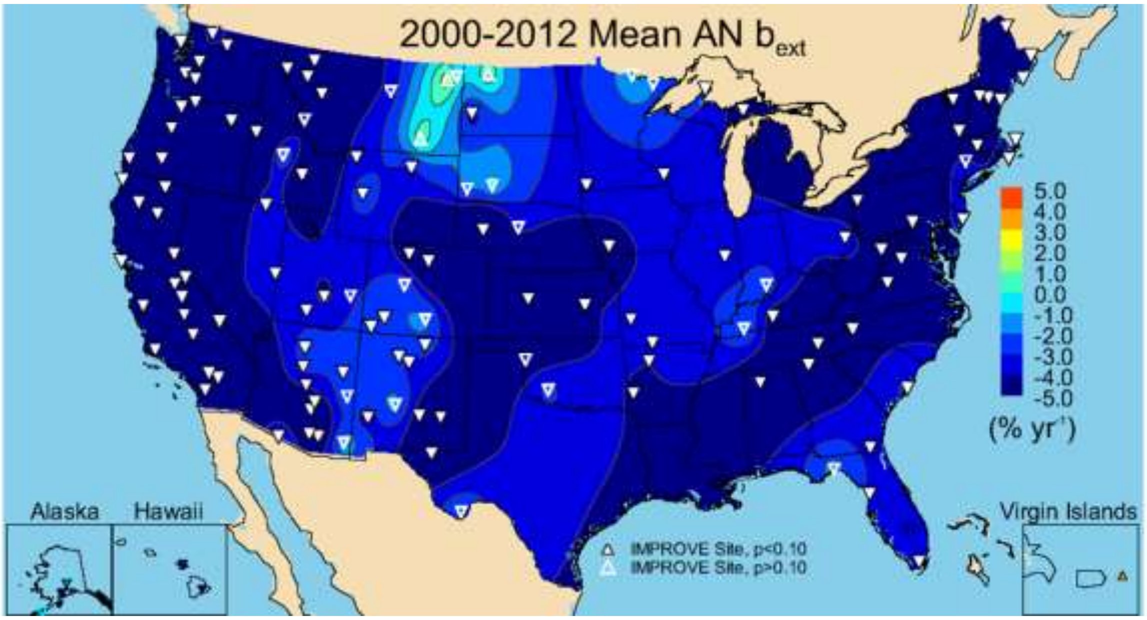

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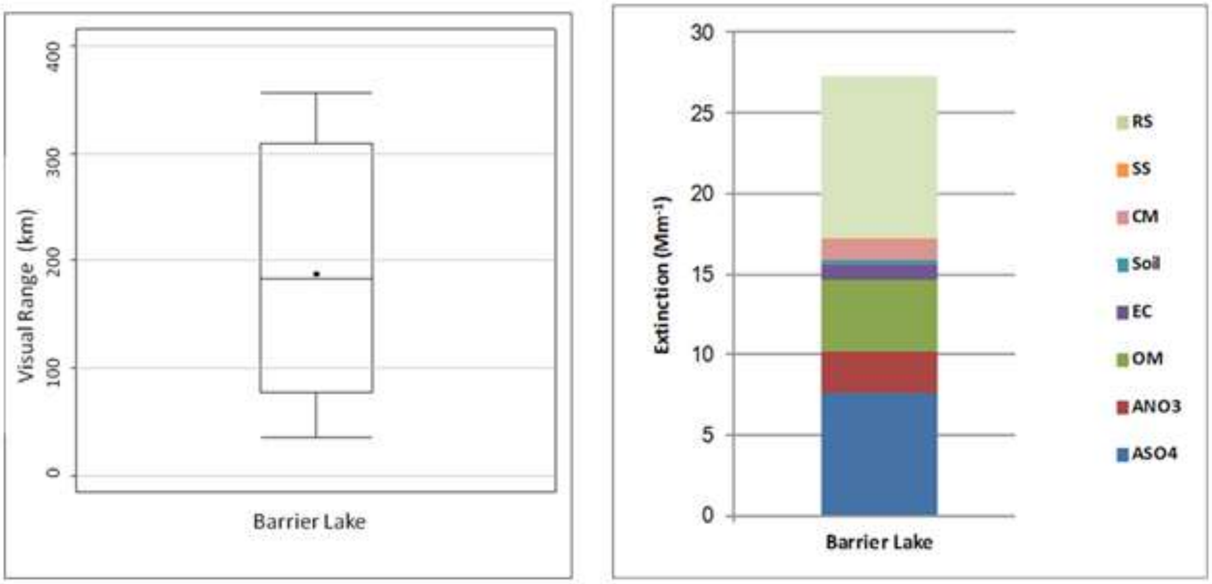
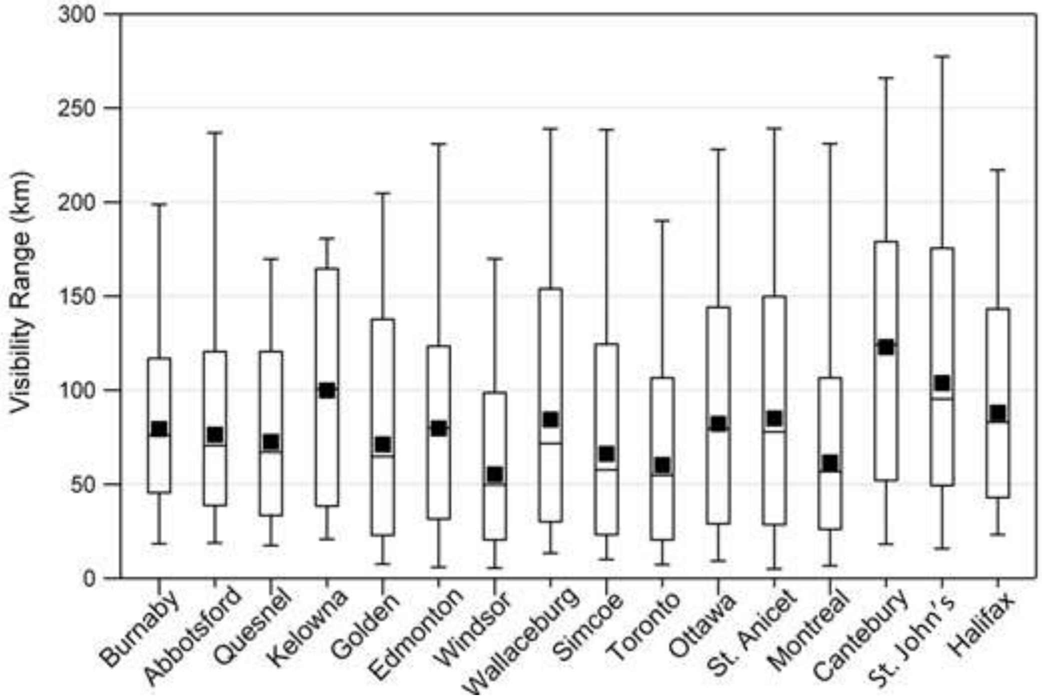
Figure 2.2.8: (a) Left: reconstructed visual range at Barrier Lake, AB, from January to December 2011; (b) Right: average contribution of PM components to extinction



Note: (a) whiskers = maximum and minimum values; box = 10th and 90th percentiles; line = median; point = mean; (b) ASO4 = ammonium sulphate; ANO3 = ammonium nitrate; OM = organic mass; EC = elemental carbon; Soil = fine soil; CM = coarse mass; SS = sea salt; RS = Rayleigh scattering

* * * * *

Figure 2.2.9: The reconstructed visual range (km) at 16 NAPS sites, presented from west to east in Canada



Note: whiskers = maximum and minimum values; box = 10th and 90th percentiles; line = median; point = mean

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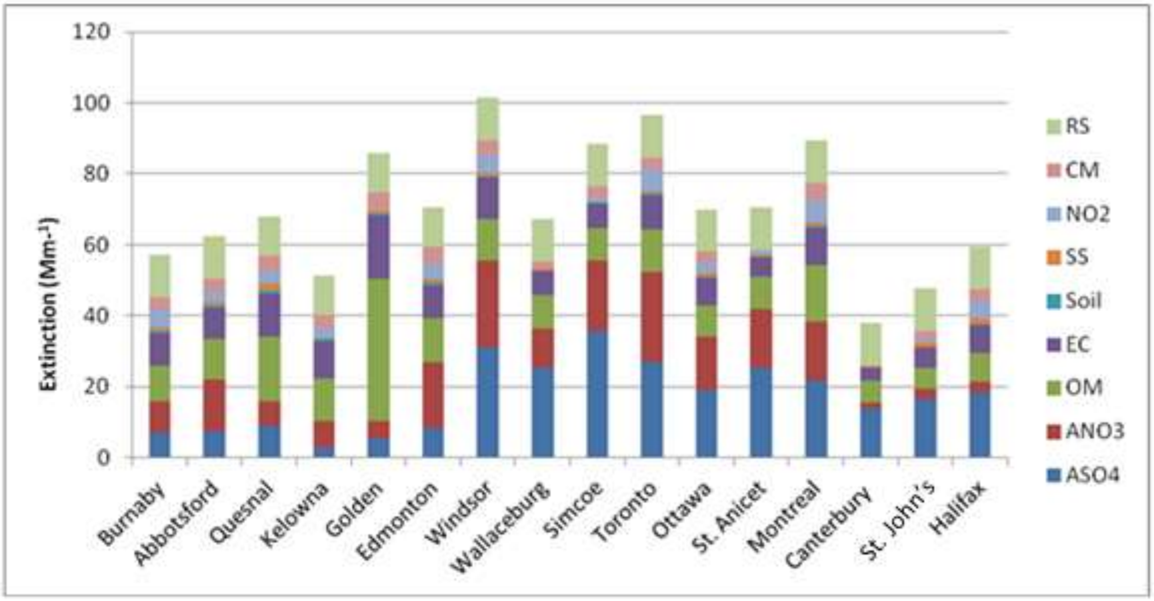
Figure 2.2.10: Average contribution of PM components to reconstructed light extinction at 16 NAPS sites, presented from west to east in Canada



Note: ASO4 = ammonium sulphate; ANO3 = ammonium nitrate; OM = organic mass; EC = elemental carbon; Soil = fine soil; SS = sea salt; CM = coarse mass; NO2 = nitrogen dioxide; RS = Rayleigh scattering

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2.3 Current Air Quality Standards

Both Canada and the U.S. have established air quality standards to protect human and ecosystem health. Each standard is defined in terms of four basic elements: indicator,[1] averaging time, form,[2] and level. These elements must be considered collectively in evaluating the health and welfare protection afforded by the PM standards.

For Canada, the Canadian Council of Ministers of the Environment (CCME) endorsed the CAAQS for $PM_{2.5}$ and ground-level O_3 in October 2012. The CAAQS replace the CWS and are established as objectives by the Government of Canada using the authority of the *Canadian Environmental Protection Act, 1999*. The CAAQS are more stringent than the existing CWS and also more comprehensive, with the inclusion of an annual average standard in addition to the 24-h standard for $PM_{2.5}$. This new annual $PM_{2.5}$ standard has been established to provide an additional level of protection for human and environmental health.

In the U.S., the EPA initially established primary (health-based) and secondary (welfare-based) NAAQS for PM under section 109 of the *Clean Air Act* in 1971. In its most recent review, the EPA revised the primary annual $PM_{2.5}$ standard by lowering the level from 15.0 $\mu g/m^3$ to 12.0 $\mu g/m^3$ to provide increased protection against the health effects associated with long- and short-term $PM_{2.5}$ exposures. The EPA also eliminated the spatial averaging provisions[3] as part of the form of the annual standard to avoid potential disproportionate impacts on at-risk populations. The agency retained the level (35 $\mu g/m^3$) and form (98th percentile) of the primary 24-h standard to continue to provide supplemental protection against health effects associated with short-term $PM_{2.5}$ exposures.

With regard to secondary standards, the EPA generally retained the annual and 24-h $PM_{2.5}$ standards and revised only the form of the secondary annual $PM_{2.5}$ standard to remove the option for spatial averaging, consistent with the revised primary annual $PM_{2.5}$ standard. PM-related visibility impairment is addressed by the secondary 24-h $PM_{2.5}$ standard, and non-visibility welfare effects (e.g. on ecosystems, materials, or climate) are addressed by both the annual and 24-h $PM_{2.5}$ standards.

The statistical forms of the comparable standards in Table 2.3.1 are identical, which allows for a direct comparison of air quality between the two countries. The calculation and evaluation procedures for the CAAQS are described in the *Guidance Document on Achievement Determination* (CCME, 2012). For the NAAQS, the Code of Federal Regulations, Title 40, Appendix N, part 50 describes the computations necessary for determining when the $PM_{2.5}$ standards are met and also addresses which measurement data are appropriate for comparison to the standards.[4]

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[1] The "indicator" of a standard defines the pollutant that is to be measured in determining whether an area attains the standard.

[2] The "form" of a standard defines the air quality statistic that is to be taken for the appropriate averaging time and compared to the level of the standard in determining whether an area attains the standard.

[3] The form of the initial 1997 $PM_{2.5}$ standard allowed the level of the standard to be compared to average measurements from multiple community-wide air quality monitoring sites (i.e. "spatial averaging") if specific requirements were satisfied. In 2006, the EPA tightened the constraints on the spatial averaging criteria by further limiting the conditions under which some areas may average measurements (71 FR 61165 to 61167, October 17, 2006) and eliminated this provision in 2013 (78 FR 3124 to 3127, January 15, 2013).

[4] See http://www.ecfr.gov/cgi-bin/text-idx?c=ecfr&SID=9d147c97512e7070f06997132251362e&rgn=div9&view=text&node=40:2.0.1.1.1.0.1.19.15&idno=40.

Table 2.3.1: Current ambient air quality standards for PM2.5 in Canada and the United States

		24-hour ($\mu g/m^3$)	**Annual ($\mu g/m^3$)**
Canada			
Canada-Wide Standards (CWS)		30[1]	None
Canadian Ambient Air Quality Standards (CAAQS)[2]	2015	28[1]	10.0[3]
	2020	27[1]	8.8[3]
United States			
Primary National Ambient Air Quality Standards (NAAQS)		35[1]	12[3]
Secondary National Ambient Air Quality Standards (NAAQS)		35[1]	15[3]

(1) The form of the standard is the 3-year average of the annual 98th percentiles of the daily 24-hour average PM$_{2.5}$ concentrations.

(2) The 2020 standards will be reviewed in 2015.

(3) The form of the standard is the 3-year average of the annual average of daily 24-hour PM$_{2.5}$ concentrations.

CHAPTER 3: CONCENTRATIONS AND TRENDS OF $PM_{2.5}$ AND CHEMICAL COMPONENTS IN CANADA AND THE UNITED STATES

Chapter Summary

- Over much of the U.S. and eastern Canada, ambient concentrations of $PM_{2.5}$ have decreased from the levels reported in the 2004 Assessment. A notable exception is the increase in winter levels of $PM_{2.5}$ over parts of western Canada.
- Long-term declines in levels of ambient SO_2, particulate SO_4^{2-}, and particulate NH_4^+ were observed in eastern North America, contributing to the declining trend of observed $PM_{2.5}$. In contrast, levels of particulate NO_3^- were more variable, with declines in some regions and no change or increases in others.
- Measurements of chemical composition indicate that $(NH_4)_2SO_4$ and NH_4NO_3, together with organic matter and EC, comprise most of the $PM_{2.5}$ mass at speciation sites across Canada and the U.S.
- In eastern Canada, the Northeast, Southeast, and Midwest U.S., particularly in the warmer months, $(NH4)_2SO_4$ is the largest contributor to $PM_{2.5}$ mass. This contribution increases during high $PM_{2.5}$ episodes in the summer. NO_3^- is an important contributor to $PM_{2.5}$ in the Northwest, North Central, and Midwest regions of the U.S. and in many parts of Canada during the cooler months.

3.1 Ambient Concentrations and Spatial-Temporal Distributions

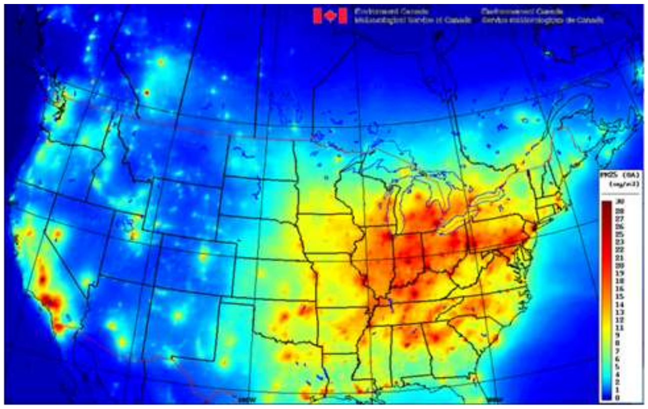
Ambient concentrations of $PM_{2.5}$ in North America have declined in many areas since the 2004 Assessment, mostly due to emission reductions and other pollution controls (as described in Chapter 4). Both the levels and chemical composition of PM have changed in the last decade, and this section presents the temporal and geographic trends that have occurred in mass and in $PM_{2.5}$ chemical components.

3.1.1 Spatial patterns and trends in $PM_{2.5}$ across North America

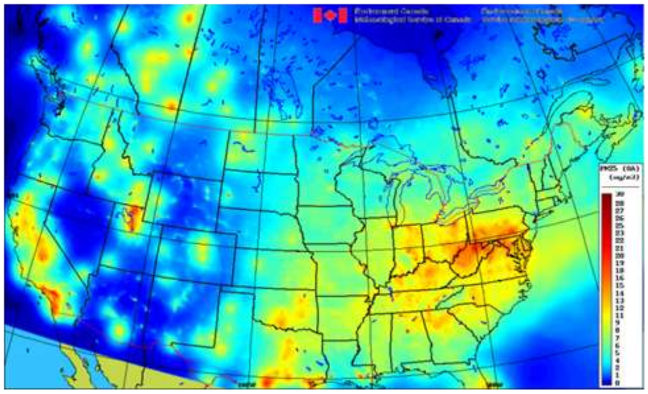
Spatially resolved maps of $PM_{2.5}$ across Canada and the U.S. for summer 2005 and summer 2012 are shown in Figures 3.1.1 (a) and (b), respectively. These maps make use of a methodology (Robichaud and Ménard, 2014), new since the 2004 Assessment, that combines air quality models and observational data. This approach provides more detailed insight into spatial variations in $PM_{2.5}$ than is possible with maps that rely on monitoring data alone. The addition of data from the model compensates for the limited spatial coverage of monitoring in both countries. Briefly, the method uses an optimal interpolation technique adapted to air pollutants that blends (by linear combination) the modelled background field (a Canadian air quality forecast model) with the U.S. EPA AIRNow system's observations of $PM_{2.5}$, plus those of extra Canadian stations monitoring $PM_{2.5}$. The model and observations are first blended hour-by-hour and then averaged across the summer months (June, July, and August). The CHRONOS model, which was used operationally in Canada until 2009, was used for the 2005 map; the Global Environmental Multi-scale–Modelling Air Quality and Chemistry (GEM-MACH) model, which replaced CHRONOS, was used to produce the 2012 map.

Figure 3.1.1: Model and observation objective analysis of North American $PM_{2.5}$ distributions for (a) 2005 and (b) 2012 in summer; and (c) the difference between the two years

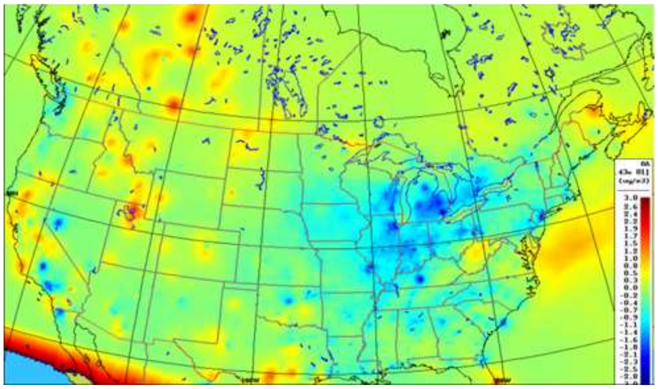
(a) JJA 2005



(b) JJA 2012



(c) Difference 2012 minus 2005



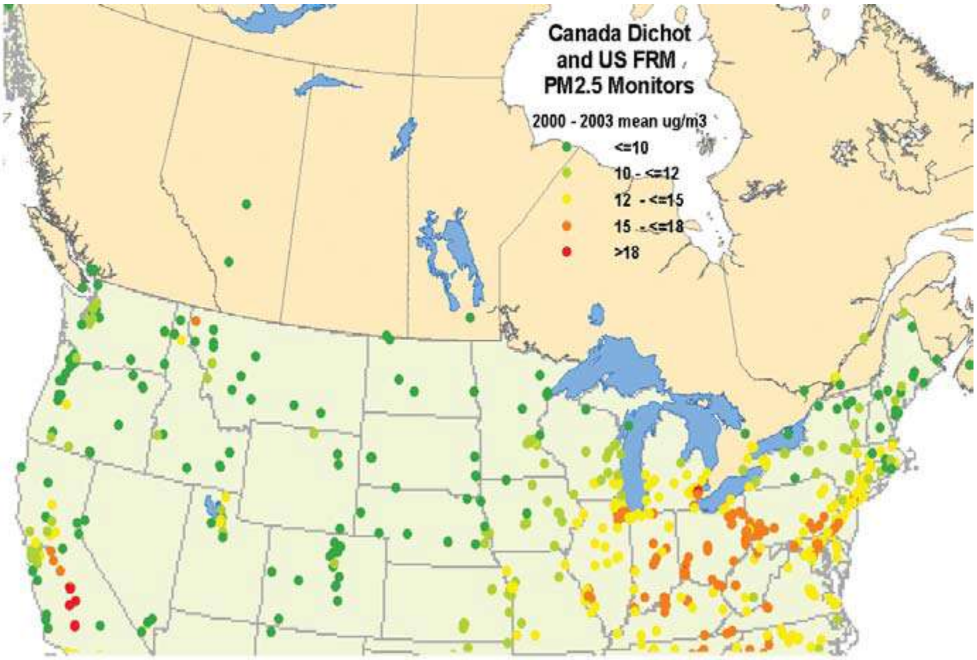
Lower PM concentrations are evident in eastern North America in 2012 (Figure 3.1.1 (a)) relative to 2005 (Figure 3.1.1.(b)). More importantly, the figures provide greater spatial detail than was available for the 2004 Assessment. The highest $PM_{2.5}$ concentrations are east of the Mississippi River from the southern Great Lakes to northern parts of the Gulf Coast states and in central and southern California.

Figure 3.1.1 (c) maps the differences between the two years, revealing considerable differences in the mean summertime concentrations. Emission reductions, which are described in Chapter 4, explain some of the change, but meteorological differences between the two years, which have varying influence across North America, are also important. Summertime concentrations of $PM_{2.5}$ were significantly lower over a broad area of eastern North America in 2012 compared to 2005; conversely, over western North America, $PM_{2.5}$ increased over some local areas, particularly in western Canada and the northwestern U.S. However, the relative contributions of meteorological factors and emission changes to the net effect are unknown. Therefore, the next several sections provide more robust information on trends in $PM_{2.5}$ and its chemical components.

3.1.2 Observations of $PM_{2.5}$ concentrations and trends across the United States

Consistent with Figure 3.1.1(c), annual average concentrations of $PM_{2.5}$ have decreased across much of the U.S. since the previous science assessment. Figure 3.1.2 (reproduced from the 2004 Assessment) shows average annual concentrations from 2000 to 2003 at all available monitoring sites. During this period, several areas in the U.S. had annual average concentrations above 12 $\mu g/m^3$, including several areas in the Midwest that had concentrations above the annual NAAQS for $PM_{2.5}$, which was 15 $\mu g/m^3$ at that time. In contrast, Figure 3.1.3 shows the design values[5] for the 2012 annual concentrations of $PM_{2.5}$ at available U.S. monitoring sites. Comparison of the two figures indicates where and by how much concentrations have decreased across the U.S. since 2000–2003. There are still some areas with annual concentrations above 12 $\mu g/m^3$, but most U.S. regions are now under this level.

Figure 3.1.2: Annual $PM_{2.5}$ (2000–2003)



(Source: 2004 Transboundary PM Science Assessment)

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[5] A design value is a statistic that describes the air quality status of a given location relative to the level of the NAAQS.

Figure 3.1.3: Design values for annual $PM_{2.5}$ concentrations (in µg/m^3) for 2012

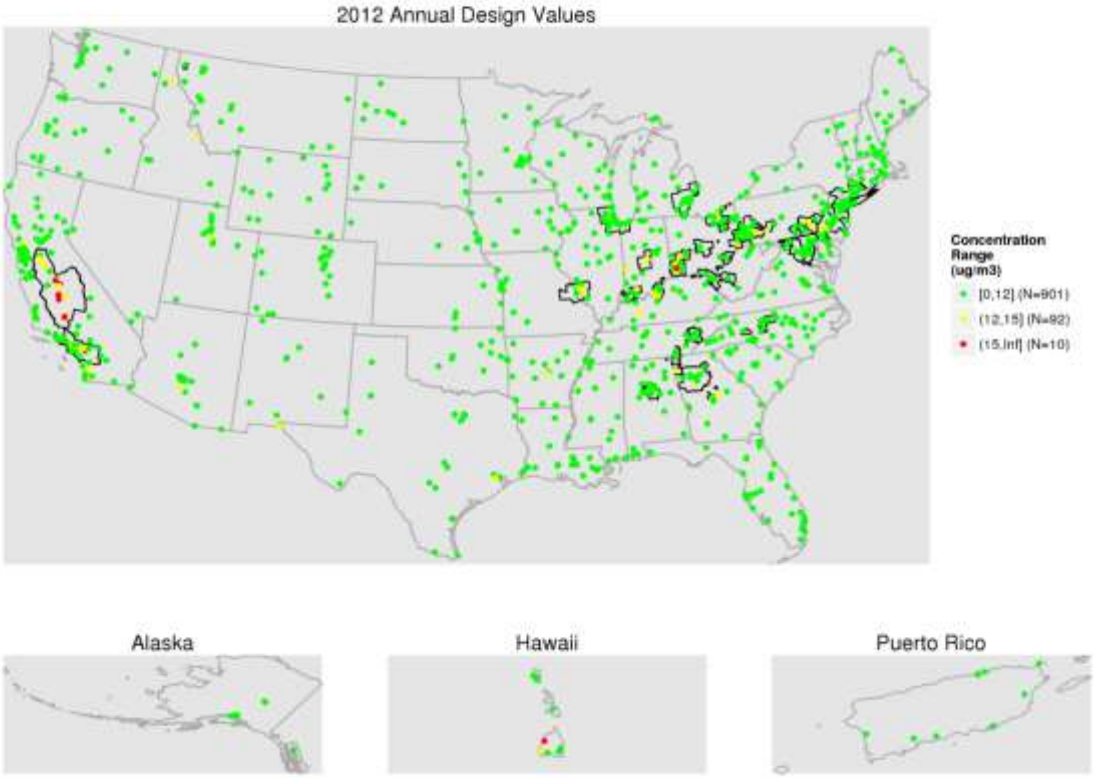
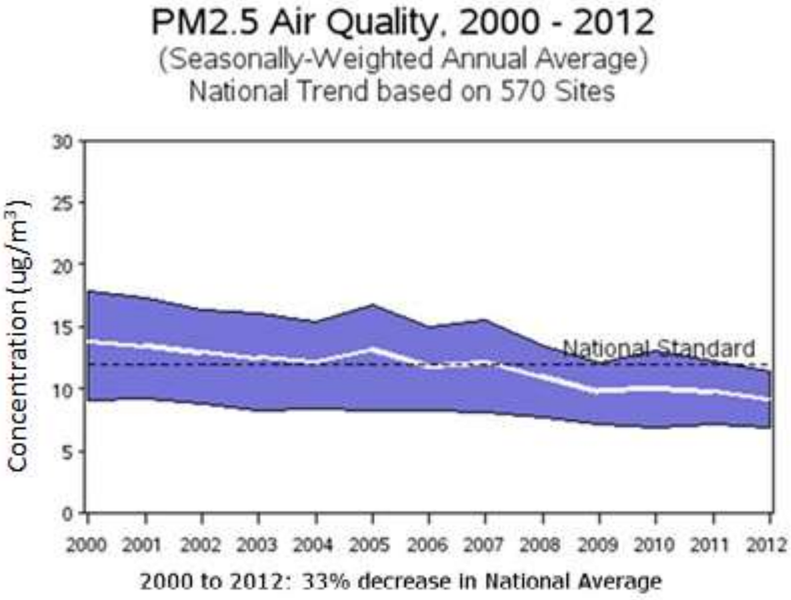


Figure 3.1.4 shows the trend in U.S. average annual concentrations of $PM_{2.5}$ from 2001 through 2012. During this period, there has been an overall decrease of 33% in the national average annual $PM_{2.5}$ concentration. In addition, most monitoring sites are currently below the annual NAAQS for $PM_{2.5}$. Comparison of Figures 3.1.5 (a) and (b) demonstrates the changes in regional annual average concentrations between 2000–2002 and 2009–2011. While annual average $PM_{2.5}$ concentrations have decreased in all regions, the same regional patterns exist: the highest concentrations occur in the industrial Midwest and the Northeast and the lowest concentrations in the Northwest and upper Midwest. Annual average concentrations in the industrial Midwest and the Northeast have decreased about 4 µg/m^3, while levels decreased in the Northwest about 2 µg/m^3 and in the upper Midwest about 1 µg/m^3. The mean annual average concentration is below the current 12 µg/m^3 NAAQS for $PM_{2.5}$ in all regions, although the highest concentrations, found in the industrial Midwest, do exceed the annual NAAQS.

Figure 3.1.6 shows design values for daily ambient $PM_{2.5}$ concentrations in 2012. Most of the U.S. sites are below the current standard of 35 µg/m^3. Figure 3.1.7 shows trends in ambient 24-h $PM_{2.5}$ concentrations, which have decreased 37% between 2001 and 2012.

Figure 3.1.4: National $PM_{2.5}$ air quality trend, 2000–2012 (annual average concentration)



(Source: 2012 EPA Air Trends Report)

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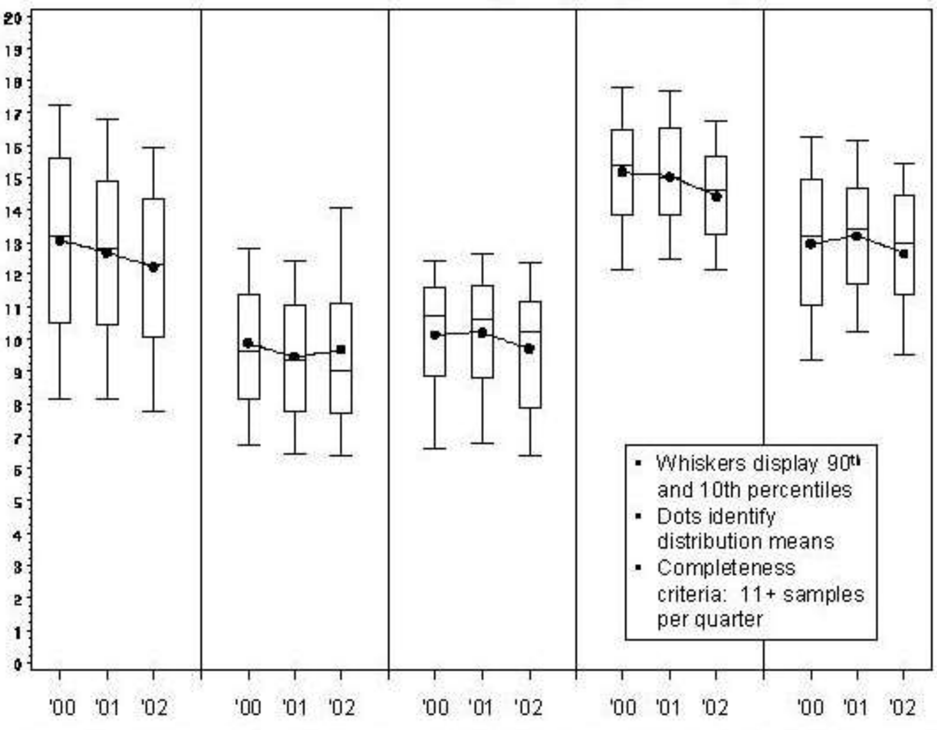
Figure 3.1.5(a): Annual $PM_{2.5}$ by region (2000–2002)



(Source: 2004 Transboundary PM Science Assessment)

Figure 3.1.5(b): Annual $PM_{2.5}$ by region (2009–2011)

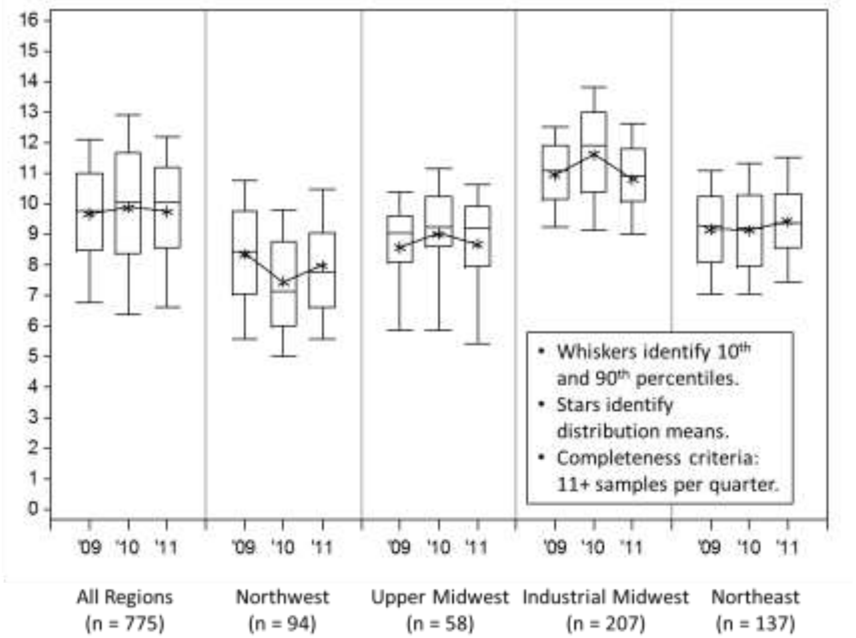


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Figure 3.1.6: Design values for daily $PM_{2.5}$ concentrations in 2012 (µg/m³)

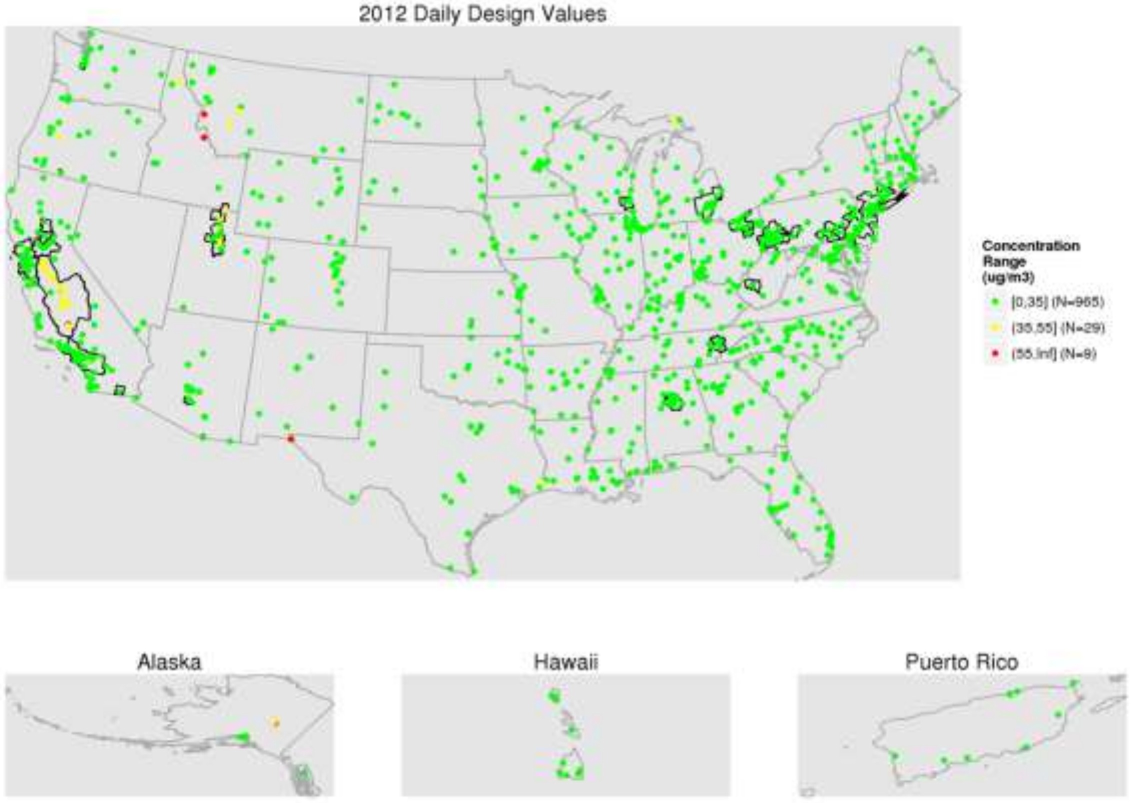
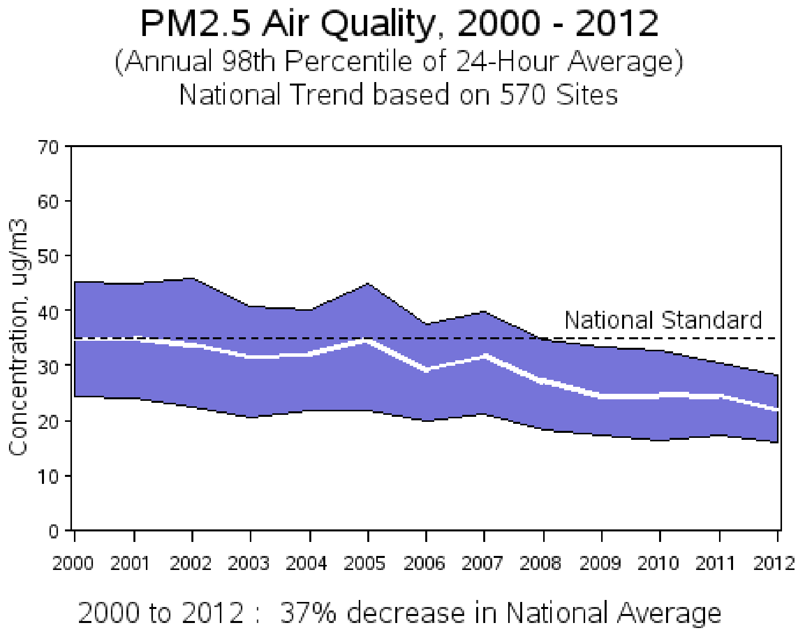


Figure 3.1.7: National $PM_{2.5}$ air quality trend, 2001–2012 (98th percentile of 24-h concentration in $\mu g/m^3$)



(Source: 2012 EPA Air Trends Report)

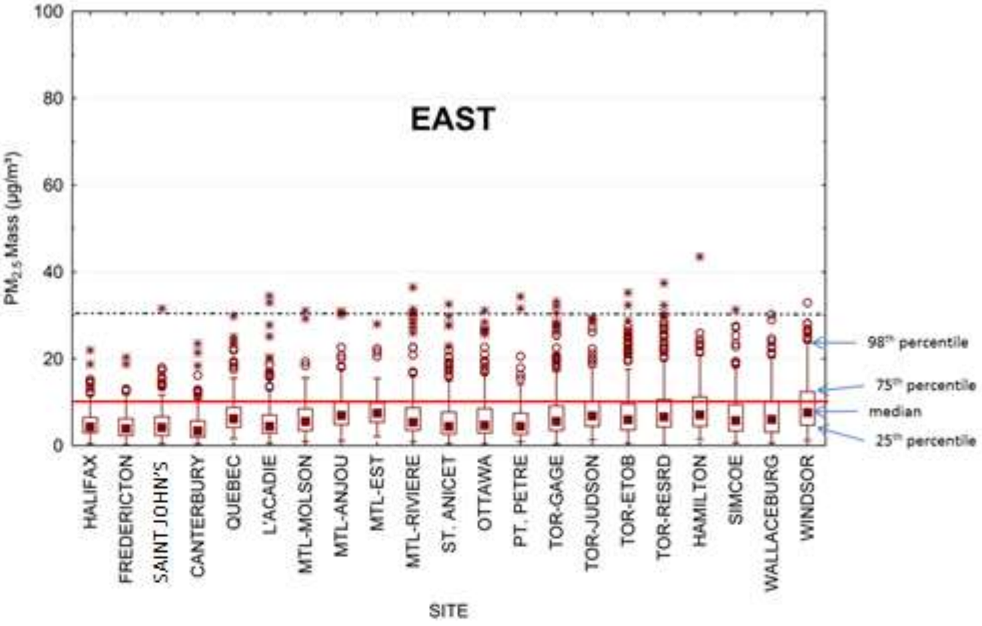
3.1.3 Observations of $PM_{2.5}$ concentrations in Canada

There are currently 21 locations in eastern Canada, 17 locations in western Canada and two locations in the North, with sufficient gravimetric $PM_{2.5}$ measurements to characterize the 24-h concentration distribution and annual averages. Figure 3.1.8(a) shows that at the eastern sites the median and 75th percentile concentrations were largest in Windsor, ON, which is adjacent to Detroit, MI. Levels tend to decrease from southwestern Ontario towards the Atlantic coast, with higher concentrations in the cities of Hamilton, ON, Toronto, ON, and Montréal, QC.

In the West (Figure 3.1.8(b)) the highest median concentration of $PM_{2.5}$ occurs within a mountainous region (Quesnel, B.C.) where the terrain (i.e. valleys) plays a role in trapping local emissions and leads to higher concentrations. Local emissions of importance include resource-based industries (e.g. forest products) as well as residential wood smoke. Wintertime temperature inversions in the mountain valleys more than double $PM_{2.5}$ concentrations in some locations. Substantial use of wood for heating contributes to the seasonal differences.

Among the larger cities of western Canada, Edmonton has the highest median $PM_{2.5}$ level, and concentrations in winter are double those in the summer. This seasonality is likely due to strong, shallow inversions that trap local emissions closer to the ground in winter, and to the increased partitioning of semi-volatile compounds (NO_3^-, organics) into the particle phase at colder temperatures.

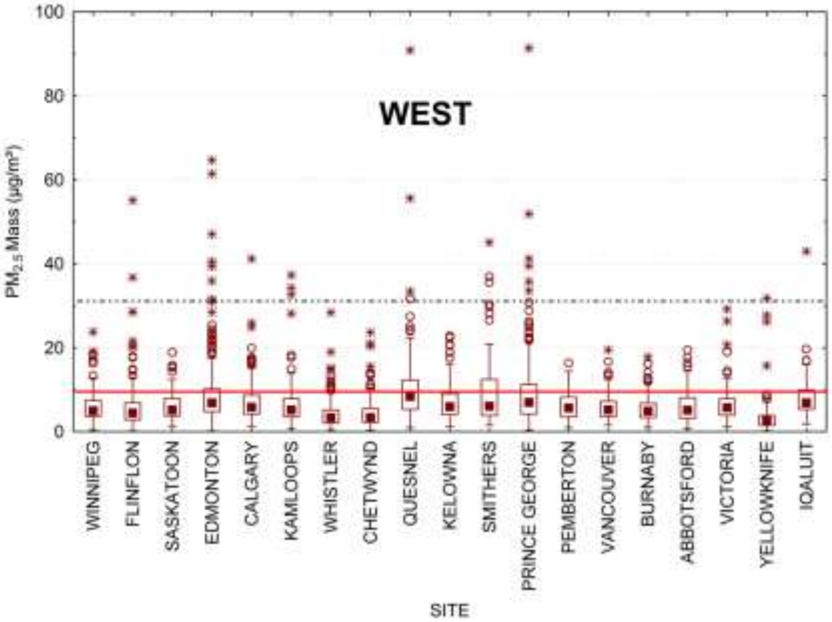
Figure 3.1.8(a): Comparison of 24-h $PM_{2.5}$ mass concentrations in eastern Canada from filter-based measurements obtained from 2008 to 2010



Note: The box plots indicate the median, 2nd, 25th, 75th and 98th percentiles, and outliers and extremes. The red line at 10 mg/m³ corresponds to the Annual Canadian Ambient Air Quality Standard to be in effect in 2015. At all sites with sufficient data to generate representative annual statistics (i.e. only sites with a minimum of 100 samples collected over the 2008–2010 period) the median concentration, which is close to the average, is seen to meet this future standard. The black dashed line indicates the current Canada-Wide Standard (30 mg/m³ for the 3-year average of the annual 98th percentile values).

* * * * *

Figure 3.1.8(b): Comparison of 24-h $PM_{2.5}$ mass concentrations in western Canada from filter-based measurements obtained from 2008 to 2010



Note: The sites on the right side of the plot are located in the north. The box plots indicate the median, 2nd, 25th, 75th and 98th percentiles, and outliers and extremes. The red line at 10 mg/m³ corresponds to the Annual Canadian Ambient Air Quality Standard to be in effect in 2015. At all sites with sufficient data to generate representative annual statistics (i.e. only sites with a minimum of 100 samples collected over the 2008–2010 period) the median concentration, which is close to the average, is seen to meet this future standard. The black dashed line indicates the current Canada-Wide Standard (30 mg/m³ for the 3-year average of the annual 98th percentile values)

3.2 Chemical Composition of $PM_{2.5}$ in Canada and the United States

3.2.1 Chemical composition in Canada

Since 1984, $PM_{2.5}$ particle mass has been measured at NAPS network sites using dichotomous filter-based samplers. Although this program has generated valuable data on $PM_{2.5}$ mass, metals, and some ions, a complete accounting of PM components could not be made because OC and EC were not measured, and NH_4NO_3 was volatilizing from the samples during analysis. In 2003, a new particle speciation program was initiated to accurately measure all the important components of $PM_{2.5}$. As of 2011, there were 38 dichotomous samplers operating at NAPS sites supplemented by an additional three EPA federal reference method samplers measuring only $PM_{2.5}$. The Teflon filters from the samplers are routinely analyzed for mass, elements, anions, and cations.

These samplers are operated once every 3 days, and samples are collected over 24 hours. Under the NAPS program, a total of 12 speciation sites (4 rural and 8 urban) are now in operation across Canada. Measured chemical components include inorganic and organic ions, total and water-soluble elements, OC and EC, levoglucosan and its isomers (since 2009), acid gases (nitric acid (HNO_3) and SO_2), and NH_3. All are quantified by well-defined and accredited analytical methods (Dabek-Zlotorzynska et al., 2011).

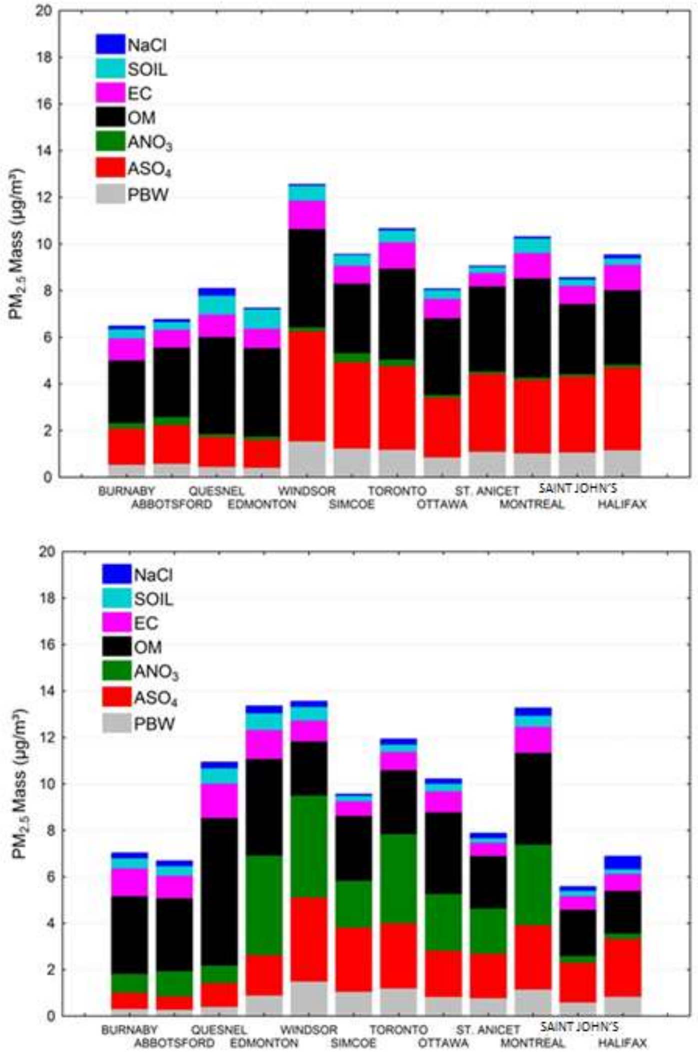
Mass reconstruction is a technique used to estimate the contributions of major components to PM mass using the measured individual chemical species, including salt, soil, EC, organic matter, particle-bound water, $(NH_4)_2SO_4$ and NH_4NO_3. The calculated total mass of the reconstructed components is then compared against the actual measured $PM_{2.5}$ mass values. Results of the $PM_{2.5}$

mass reconstruction at locations across Canada during the summer and winter months from 2007 to 2009 are shown in Figure 3.2.1.

Secondary inorganic species, specifically $(NH_4)_2SO_4$ and NH_4NO_3, together accounted for 32−43% of total $PM_{2.5}$ mass at the eastern speciation sites during the summer and for 36−59% in the winter. At the western sites, the contribution was smaller but still important at 18−30% in the summer and 22−45% in the winter.

Organic matter is the next most important contributor to $PM_{2.5}$ mass after the two secondary inorganic species except for the Edmonton and the three British Columbia sites, where it is the largest contributor. Organic matter contributions to $PM_{2.5}$ mass for urban and rural sites range from 31% to 52% in summer and from 23% to 58% in winter. Interior locations in British Columbia, such as Quesnel and Golden, have the largest contributions of organic matter to $PM_{2.5}$ mass, particularly in the winter (Dabek-Zlotorzynska et al., 2011). Wood combustion, either from industry and residential heating or from wildfires, is the key contributor in these areas.

Figure 3.2.1: 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)



Note: NaCl = salt, EC = elemental carbon, OM = organic matter, ANO3 = ammonium nitrate, ASO4 = ammonium sulphate, PBW = particle-bound water.

For the 10 highest days in summer, $PM_{2.5}$ mass at the eastern NAPS speciation sites is 70–80% $(NH_4)_2SO_4$ and organic matter (GoC, 2012a). $(NH_4)_2SO_4$, which is from anthropogenic sources, is responsible for approximately 50% of the total $PM_{2.5}$ mass. Typically these high episodes involve southerly flow, implicating sources from the U.S., although depending upon the location, emissions of SO_2 in Canada also contribute. At western sites other than Golden, NH_4NO_3 is also an important contributor during summer episodes.

For winter days when the highest $PM_{2.5}$ concentrations were observed, NH_4NO_3 and organic matter 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, particularly Halifax, NS. Given the reduction in both biological activity and wildfire frequency during winter, the increased importance of organic matter during winter PM episodes is inferred to result from the build-up of anthropogenic emissions of primary organic matter and its precursors (gas-phase volatile organic compounds (VOCs)).

3.2.2 Chemical composition in the United States

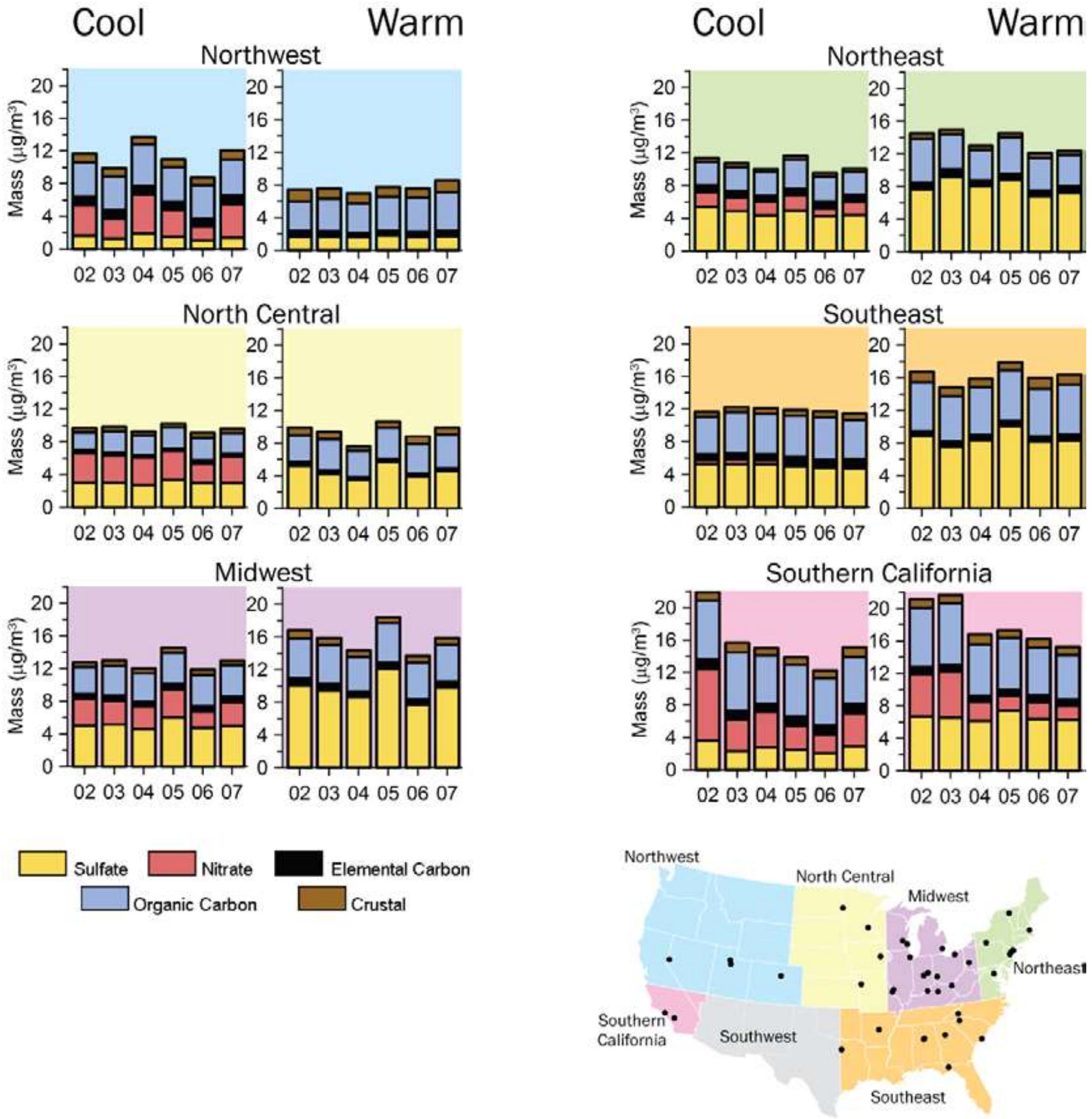
The chemical composition of $PM_{2.5}$ is measured at two primary networks in the U.S.: the Chemical Speciation Network (CSN) and the IMPROVE network. The CSN contains about 200 sites, mostly in urban locations, while the IMPROVE network contains 160 sites, mostly in rural and remote locations. Most of the sites in the CSN and the IMPROVE networks collect integrated 24-h filter-based measurements every third day, although some collect every sixth day.

Another network that provides information on $PM_{2.5}$ species is the Clean Air Status and Trends Network (CASTNET). CASTNET is a long-term, rural monitoring network that collects weekly measurements of nitrogen and sulphur species, as well as O_3, at 90 sites in the U.S. and one site in Ontario, Canada.

The chemical makeup of fine particles varies across the U.S., as illustrated in Figure 3.2.2. For example, higher regional emissions of SO_2 in the east result in higher levels of sulphates than are found in the western U.S. Fine particles in southern California generally contain more nitrates than other areas of the country. EC and organic compounds are a substantial component of fine particles everywhere. Fine particles can also have a seasonal pattern.

As shown in Figure 3.2.2, $PM_{2.5}$ values in the eastern half of the U.S. are typically higher in warmer weather, when meteorological conditions favour the formation and build-up of sulphates due to higher SO_2 emissions from power plants in that region. Fine particle concentrations tend to be higher in the cooler calendar months in urban areas in the west, in part because fine particle nitrates and carbonaceous particles are more readily formed in cooler weather, and wood stove and fireplace use increases direct emissions of carbon.

Figure 3.2.2: Regional and seasonal trends in annual $PM_{2.5}$ composition from 2002 to 2007



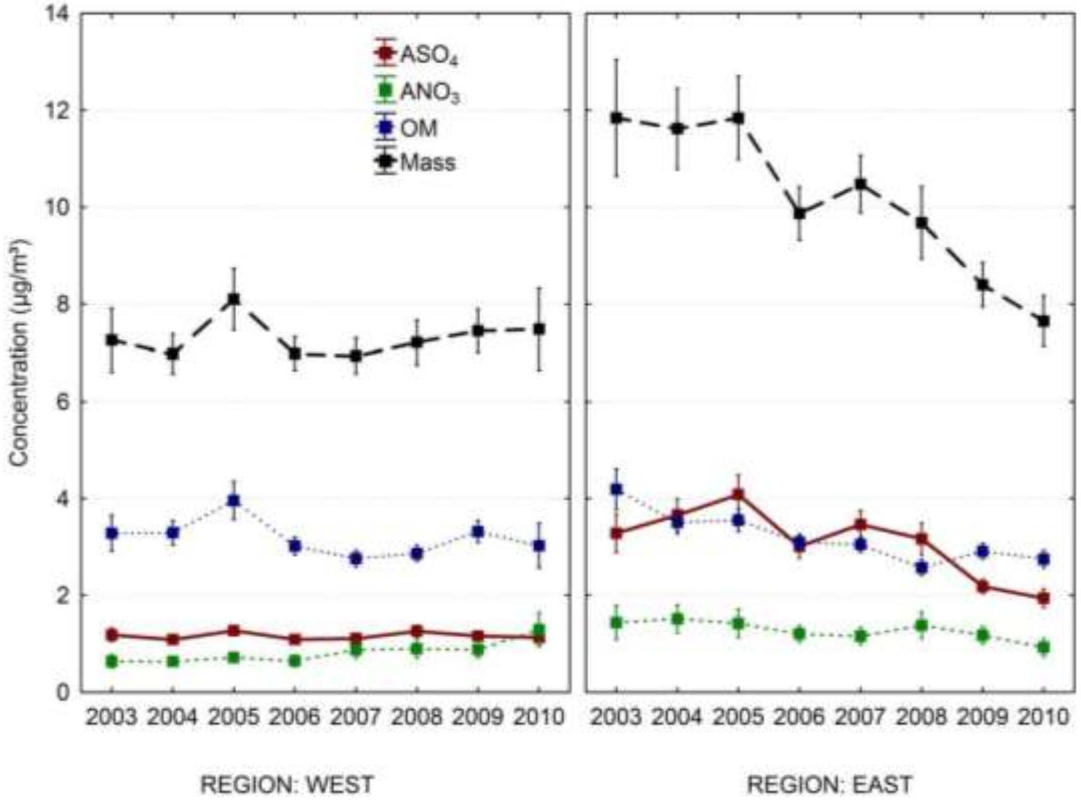
Note: Data from the 42 monitoring locations shown on the map were stratified by region and season, including cool months (October–April) and warm months (May–September).
(Source: Regulatory Impact Analysis for the Final Revisions to the NAAQS for PM (2012))

3.2.3 Analysis of trends in $PM_{2.5}$ and components in eastern Canada and the United States

Between 2005 and 2011 fine particle SO_4^{2-} declined at the eastern Canadian NAPS sites. East-to-west differences in SO_4^{2-} levels are now smaller than 10 years ago. Annual average concentrations of NH_4NO_3 have not changed markedly across the Canadian NAPS sites over this period despite a significant decrease in the ambient NO_x concentrations recorded at most locations across the country (Brook et al., 2013).

Figure 3.2.3 shows that, overall, annual average $PM_{2.5}$ mass has decreased significantly among the eastern NAPS speciation sites, with reductions in SO_4^{2-} and organic matter mass being responsible for the majority of the decline. However, among the western Canadian NAPS sites, $PM_{2.5}$ and its main components have remained stable.

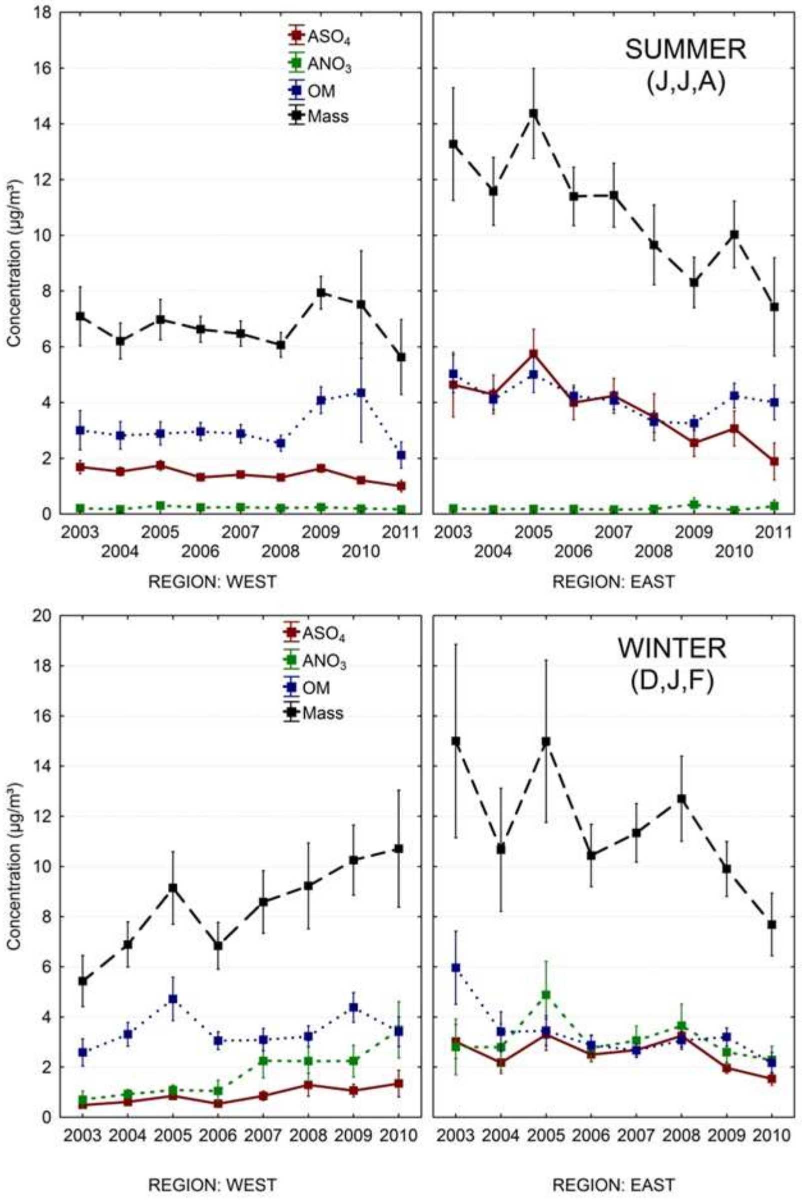
Figure 3.2.3: Trends in annual $PM_{2.5}$ mass, ammonium sulphate, ammonium nitrate, and organic matter from Canadian speciation network sites (2003–2011)



Note: Bars represent the 90th percentile confidence interval around the mean concentration. ASO_4 = ammonium sulphate, ANO_3 = ammonium nitrate, and OM = organic matter.

Figure 3.2.4 shows the eastern and western annual average trends (2003–2010) broken into the summer and winter months. For the winter months in the east, when NH_4NO_3 is a larger contributor to the mass, there was a small drop in this component in the later years. Most importantly, this figure shows that for western sites, wintertime $PM_{2.5}$ has been increasing from 2003 to 2010. This change appears to be related to rises in $(NH_4)_2SO_4$ and NH_4NO_3.

Figure 3.2.4: Trends in summer (June, July, August) and winter (December, January, February) for $PM_{2.5}$ mass, ammonium sulphate, ammonium nitrate, and organic matter

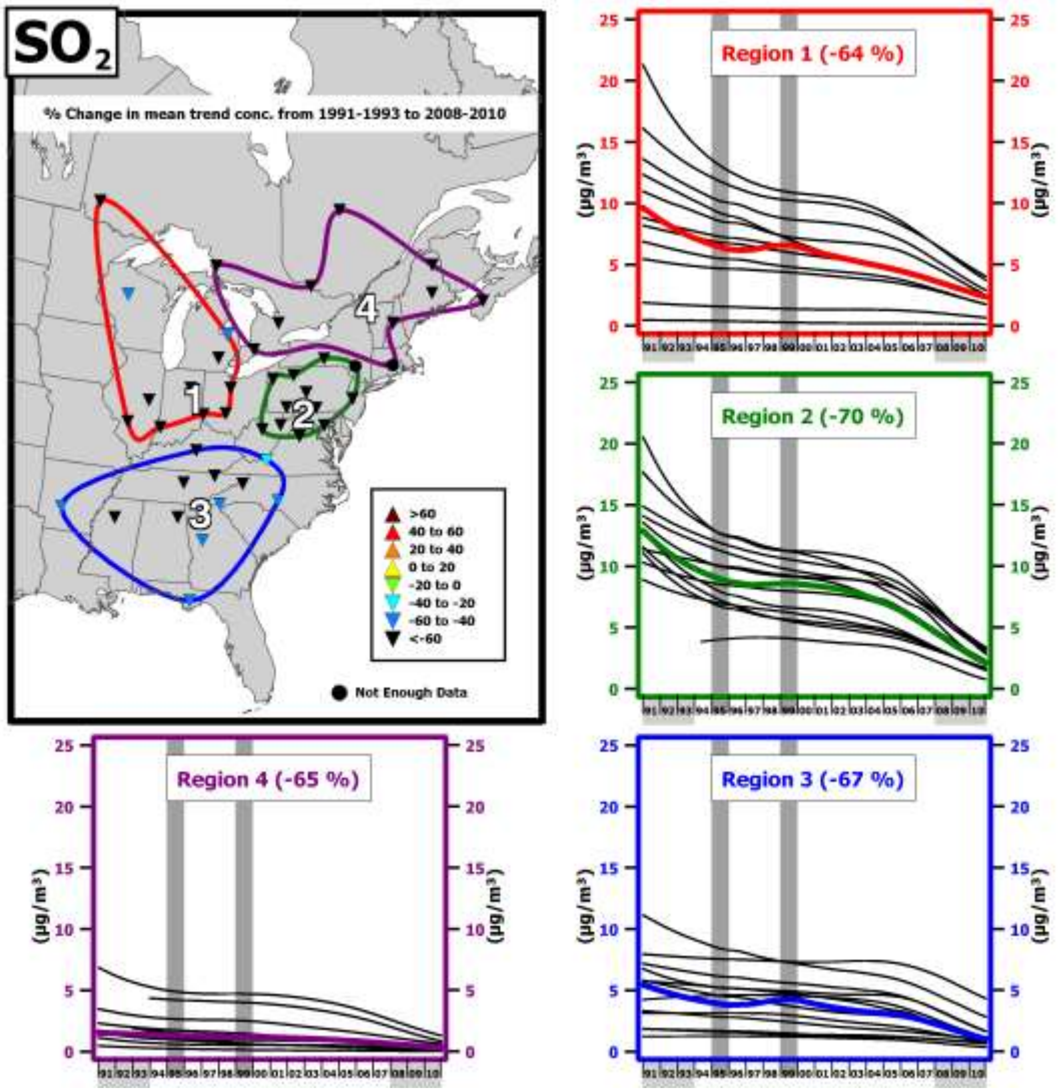


Note: Bars represent the 90th percentile confidence interval around the mean concentration. ASO_4 = ammonium sulphate, ANO_3 = ammonium nitrate, and OM = organic matter.

The $PM_{2.5}$ mass reduction in eastern Canada between 2003 and 2010 is generally consistent with the reductions observed in the upper Midwest and Northeast U.S., as shown by comparing Figures 3.1.5(a) and 3.1.5(b). The observed trends of $PM_{2.5}$ and its chemical composition are consistent with the emission trends discussed in Chapter 4.

Regionally representative long-term trends of ambient SO_2, particulate SO_4^{2-}, particulate NH_4^+ and particulate NO_3^- were assessed from data collected at rural and remote sites in the Canadian Air and Precipitation Monitoring Network (CAPMoN) and in CASTNET. These sites are located in areas with little influence from local emission sources, and their trends represent regional-scale changes to PM and its precursors. Figures 3.2.5 to 3.2.8 show the time trends of SO_2, particulate SO_4^{2-}, particulate NH_4^+, and particulate NO_3^- in four regions of eastern North America over the period 1991–2010. The boundaries of the four regions were determined from principal component analysis, which separated the sites into groups with common spatial and temporal behaviour (based on particulate SO_4^{2-} data and applied to the other species) using the technique described in Chan (2009). Smoothed trend curves in the figures were generated using a technique described in Cleveland et al. (1988). Coloured triangles on the maps show the changes in 3-year average concentrations between the time intervals of 1991−1993 and 2008−2010 based on the time-series curves.

Figure 3.2.5: Map of CAPMoN and CASTNET sites showing the percentage changes in the 3-year average SO_2 concentrations from 1991–1993 to 2008–2010



Note: The percentage changes shown in the triangles are based on the trend curves in the regional boxes. The black trend lines in the boxes show the site-by-site trend lines in the U.S. Midwest (Region 1), the U.S. Eastern Seaboard (Region 2), the U.S. Southeast (Region 3) and eastern Canada/the U.S. Northeast (Region 4), while the coloured trend lines show the group-mean trends.

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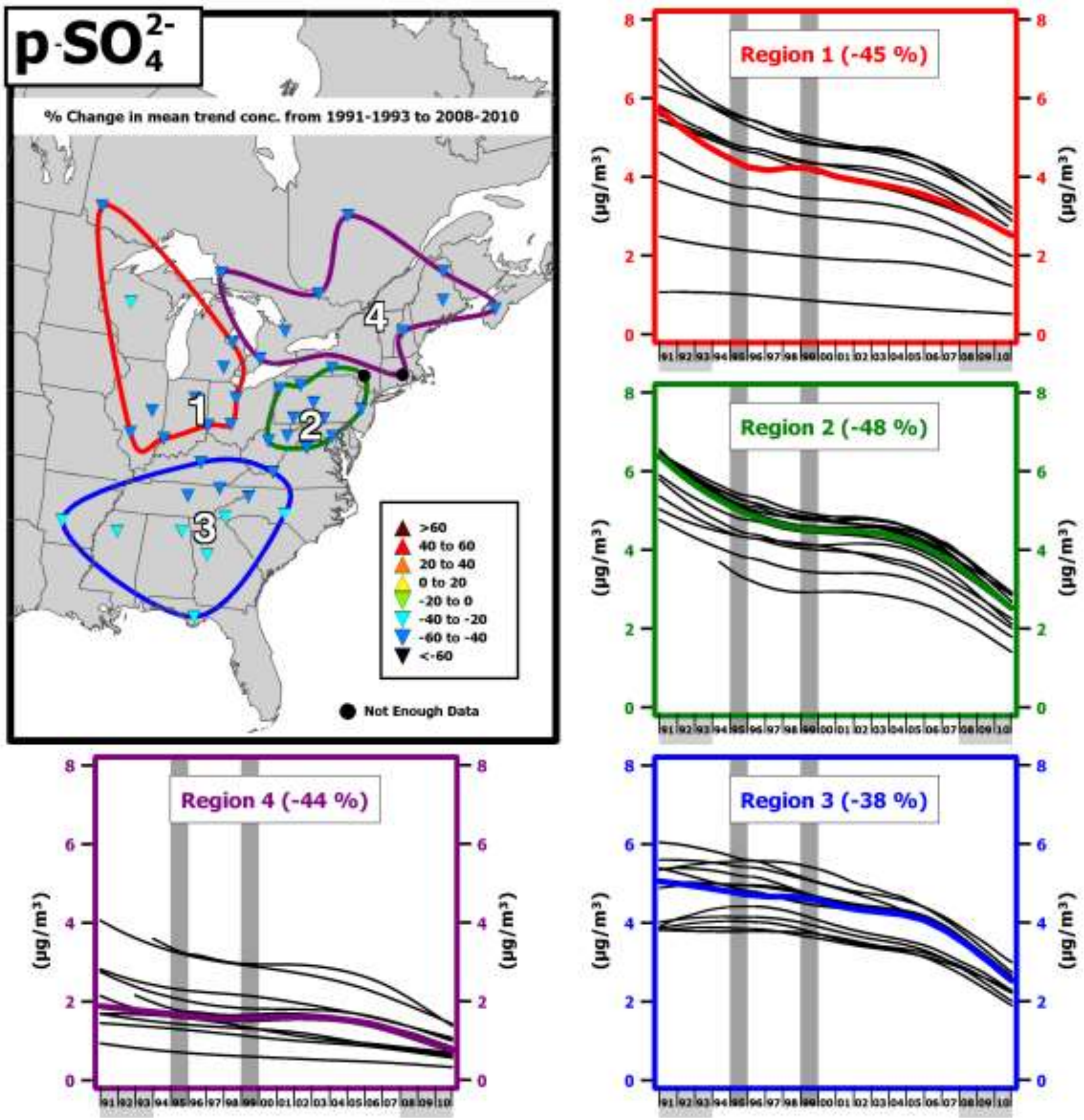
Figure 3.2.6: Map of CAPMoN and CASTNET sites showing the percentage changes in the 3-year average particulate SO_4^{-2} concentrations from 1991–1993 to 2008–2010



Note: The percentage changes shown in the triangles are based on the trend curves in the regional boxes. The black trend lines in the boxes show the site-by-site trend lines in the U.S. Midwest (Region 1), the U.S. Eastern Seaboard (Region 2), the U.S. Southeast (Region 3) and eastern Canada/the U.S. Northeast (Region 4), while the coloured trend lines show the group-mean trends.

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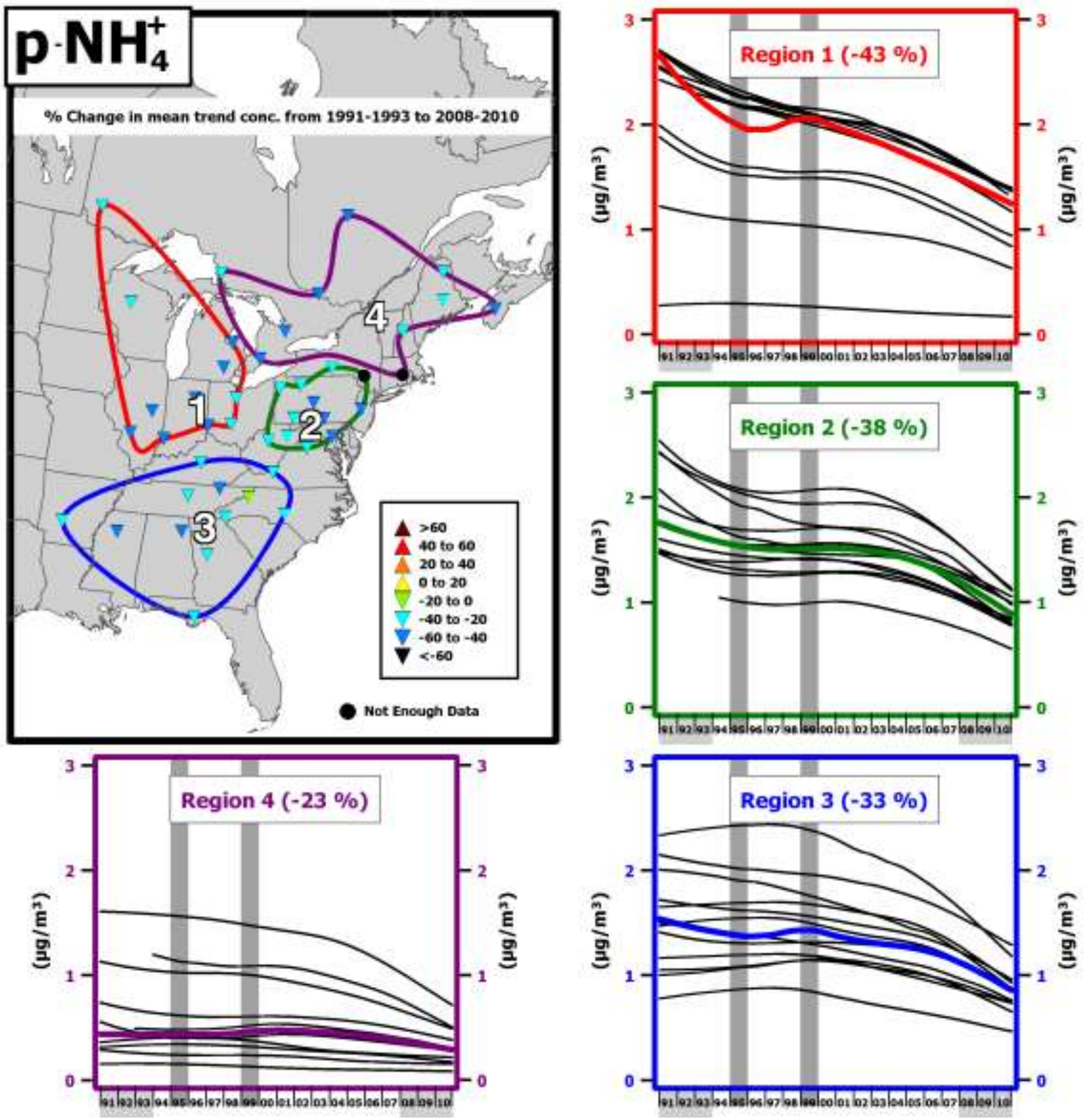
Figure 3.2.7: Map of CAPMoN and CASTNET sites showing the percentage changes in the 3-year average particulate NH_4^+ concentrations from 1991–1993 to 2008–2010



Note: the percentage changes shown in the triangles are based on the trend curves shown in the regional boxes. The black trend lines in the boxes show the site-by-site trend lines in the U.S. Midwest (Region 1), the U.S. Eastern Seaboard (Region 2), the U.S. Southeast (Region 3) and eastern Canada/ the U.S. Northeast (Region 4), while the coloured trend lines show the group-mean trends.

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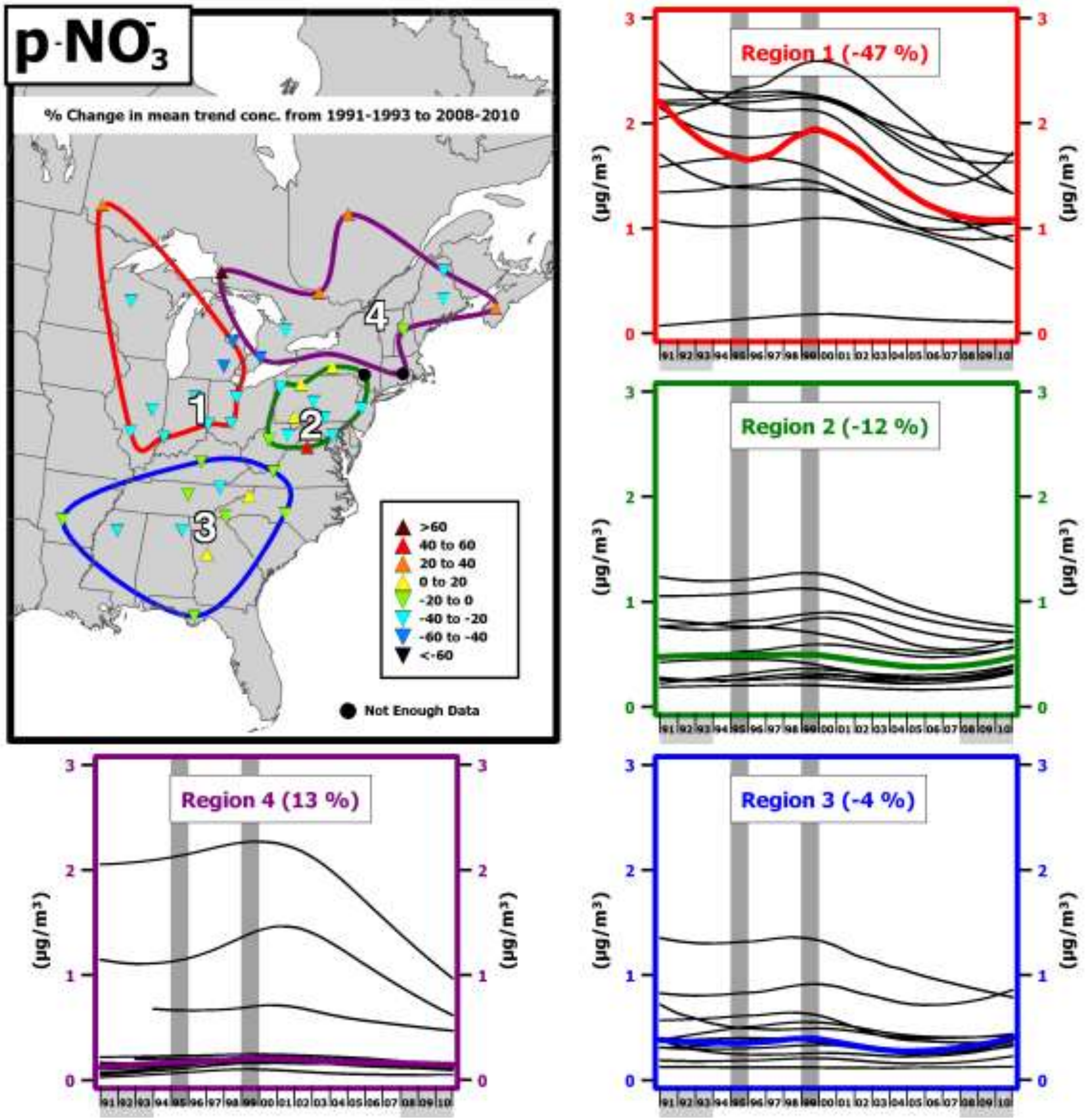
Figure 3.2.8: Map of CAPMoN and CASTNET sites with triangles showing the percentage change in the 3-year average particulate NO_3^- concentrations between 1991 - 1993 and 2008 - 2010



Note*:* the percentage changes shown in the triangles are based on the trend curves shown in the regional boxes. The black trend lines in the boxes show the site-by-site trend lines in the U.S. Midwest (Region 1), the U.S. Eastern Seaboard (Region 2), the U.S. Southeast (Region 3) and eastern Canada/the U.S. Northeast (Region 4), while the coloured trend lines show the group-mean trends.

SO_2 concentrations decreased markedly from 1991 to 2010 at all sites in the four regions (Figure 3.2.5), although there was a period of relatively small change in the late 1990s and early 2000s. Trends for two sites (shown by circles in Figure 3.2.5) could not be determined due to insufficient data. The group-mean 3-year average concentrations of SO_2 in the four regions (coloured trend lines in the figures) decreased 64– 70% between 1991−1993 and 2008−2010, suggesting that SO_2 emission reductions from the early 1990s though 2010 in eastern Canada and the eastern U.S. were highly effective at decreasing ambient SO_2.

Particle SO_4^{2-} concentrations (Figure 3.2.6) also decreased markedly from 1991 to 2010 at sites in the four regions, with a group-mean reduction of 38–48% in the 3-year average concentrations between 1991−1993 and 2008−2010. These decreases are consistent with major reductions in SO_2 precursor emissions during the time period and with the general PM trends described in the other trend figures shown in this chapter.

Consistent with the SO_2 and particulate SO_4^{2-} trends, particle NH_4^+ concentrations also decreased from 1991 to 2010 at all sites (except for two sites with insufficient data) (Figure 3.2.7). The regional 3-year average concentrations decreased 23−43%, with Region 1 in the midwestern U.S. showing the greatest decline. For the Great Lakes states within Regions 1 and 2, there was a steady reduction at all sites from 1991 to 1995−1996, followed by a levelling off until about 2002−2003 and then a final decline to the year 2010. Regions 3 and 4 were different in that Region 3 sites in the southeastern U.S. began to trend downward earlier (i.e. in 1999−2000), while Region 4 sites in Ontario, Quebec and the eastern seaboard of both countries exhibited variable trends.

Trends in particle NO_3^- (Figure 3.2.8) differed from those of the other PM precursors and components. Many sites in the four regions showed decreasing trends of 0% to 60% between 1991−1993 and 2008−2010, while many others showed either no change or increasing trends of 0% to 60%. The sites in the midwestern U.S. (Region 1) showed consistent declines, with a 47% drop in regional group-means between the two periods. It is worth noting that particle NO_3^- can undergo volatilization in the atmosphere and after being collected on filters, which suggests that the filter-based concentrations shown in Figure 3.1.8 may be lower than actual. Nevertheless, particle NO_3^- trends are shown here because particle NO_3^- constitutes an important part of total $PM_{2.5}$ mass in North America. The trend lines in Figure 3.1.8 are considered reasonable, given the assumption that particle NO_3^- volatilization is relatively constant over the long term.

Interestingly, many sites in the four regions exhibited an upward bump in the trends in the late 1990s and early 2000s, although the timing of the maximum value varied from site to site. This particular period was characterized by a major drop in SO_2 emissions with little change in NO_x emissions. The drivers of this upward particulate NO_3^- trend are related to the complex interplay among sulphates, nitrates, NH_3 and temperature and changing SO_2, NO_x and NH_3 emissions (both intra-annual and inter-annual). Decreasing SO_2 emissions and levels and subsequent decreases in its secondary product, sulphuric acid (H_2SO_4), lead to more of the ambient NH_3 being available to react with secondary products of the NO_x emissions, mostly HNO_3, which, when the temperature and relative humidity are favourable, forms particle NO_3^-. Thus, without concurrent NO_x emission reductions, there were increases in particle NO_3^-. The surprising observation is that at a number of the sites, particle NO_3^- increased over the period 2008–2010 when NO_x emissions were likely to be decreasing. The reasons for this behaviour have not been determined and thus require further study.

3.2.4 Relationships between chemical composition and sources

Source apportionment studies use measurements of the chemical composition of $PM_{2.5}$ to provide insights into the sources contributing to its concentrations. These studies are typically able to be more specific for linking the primary fraction of the $PM_{2.5}$ to likely sources and are much less able to identify the sources of the secondary inorganic or organic $PM_{2.5}$. The results of source apportionment studies relevant to transboundary $PM_{2.5}$ were summarized in the 2004 Assessment.

Jeong and colleagues (2008a) applied receptor models to the data from five of the locations in Figure 3.2.1. For each location, the $PM_{2.5}$ was apportioned into eight or nine "sources" related to both local and/or more distant emissions. (However, given the capabilities inherent in receptor modelling approaches, the goal of this work was not to quantify sources according to their country of origin.) Not surprisingly, secondary SO_4^{-2} and NO_3^- were the most significant contributors to $PM_{2.5}$ mass, accounting for ≈59% in Windsor, ≈56% in Toronto, ≈46% in Montréal, ≈49% in Halifax, and ≈40% in Edmonton. With the exception of Edmonton, these secondary factors were assumed, due to the relatively large SO_2

and NO_x emissions in these areas, to be related to regional sources, many being located in the U.S. in Ohio, Pennsylvania, and New York. The combined contribution of motor vehicle-related sources (traffic and road dust factors) ranged from 13% to 20% across the five locations studied, with the majority of the emissions expected to be from local activities (i.e. within each city). Given its northern location and the limited regional emission sources, the secondary NO_3^- and SO_4^{-2} in Edmonton were also assumed to be mostly a result of local sources.

Source apportionment for the five Canadian cities was recently updated (Jeong et al., 2011). Some additional but relatively minor sources of primary $PM_{2.5}$ were uncovered, given the availability of more data and possible changes in the sources. However, there was still a large contribution from inorganic secondary $PM_{2.5}$ ($(NH_4)_2SO_4$ and NH_4NO_3) for which specific sources could not be identified. Given that emissions of SO_2 and NO_x are relatively well known, including the locations of the major emissions, it is possible to infer which sources are most important (e.g. combustion of sulphur-rich fuels such as coal). Estimates of contributions from Canada versus the U.S. are also possible by comparing observations among Canadian sites, including those situated close to the border (Brook et al., 2002). However, Jeong and colleagues (2011) did not clearly resolve the fraction of $PM_{2.5}$ related to secondary organic aerosols (SOAs). This was likely due to a larger range of sources contributing to the precursor VOCs and the fact that there is a relatively limited understanding of SOA formation mechanisms. As a result of this knowledge gap, SOA formation has been the subject of considerable research since the 2004 Assessment (Goldstein and Galbally, 2007; de Gouw and Jimenez, 2009); some of this research relevant to the Canada – U.S. border region is highlighted below.

On average, SOAs are estimated to be a significant fraction, on the order of 60%, of the organic matter (de Gouw et al., 2008). However, there remains considerable uncertainty about this estimate; while knowledge continues to evolve it is not possible to determine if the recently published value of 60% is likely to be an over- or underestimate. The SOA contribution depends upon proximity to populated areas and time of year, and it will increase as air masses age in the atmosphere (Morgan et al., 2010). Given these uncertainties and assuming that the Canada – U.S. border region is influenced by a considerable number of large cities and industrial regions, an order of magnitude assumption is that half the organic matter is SOAs. Thus at least half the $PM_{2.5}$ over southern border regions of Canada, particularly in the eastern part of the country, originates from gaseous emissions and subsequent atmospheric processes (i.e. is secondary PM).

In contrast to $(NH_4)_2SO_4$ and NH_4NO_3, which arise mostly from anthropogenic SO_2 and NO_x emissions, SOAs are a result of both anthropogenic and biogenic emissions of VOCs. The relative importance of these two sources of VOCs to SOAs depends upon meteorological conditions and proximity to urban areas or urban plumes, as has been demonstrated for southern Ontario (Liggio et al., 2012; Slowik et al., 2010, 2011; Sjostedt et al., 2011). Overall, anthropogenic VOCs are estimated to contribute 55% of the total SOA fraction (de Gouw et al., 2008) over the U.S. (particularly in the northeast, and likely in populated areas of eastern Canada). Also, considering biomass burning emissions of organic matter (de Gouw et al., 2008) and assuming that they are predominantly natural leads to an estimate that half the observed organic matter is of anthropogenic origin. As with SOAs, this value varies temporally, from daily to seasonally, and also can be expected to vary geographically (i.e. western versus eastern Canada), though knowledge of such variations remains limited. However, estimates of the fraction of organic matter that is SOAs and the fraction of SOAs that is biogenic are uncertain and many questions remain, such as the role of cloud processing and surface uptake of gases on to aerosols (e.g. Li et al., 2011).

The uncertainty and the evolving knowledge regarding organic matter complicate efforts to determine how much of ambient $PM_{2.5}$ is due to controllable emissions. While modelling of the atmospheric concentration of organic matter is improving, our ability to reliably apportion organic matter to its multitude of sources remains limited. Lack of knowledge limits the representation of secondary formation processes in the models, and there is substantial uncertainty in estimates of natural emissions (which include biogenic VOCs, wildfires and primary particles made of biological matter). Primary anthropogenic emissions of organic matter are also relatively poorly characterized (Stroud et al, 2012). As a result, current models cannot accurately attribute the organic matter portion of the $PM_{2.5}$ shown in Figure 3.2.1 to sources, or clarify how much originates in Canada versus the U.S. Thus, given that non-urban measurements at Canadian sites closer to the border show that $PM_{2.5}$ entering Canada from the U.S. contains a considerable amount of organic matter, the best estimate of the contribution of U.S. anthropogenic emissions (i.e. controllable), is approximately 50%, based on the observation-based approach used in de Gouw et al. (2008).

Several aspects of the 2007 Border Air Quality and Meteorological Study (BAQS-Met) field campaign (Brook et al., 2013) have recently assessed the sources and processes affecting $PM_{2.5}$ in southwestern Ontario through measurements and modelling with AURAMS. A unique aspect of this work was the focus on a small domain, fine scale (2.5-km) modelling and the impacts on $PM_{2.5}$ from sources closer to the border region in both Canada and the U.S.

McGuire et al. (2011) deployed a single particle mass spectrometer at a rural location close to the border to obtain high resolution measurements for receptor modelling. This effort revealed the complexity of the particles in this region, finding 33 unique particle types, which, due to similarities in temporal behaviour, could be grouped into seven particle families. There were distinct differences among all 33 in their composition, potential origin, and characteristic sizes, with variations in the extent of external versus internal mixing and the degree of atmospheric processing. However, despite the level of detail possible with such advanced measurement techniques, including use of high-resolution back-trajectories, linking the particles or particle families back to specific sources in either country could not be done with a high degree of confidence. Using BAQS-Met measurements Ellis et al (2011) and Markovic et al. (2011) focused on modelling NH_4NO_3, and Stroud et al. (2012) concentrated on organic matter to identify deficiencies in AURAMS and/or the emissions information. Their results further support the assertion mentioned above that there remains considerable uncertainty in model results, especially for NH_4NO_3 (in summer) and for organic matter. Consequently, despite ongoing improvement in model applications, attempts to quantify source–receptor relationships must be interpreted with care, particularly if the goal is to identify specific sources in Canada or the U.S. in closer proximity to the border for greater attention with respect to emission reductions.

CHAPTER 4: EMISSIONS AND EMISSION TRENDS OF POLLUTANTS CONTRIBUTING TO $PM_{2.5}$ CONCENTRATIONS IN THE U.S. AND CANADA

Chapter Summary

- National emission inventories indicate that total anthropogenic emissions of primary $PM_{2.5}$ and NH_3 have remained fairly stable in Canada and the U.S. from 2002 to 2010, while emissions of SO_2, NO_x, and VOCs have all declined over this period.
- In Canada, reductions of SO_2 emissions by 40% since 2002 are attributable to technological and process changes in base metal smelting industries, regulatory changes in the upstream oil and gas sector in Alberta, as well as the economic slowdown and facility closures through 2010. Lower NO_x emissions are attributable to more stringent emission regulations for the transportation and electric power generation sectors. Improved standards for on-road and off-road vehicle engine emissions have contributed to reductions in VOC emissions.
- In the U.S., emissions of $PM_{2.5}$ precursors (SO_2, NO_x, VOCs) have declined steadily since 2000, while primary emissions of $PM_{2.5}$ and NH_3 have remained relatively constant. The most significant decreases have come from reductions in SO_2 and NO_x from the fuel combustion sector and the non-road mobile sector, as well as declines in VOCs from highway vehicles and other miscellaneous sources. Conversely, emissions of all pollutants from wildfires have increased in the 2008 National Emissions Inventory (NEI).
- Anthropogenic emissions of SO_2, NO_x and VOCs are projected to decrease through about 2020 in both countries, while emissions of $PM_{2.5}$ and NH_3 are expected to change much less during this period. Beyond 2020, decreases in all emissions are expected to be smaller.

4.1 Summary of Major Sources of $PM_{2.5}$

Table 4.1 below summarizes anthropogenic and natural sources for the major primary and secondary aerosol constituents of fine particles. Anthropogenic sources can be further divided into stationary and mobile sources. Stationary sources include fuel combustion for electrical utilities, residential space heating and cooking; industrial processes; construction and demolition; metal, mineral, and petrochemical processing; wood products processing; mills and elevators used in agriculture; erosion from tilled lands; waste disposal and recycling; and biomass combustion. Biomass combustion encompasses many emission activities, including burning of wood for fuel, as well as burning of vegetation to clear land for agriculture and construction, to dispose of agricultural and domestic waste, to control the growth of animal or plant pests, or to manage forest resources. Wildlands also burn due to lightning strikes and arson. Mobile or transportation-related sources include emissions of primary PM and secondary PM precursors from on-road vehicles and non-road sources, as well as fugitive dust from paved and unpaved roads. Also shown in Table 4.1 are sources for several precursor gases, the oxidation of which can form secondary PM.

Table 4.1: Summary of major sources of $PM_{2.5}$

Primary (PM <2.5 µm)			Secondary PM Precursors (PM <2.5 µm)	
Aerosol Species	Natural	Anthropogenic	Natural	Anthropogenic
Sulfate(SO_4^{2-})	Sea spray	Fossil fuel combustion	Oxidation of reduced sulfur gases emitted by the oceans and wetlands and SO_2 and H_2S emitted by volcanism and forest fires	Oxidation of SO_2 emitted from fossil fuel combustion
Nitrate(NO_3^-)	-	Mobile source exhaust	Oxidation of NO_X produced by soils, forest fires, and lightning	Oxidation of NO_X emitted from fossil fuel combustion and in motor vehicle exhaust
Minerals	Erosion and re-entertainment	Fugitive dust from paved and unpaved roads, agriculture, forestry, construction, and demolition	-	-
Ammonium (NH_4^+)	-	Mobile source exhaust	Emissions of NH_3 from wild animals, and undisturbed soil	Emissions of NH_3 from motor vehicles, animal husbandry, sewage, and fertilized land
Organic Carbon (OC)	Wildfires	Prescribed burning, wood burning, mobile source exhaust, cooking, tire wear and industrial processes	Oxidation of hydrocarbons emitted by vegetation (isoprene, terpenes, waxes) and wild fires	Oxidation of hydrocarbons emitted by motor vehicles, prescribed burning, wood burning, solvent use and industrial processes
Elemental Carbon (EC)	Wildfires	Mobile source exhaust (mainly diesel), wood biomass burning, and cooking	-	-
Metals	Volcanic activity	Fossil fuel combustion, smelting and other metallurgical processes, and brake wear	-	-
Bioaerosols	Viruses and bacteria	-	-	-

(Source: EPA, PM ISA)

4.2 Historical Trends in PM and Precursor Emissions Contributing to Ambient $PM_{2.5}$ Concentrations in Canada

4.2.1 Canadian emissions inventory

Canada compiles comprehensive emission inventories for the following air pollutants, including precursors and contributors to ambient particulate formation:

- criteria air contaminants: total PM, PM_{10}, $PM_{2.5}$, sulphur oxides (SO_x), NO_x, VOCs, carbon monoxide (CO), and NH_3
- heavy metals: mercury, cadmium and lead
- persistent organic pollutants: four polycyclic aromatic hydrocarbons, dioxins and furans, and hexachlorobenzene

The National Pollutant Release Inventory (NPRI) and Air Pollutant Emissions Inventory (APEI) are the two main sources of air pollutant data compiled by Environment and Climate Change Canada. Facility emissions of air pollutants (from contiguous, portable, pipeline and offshore installations) have been reported under the NPRI since 2002; they form part of the comprehensive air pollutant emissions inventory, which also includes area or non-point source data. Further information and data are available from https://www.ec.gc.ca/inrp-npri/.

Significant changes have been made to the emissions methodologies throughout the 2005–2010 timeframe. The notable changes noted below are reflected in the trends.

- Agricultural NH_3 methodology was changed to the National Agri-Environmental Standards Initiatives method, causing increases in 2006.
- Marine transportation emissions methodologies were changed in 2006.
- Wood product-related PM decreased due to modifications in 2007 as a result of a joint Environment and Climate Change Canada-industry initiative, which revealed that previous methodologies overestimated PM emissions.
- Asphalt VOCs increased due to the inclusion of paving emissions in 2008.
- VOC emissions from refuelling were included since 2008.
- VOC emissions from gasoline service stations were included since 2008.

Emission estimates contain uncertainties that vary considerably from one sector to another and from one pollutant to the next. Open sources (e.g. road dust, construction dust, agriculture) are particularly difficult to quantify because the emissions are highly variable among sites and over time. Sector-specific emissions are often estimated using an emission factor and appropriate activity data. Efforts are being made to reduce uncertainties through continuous monitoring, comparisons to observations, testing and collaboration with provincial agencies and industry.

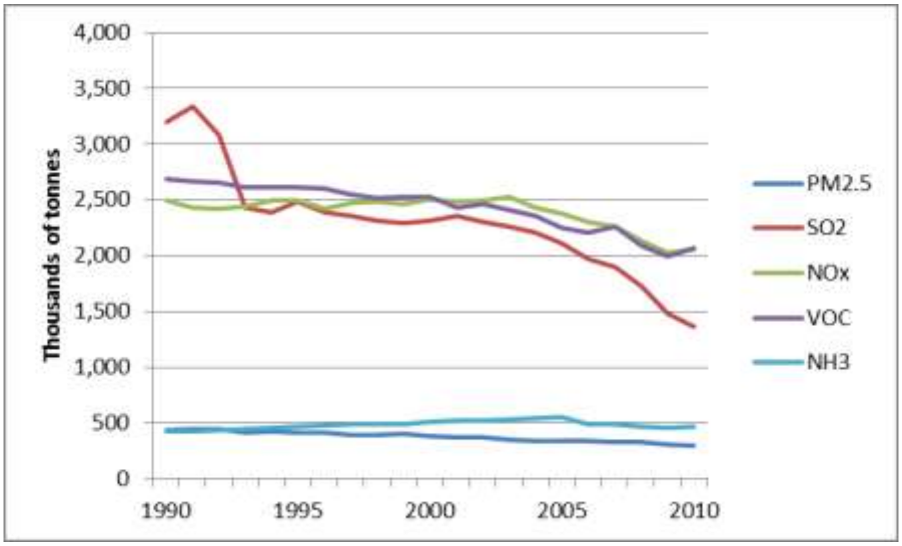
4.2.2 General trends in Canadian emissions

Emissions of most $PM_{2.5}$ precursors decreased between 2002 and 2010, except for NH_3. Total anthropogenic primary $PM_{2.5}$ emissions have remained relatively stable over the same period, as reductions from transportation and some industrial sources are masked by estimated increases in fugitive $PM_{2.5}$ emissions from road dust and construction (Figure 4.2.1).

Large SO_2 reductions between 2002 and 2010 are attributable to technological and process changes in primary non-ferrous (base metal smelting) industries, regulatory changes in the upstream oil and gas sector (oil and gas development, extraction production excluding oil sands prior to refinement) in Alberta, as well as the economic slowdown and facility closures late in the period. Specifically, improvements to milling and smelting processes have resulted in high sulphur containment and chemical conversion of SO_2 to H_2SO_4 for commercial resale. Fuel switching, decreased sulphur in fuels and many other small changes have occurred to further the SO_x emissions reduction.

NO_x emissions steadily declined between 2002 and 2010. This reduction is attributable to more stringent emission regulations in the transportation and electric power generation sectors. However, some of the decreases are currently being masked by increasing emissions from off-road diesel and the upstream (upstream oil and gas and oil sands) and downstream petroleum sectors (refineries and bulk fuel distribution), mainly in Alberta.

Figure 4.2.1: Trends in Canadian emissions of $PM_{2.5}$ and its precursors from 1990 to 2010



Emissions of VOCs also steadily decreased over the same period, largely due to improved on-road and off-road vehicle and engine emission standards. On-road vehicles are fossil fuel-powered cars, trucks, motorcycles, etc. licensed for travel on public roads. Off-road vehicles and engines are fossil fuel-powered equipment and engines ranging from lawnmowers to ATVs to farm tractors to heavy mine haulage trucks and much other engine-powered machinery.

NH_3 emission reductions from non-agricultural sources have occurred in the industrial sources category, mostly in the chemical production sector, which has seen lowered production over the period (Statistics Canada, 2010). Efforts to reduce industrial and commercial NO_x emissions have often led to NH_3 reductions, although increases are being seen from on-road transportation sources.

4.2.3 $PM_{2.5}$ emission trends in Canada

When excluding natural sources, the total emissions of $PM_{2.5}$ in Canada have remained relatively steady: 997,257 tonnes in 2002 and 1,113,515 tonnes in 2010 (Table 4.2.1). Road dust accounted for 48.8% of $PM_{2.5}$ emissions in 2002, and 53.3% in 2010. It should be noted that estimates of $PM_{2.5}$ emissions from road, agricultural and construction dusts are subject to a high degree of uncertainty. If both natural and open sources are excluded, the major emitting sources have decreased from 368,442 tonnes in 2002 to 298,447 tonnes in 2010 (Table 4.2.1).

Other than road dust and construction, the largest sources of primary $PM_{2.5}$ are residential wood combustion and the industrial and agricultural sectors, accounting for approximately two-thirds of the national total. Another important anthropogenic source is off-road diesel. One area of high-density $PM_{2.5}$ emissions is the Windsor – Québec City corridor, resulting mainly from industrial activities, the transportation sector, and residential wood combustion. Major urban centres in western Canada and along Alberta's Calgary – Edmonton corridor are also areas of high-density $PM_{2.5}$ emissions, again likely the result of emissions from the transportation sector.

Natural sources are also important contributors to primary $PM_{2.5}$ emissions. They include forest fires (including arson-related fires), windblown soil and sea salt spray. Forest fires can contribute to primary $PM_{2.5}$ emissions in the boreal forest, and sea salt has an important influence in coastal areas.

Table 4.2.1: Canadian emission inventory of $PM_{2.5}$ in 2002 and 2010 by sector

(a) Excluding natural sources

Sectors	2002		2010	
	Tonnes	%	Tonnes	%
Road Dust	486,196	48.8	593,025	53.3
Construction Operations	132,549	13.3	218,012	19.6
Industrial (incl. fugitives)	101,543	10.2	67,135	6.0
Residential Fuel Wood Combustion	109,778	11.0	104,087	9.3
Agriculture	53,539	5.4	44,082	4.0
Transportation	71,094	7.1	61,062	5.5
Other (Excluding Natural)	42,558	4.3	26,113	2.3
Total:	997,257	100.0	1,113,515	100.0

(b) Excluding natural and open sources

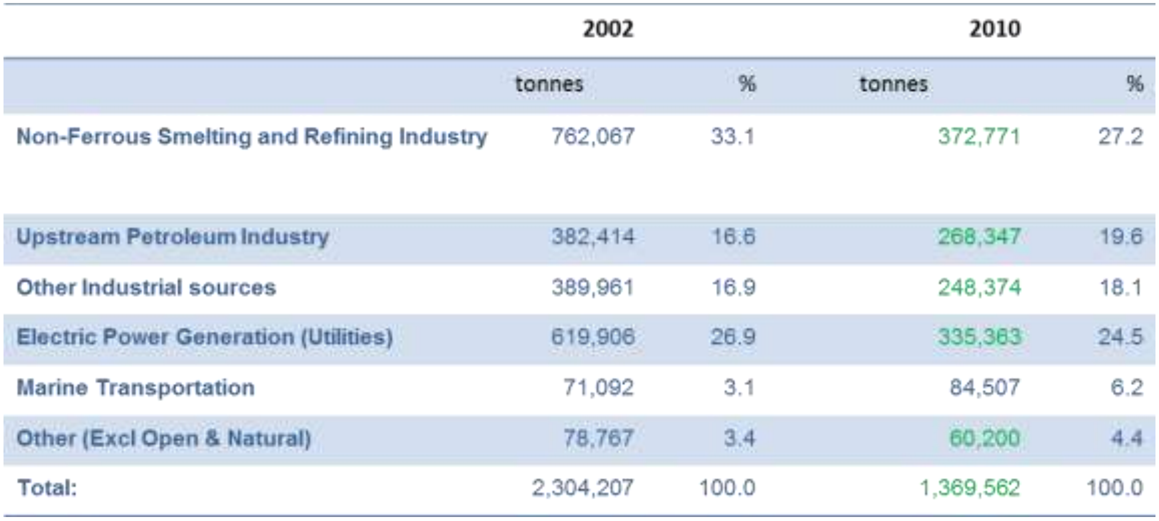
Sectors	2002		2010	
	Tonnes	%	Tonnes	%
Pulp and Paper Industry	17,325	4.7	8,250	2.8
Mining and Rock Quarrying	12,872	3.5	12,193	4.1
Other Industrial	71,346	19.4	46,692	15.6
Residential Fuel Wood Combustion	109,778	29.8	104,087	34.9
Off-road use of diesel	39,810	10.8	31,153	10.4
Other Transportation	31,284	8.5	29,909	10.0
Agriculture	53,539	14.5	44,082	14.8
Other (Excl Open & Natural)	32,488	8.8	22,081	7.4
Total:	368,442	100.0	298,447	100.0

Note: Percentages correspond to percentage of the total emissions. Green values highlight 2010 emissions that are lower than 2002.

4.2.4 $PM_{2.5}$ precursor emission trends in Canada

SO_x: Non-ferrous smelting and refining is the largest source sector of national SO_2 emissions, followed by electricity generation and the upstream and downstream petroleum sectors. Collectively, they account for approximately 96% of Canada's total SO_2 emissions. The highest density of emissions occurs in the Prairies and the Windsor – Québec City corridor; other high emission areas occur where non-ferrous and upstream petroleum industries are located across Canada (Table 4.2.2 below). Emissions of SO_x decreased around 40% between 2002 and 2010.

Table 4.2.2: Canadian emission inventory of SO_x in 2002 and 2010 by sector



	2002		2010	
	tonnes	%	tonnes	%
Non-Ferrous Smelting and Refining Industry	762,067	33.1	372,771	27.2
Upstream Petroleum Industry	382,414	16.6	268,347	19.6
Other Industrial sources	389,961	16.9	248,374	18.1
Electric Power Generation (Utilities)	619,906	26.9	335,363	24.5
Marine Transportation	71,092	3.1	84,507	6.2
Other (Excl Open & Natural)	78,767	3.4	60,200	4.4
Total:	2,304,207	100.0	1,369,562	100.0

Note: Green values highlight 2010 emissions that are lower than 2002.

NO_x: In Canada, transportation accounted for two-thirds of national NO_X emissions in 2010. Upstream oil and gas, and electric power generation are also important source sectors. The highest densities of NO_X emissions are seen in Alberta and the Windsor – Québec City corridor, where the oil and gas and transportation sectors, respectively, are the most prominent sources. (The Windsor–Québec City corridor is Canada's pollutant emission management area under the Ozone Annex of the Air Quality Agreement.) Emissions of NO_x were reduced by 17% between 2002 and 2010 (Table 4.2.3).

Table 4.2.3: Canadian emission inventory of NO_x in 2002 and 2010 by sector

	2002		2010	
	tonnes	%	tonnes	%
Upstream Petroleum Industry	475,538	19.1	434,022	21.1
Heavy-duty diesel vehicles	302,144	12.1	188,913	9.2
Marine Transportation	103,499	4.1	121,642	5.9
Off-road use of diesel	384,723	15.4	425,039	20.6
Other Transportation	556,890	22.3	402,829	19.6
Electric Power Generation (Utilities)	278,874	11.2	208,715	10.1
Other (Excl Open & Natural)	394,565	15.8	278,931	13.5
Total:	2,496,233	100.0	2,060,091	100.0

Note: Green values highlight 2010 emissions that are lower than 2002.

Improved on-road vehicle emission standards have contributed to significant reductions in NO_x emissions in recent years. On-road heavy-duty diesel vehicles have seen increasingly stringent emissions standards for the model-years 1989, 1995, 1998, 2004/2007 (Phase 1) and 2010 (Phase 2).

Fleet emissions will continue to decrease as an increasingly larger percentage of vehicles comply with these standards (due to fleet turn-over). Other regulations implemented during the same period for light duty and off-road engines have also contributed to this overall decrease.

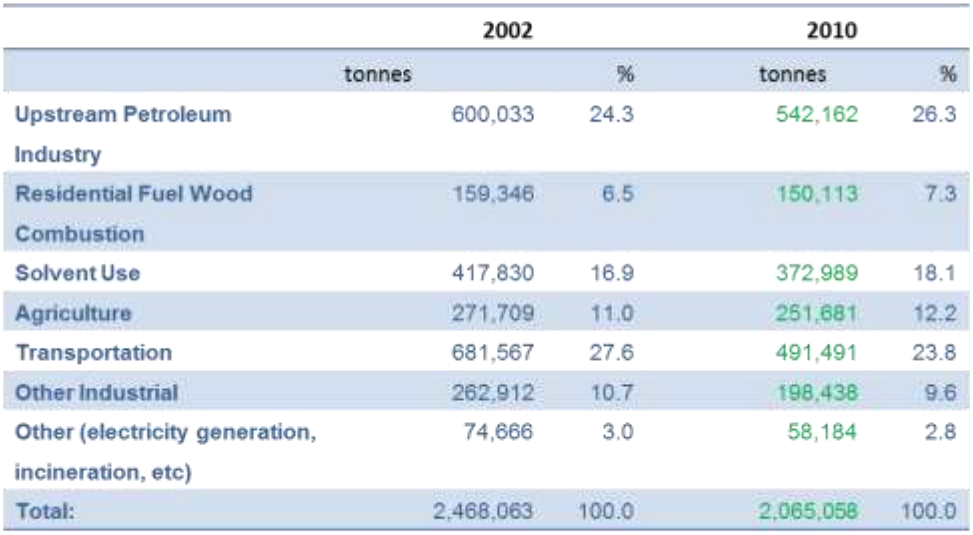
VOCs: These emissions originate predominately from biogenic sources; they represent 85% and 91% of the estimated emissions for 2002 and 2010, respectively. Biogenic sources (e.g. forests, scrub brush, crops, and grasses) contribute to ambient VOC levels. This is especially the case in rural or forested areas, where the proportion of VOCs from natural sources is much higher than that from anthropogenic sources. The upstream petroleum industry, solvent use, and agriculture are the major anthropogenic sources (Table 4.2.4). Overall, anthropogenic sources contribute approximately 9% of national emissions. Geographically, these emission sources are concentrated in Alberta and southern Saskatchewan, primarily due to the petroleum industry.

Table 4.2.4: Canadian emission inventory of VOCs in 2002 and 2010 by sector

(a) Including natural and open sources

	2002		2010	
	tonnes	%	tonnes	%
Biogenics (Vegetation, soils)	16,158,619	84.8	23,032,699	91.3
Upstream Petroleum Industry	600,033	3.1	542,162	2.1
Solvent Use	417,830	2.2	372,989	1.5
Agriculture	271,709	1.4	251,681	1.0
Other	1,602,374	8.4	1,035,363	4.1
Total:	19,050,565	100.0	25,234,894	100.0

(b) Excluding open and natural sources



	2002		2010	
	tonnes	%	tonnes	%
Upstream Petroleum Industry	600,033	24.3	542,162	26.3
Residential Fuel Wood Combustion	159,346	6.5	150,113	7.3
Solvent Use	417,830	16.9	372,989	18.1
Agriculture	271,709	11.0	251,681	12.2
Transportation	681,567	27.6	491,491	23.8
Other Industrial	262,912	10.7	198,438	9.6
Other (electricity generation, incineration, etc)	74,666	3.0	58,184	2.8
Total:	2,468,063	100.0	2,065,058	100.0

Note: Green values highlight 2010 emissions that are lower than 2002.

The reported 43% increase in total VOC emissions between 2002 and 2010 (Table 4.2.4a) is due to modifications in the biogenic VOC estimation method. Excluding biogenic sources, anthropogenic VOC emissions have decreased by about 16% (Table 4.2.4b).

Emissions from the upstream petroleum industry decreased the most between 2002 and 2010, followed by the transportation sector. This was due to changes in on-road light-duty gasoline vehicles (cars, trucks and motorcycles), which were subject to increasingly stringent emission standards for auto model years 1988 (Tier 0), 1994/1996 (Tier 1), 2004/2007 (Tier 2) and year 2006 for motorcycles. Fleet emissions, particularly from heavy-duty diesel on-road vehicle emissions, have also decreased from 2002 to 2010.

A series of regulations came into effect between the model years 1997 and 2010 for small spark-ignition off-road engines. These engines are under 25 horsepower and are powered by fuels that require a spark ignition (as opposed to compression ignition), such as gasoline and natural gas. As engines that comply with the new standards have become an increasingly large proportion of the fleet (due to fleet turnover), VOC emissions have decreased by an estimated 10%.

The NPRI requires reporting of industrial emissions of 60 VOC species to support regulatory efforts and air quality modelling. Environment and Climate Change Canada models are supported by additional data processing using methodologies consistent with those of the EPA. More research and monitoring of individual VOCs and their reactivity is required to more effectively target and reduce their contribution to $PM_{2.5}$.

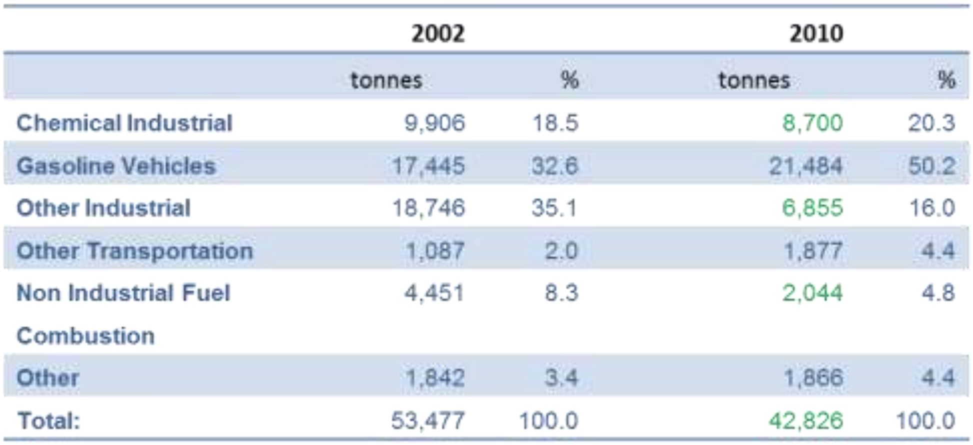
NH_3: The agriculture sector is the most important contributor of NH_3, accounting for 92% of national emissions in 2010. Areas of intense agricultural activity include southern Ontario and Quebec, southern British Columbia, Alberta and Saskatchewan. Nationally, the reduction in NH_3 emissions from 2002 to 2010 was around 13% (Table 4.2.5a).

Table 4.2.5: Canadian emission inventory of NH_3 in 2002 and 2010 by sector

(a) All sources

	2002		2010	
	tonnes	%	tonnes	%
Agriculture (Animals and Fertilizer)	472,753	87.9	420,176	89.6
Transportation	18,532	3.4	23,361	5.0
Industrial Sources	28,652	5.3	15,555	3.3
Other	18,039	3.4	10,108	2.2
Total:	537,976	100.0	469,200	100.0

(b) Excluding natural sources



	2002		2010	
	tonnes	%	tonnes	%
Chemical Industrial	9,906	18.5	8,700	20.3
Gasoline Vehicles	17,445	32.6	21,484	50.2
Other Industrial	18,746	35.1	6,855	16.0
Other Transportation	1,087	2.0	1,877	4.4
Non Industrial Fuel Combustion	4,451	8.3	2,044	4.8
Other	1,842	3.4	1,866	4.4
Total:	53,477	100.0	42,826	100.0

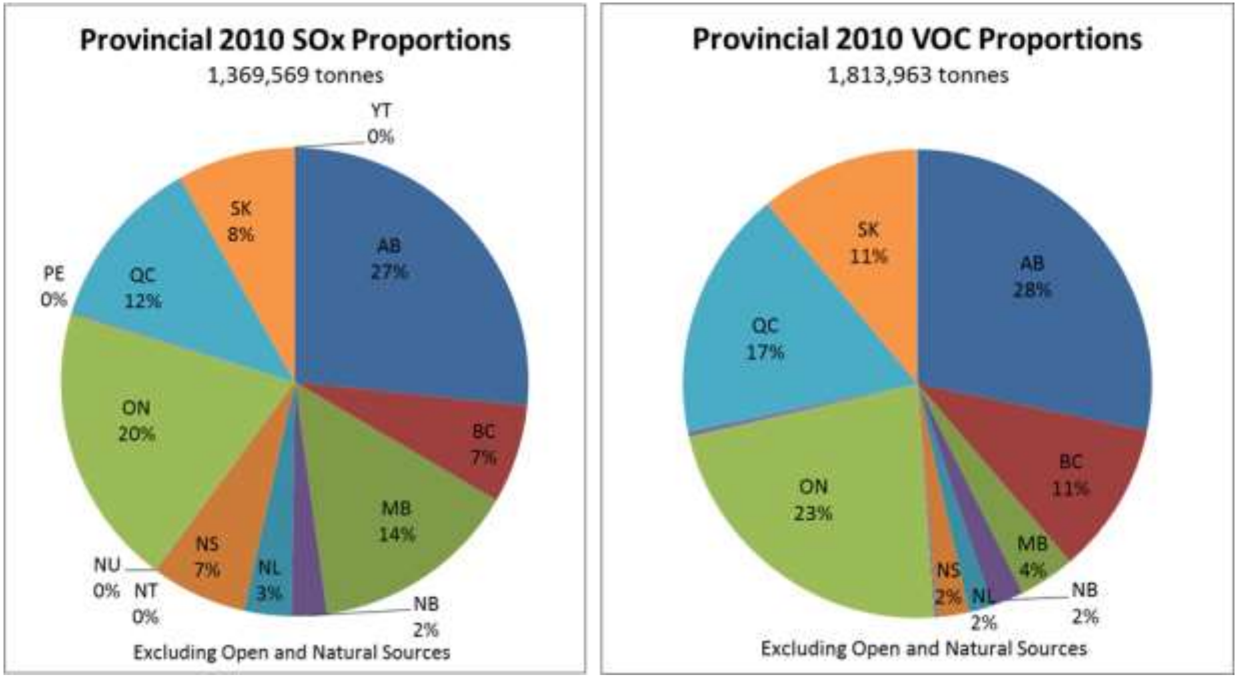
Note: Green values highlight 2010 emissions that are lower than 2002.

Non-agricultural sources of NH_3 emissions include the chemical, industrial, and light-duty transportation sectors, as well as other industrial sources. The 20% reduction from industrial sources (Table 4.2.5b) is driven by the chemical sector's lower production. According to Statistics Canada (2010), total manufacturing sales were lowest in 2009 for the period 2005–2009.

The increase in light-duty vehicle emissions over the period is a function of growth in the number of light-duty vehicles in Canada, which rose from 10,929,000 in 2002 to 12,130,000 in 2010 (Environment Canada, 2012). NH_3 is a by-product of fossil fuel combustion and catalytic converters.

VOC and SO_X by province in 2010: In 2010, over one-quarter of SO_x and VOC emissions in Canada originated in Alberta. Ontario was the second-largest contributor to national emissions, followed by Manitoba for SO_x and Quebec for VOCs. The concentration of Canada's industrial activity in Alberta and Ontario accounts for the relative contributions from each province (Figure 4.2.2).

Figure 4.2.2: Provincial contributions to anthropogenic emissions of SO_x and VOCs in Canada



4.3 Historical Trends in PM and Precursor Emissions Contributing to Ambient $PM_{2.5}$ Concentrations in the United States

4.3.1 United States emissions inventory

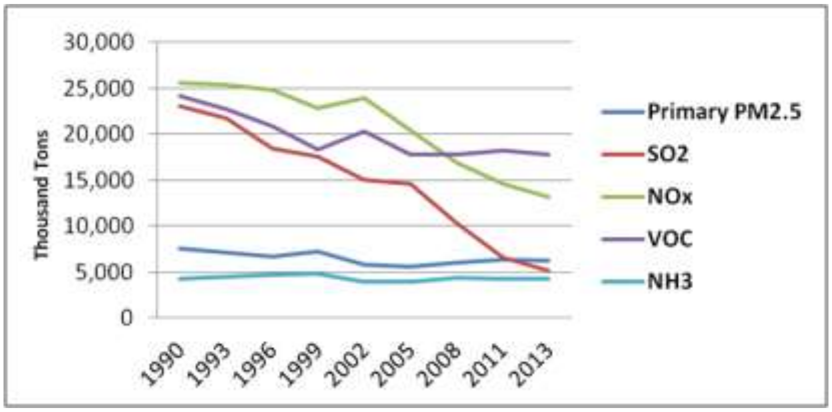
The U.S. compiles national air emissions data in the National Emissions Inventory (NEI). The NEI is a comprehensive and detailed estimate of air emissions classified as Criteria and Hazardous air pollutants (CAPs and HAPs) from all air emission sources. The EPA prepares the NEI every three years, based primarily upon emission estimates and emission model inputs provided by state, local, and tribal air agencies for sources in their jurisdictions. These are supplemented by data from EPA emission programs, including the Toxics Release Inventory, emissions trading programs such as the ARP, and data gathered for EPA development of regulations to reduce air toxic emissions. Using quality assurance procedures, the data from multiple sources are blended together to complete the NEI. The latest version of the NEI available for this Assessment is for the year 2011. Additional information on the NEI and data are available on the NEI website (https://www.epa.gov/air-emissions-inventories).

4.3.2 General trends in United States emissions

Figure 4.3.1 summarizes the trends in primary $PM_{2.5}$ and the major precursors of secondary $PM_{2.5}$ since 1990. Most of the pollutant levels have decreased significantly during this period, with the exception of primary $PM_{2.5}$ and NH_3, which show little change. Since the late 1990s, the rate of decrease is highest for NO_x and SO_2. The EPA updated the mobile source emissions for specific years starting in 2002, using the 2013 version of the emission estimation model called the Motor Vehicle Emissions Simulator. The apparent increase in NO_x and VOCs for 2002 is a methodological artifact. EPA emission control programs that are currently helping areas meet NAAQS and that influence such pollutant reductions include:

- NO_x Budget Program and the Clean Air Interstate Rule;
- Mercury and Air Toxics Standards, which have a significant SO_2 reduction co-benefit;
- Maximum Achievable Control Technology standards, which though intended to reduce HAP emissions have co-benefits for VOC and PM emission reductions;
- Motor vehicle programs for cleaner fuels and engines;
- Non-road engine control and clean fuel programs for small engines, commercial marine vessels, and locomotives.

Figure 4.3.1: Emission trends of primary $PM_{2.5}$ and its precursors in the U.S.

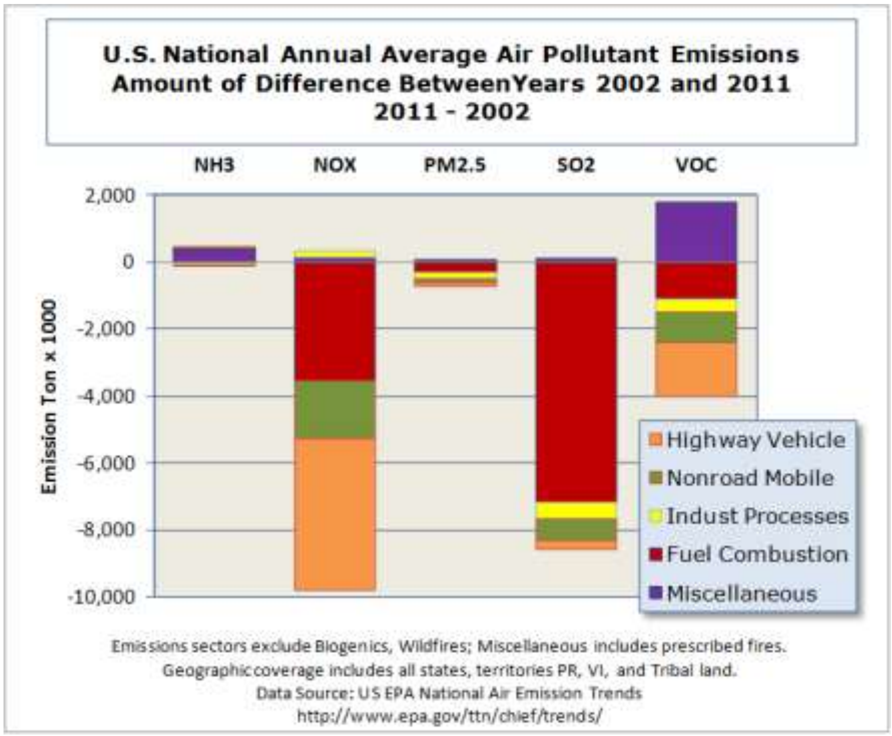


(Source: EPA National Emissions Inventory)

4.3.3 $PM_{2.5}$ emission trends in United States sectors

Figure 4.3.2 below compares emission inventories of primary $PM_{2.5}$ and major precursors of secondary $PM_{2.5}$ for 2002 and 2011. The y-axis shows the emission differences as estimated by subtracting the 2002 emissions from the 2011 emissions. Values greater than zero indicate that 2011 emissions are larger than 2002 values. In addition, Figure 4.3.2 compares emissions for major sectors (excluding wildfires and biogenic emissions). Table 4.3.1 describes the emission changes for each pollutant/sector combination, and Table 4.3.2 identifies the source within the sector that drives the decrease or increase observed by pollutant /sector combination and notes where some differences are also due to method changes. Figure 4.3.2, Table 4.3.1 and Table 4.3.2 illustrate that overall emissions are lower in 2011 than in 2002, though NH_3 shows a slight increase. The exceptions by sector and pollutant are increases in $PM_{2.5}$, SO_2, and NH_3 from the miscellaneous sector and small increments in NO_x from the miscellaneous sector and industrial processes. The $PM_{2.5}$ change in the miscellaneous sector results from an increase in emissions from prescribed burning.

Figure 4.3.2: Differences in emissions by major sector in the U.S. between 2002 and 2011



Note: NH_3 = ammonia, NO_x = nitrogen oxides, SO_2 = sulphur dioxide, VOC = volatile organic compounds

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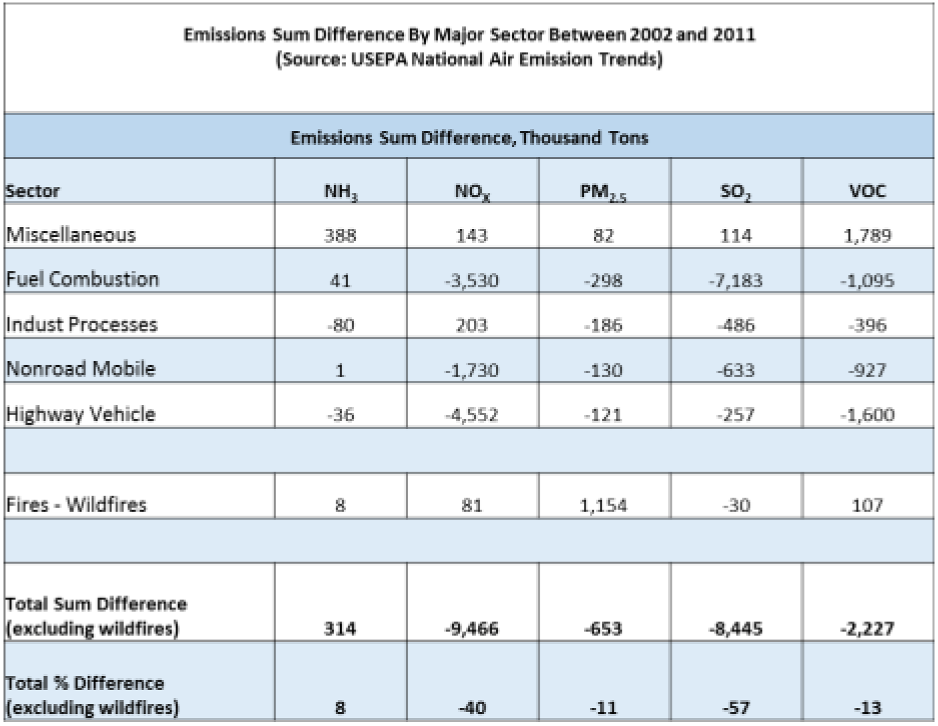
Table 4.3.1: Emissions sum difference by major sector between 2002 and 2011

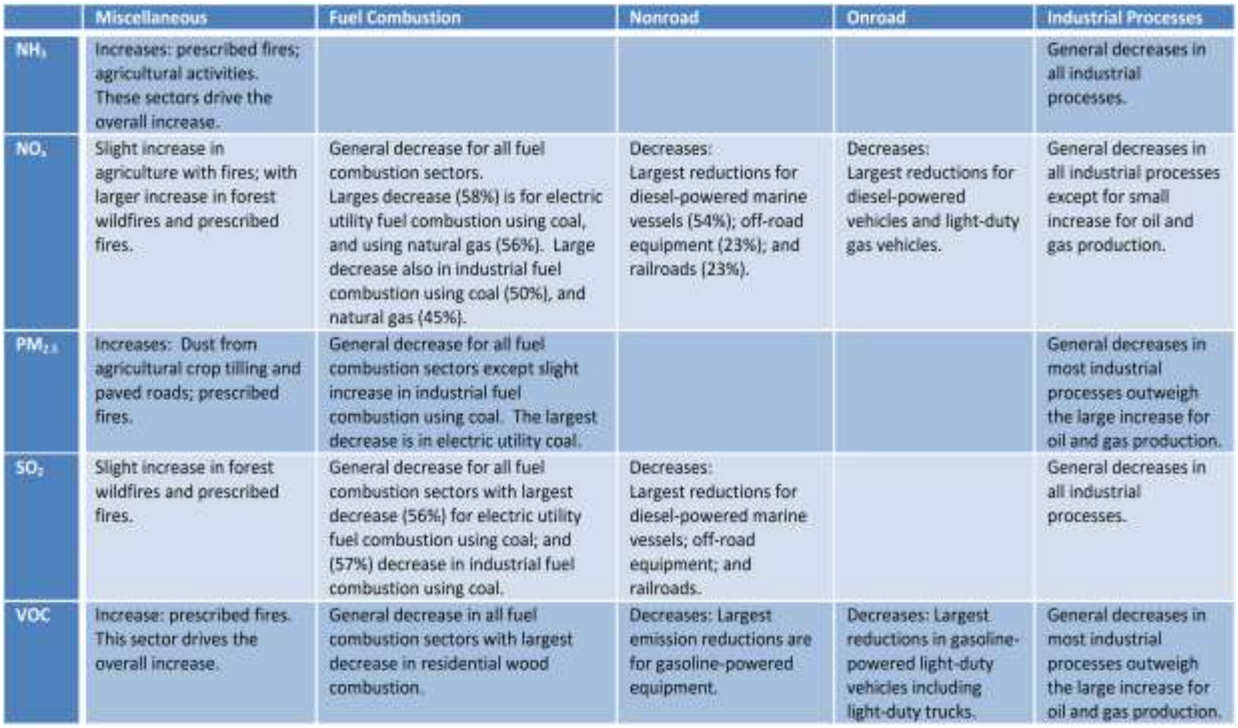


Emissions Sum Difference By Major Sector Between 2002 and 2011
(Source: USEPA National Air Emission Trends)

Emissions Sum Difference, Thousand Tons					
Sector	NH_3	NO_x	$PM_{2.5}$	SO_2	VOC
Miscellaneous	388	143	82	114	1,789
Fuel Combustion	41	-3,530	-298	-7,183	-1,095
Indust Processes	-80	203	-186	-486	-396
Nonroad Mobile	1	-1,730	-130	-633	-927
Highway Vehicle	-36	-4,552	-121	-257	-1,600
Fires - Wildfires	8	81	1,154	-30	107
Total Sum Difference (excluding wildfires)	**314**	**-9,466**	**-653**	**-8,445**	**-2,227**
Total % Difference (excluding wildfires)	**8**	**-40**	**-11**	**-57**	**-13**

(Source: EPA National Air Emission Trends)

Table 4.3.2: Explanation of the differences in emissions between 2002 and 2011



	Miscellaneous	Fuel Combustion	Nonroad	Onroad	Industrial Processes
NH_3	Increases: prescribed fires; agricultural activities. These sectors drive the overall increase.				General decreases in all industrial processes.
NO_x	Slight increase in agriculture with fires; with larger increase in forest wildfires and prescribed fires.	General decrease for all fuel combustion sectors. Larges decrease (58%) is for electric utility fuel combustion using coal, and using natural gas (56%). Large decrease also in industrial fuel combustion using coal (50%), and natural gas (45%).	Decreases: Largest reductions for diesel-powered marine vessels (54%); off-road equipment (23%); and railroads (23%).	Decreases: Largest reductions for diesel-powered vehicles and light-duty gas vehicles.	General decreases in all industrial processes except for small increase for oil and gas production.
$PM_{2.5}$	Increases: Dust from agricultural crop tilling and paved roads; prescribed fires.	General decrease for all fuel combustion sectors except slight increase in industrial fuel combustion using coal. The largest decrease is in electric utility coal.			General decreases in most industrial processes outweigh the large increase for oil and gas production.
SO_2	Slight increase in forest wildfires and prescribed fires.	General decrease for all fuel combustion sectors with largest decrease (56%) for electric utility fuel combustion using coal; and (57%) decrease in industrial fuel combustion using coal.	Decreases: Largest reductions for diesel-powered marine vessels; off-road equipment; and railroads.		General decreases in all industrial processes.
VOC	Increase: prescribed fires. This sector drives the overall increase.	General decrease in all fuel combustion sectors with largest decrease in residential wood combustion.	Decreases: Largest emission reductions are for gasoline-powered equipment.	Decreases: Largest reductions in gasoline-powered light-duty vehicles including light-duty trucks.	General decreases in most industrial processes outweigh the large increase for oil and gas production.

The EPA believes that the changes in fire emissions are based primarily on actual changes seen in fire activity, though some method differences exist between 2002 and 2011. Figure 4.3.3 shows the trend (lower 48 states only) in $PM_{2.5}$ emissions from prescribed fires and wildfires (together called Wildland fires) from 2007 to 2011, all based on the “SMARTFIRE2” model.

The emission changes reflect actual activity differences in area burned, as identified by either national default databases or by activity data that individual states submit. The 2011 data benefited from a significant review and contribution of activity data from state forestry agencies. The U.S. Forest Service National Interagency Fire Center database[6] shows activity data on a year-to-year basis and confirms the emission trends from fires seen in the NEI data.

Figure 4.3.3 additionally shows that 2011 saw the second highest $PM_{2.5}$ fire emissions over the time range shown and reveals that from year to year, the variation in total fire emissions is caused primarily by changes in wildfire activity.

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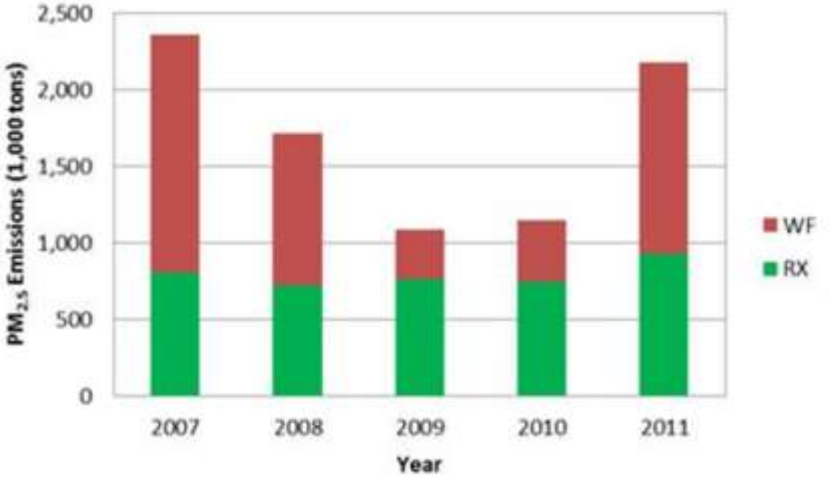
[6] The database is available online at https://www.nifc.gov/fireInfo/fireInfo_statistics.html.

Figure 4.3.3: PM2.5 trends from wild/natural and prescribed fires, 2007–2011

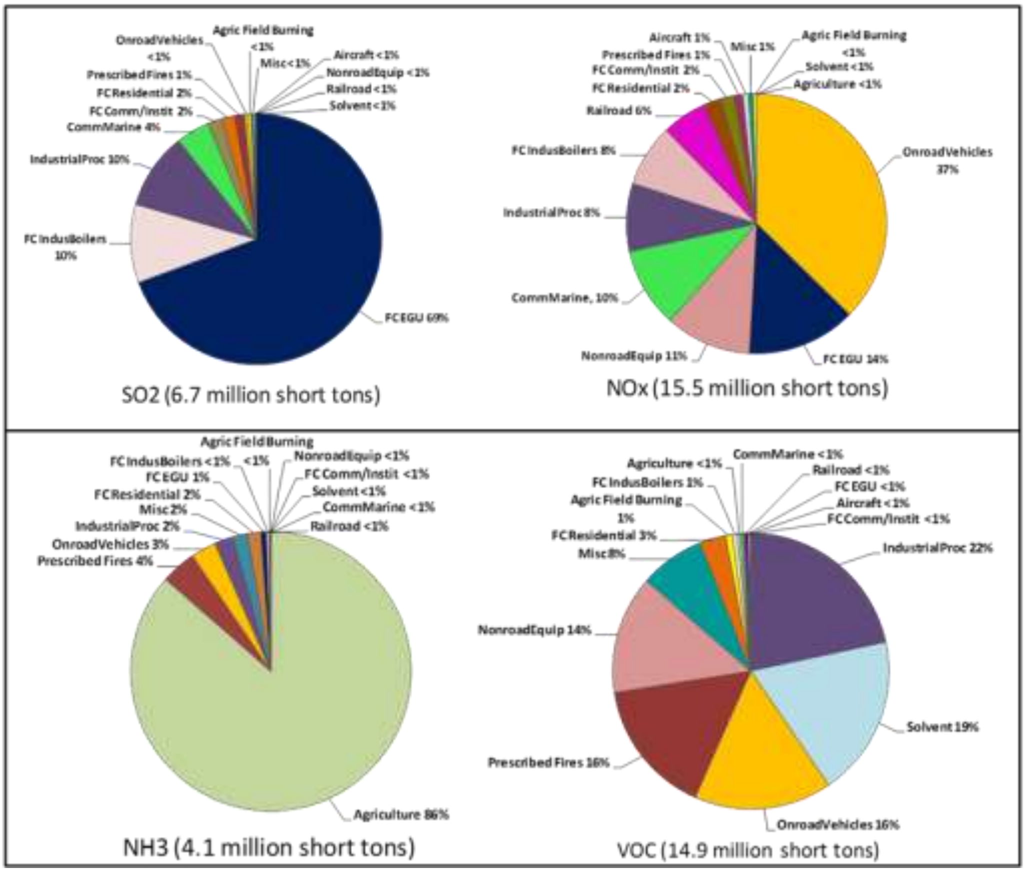


Note: WF = wild/natural fires, RX = prescribed fires

4.3.4 PM$_{2.5}$ precursor emissions in the United States

Figure 4.3.4 shows, in detail, the sources contributing to emissions of PM precursors: SO_2, NO_x, NH_3, and VOCs, according to the 2011 NEI, version 1 (EPA, 2014). The largest 2011 U.S. source of SO_2 emissions by far (69%) was electric generating units. They were also a significant contributor (14%) to NO_x emissions in 2011, but mobile sources were the largest contributors, with on-road mobile sources contributing 37% and other mobile sources (non-road equipment, aircraft, commercial marine, and railroad) contributing another 28%. For NH_3, the largest source by far was agricultural practices, while several different sources (industrial processes, solvent use, on-road vehicles, non-road equipment, and agricultural/prescribed fires) contributed significantly to emissions of VOCs.

Figure 4.3.4: U.S. national average emissions of gaseous precursor species by source category for 2011



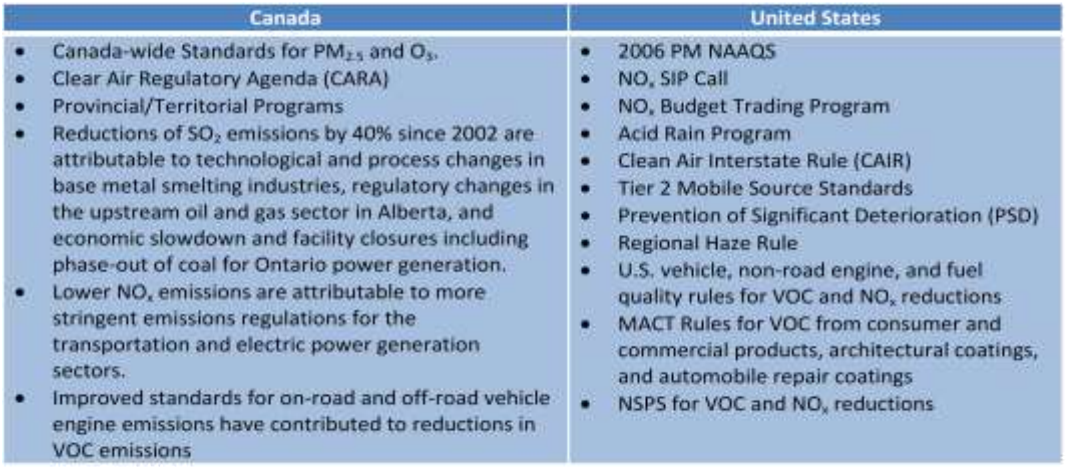
Note: SO_2 = sulphur dioxide, NO_X = nitrogen oxides, NH_3 = ammonia, VOCs = volatile organic compounds

4.4 Comparison Between the 2004 Projections and the Observed Emission Trends

Linking past policies to observed changes in air pollutant concentrations and deposition amounts is an important step, providing a measure of accountability and lending credibility to the past emission projections. Hidy et al. (2011b) assessed this issue in relation to emission reduction policies and the resulting changes in O_3, acidic deposition, PM and toxics from the 1980s or 1990s and the early 2000s. They also assessed whether it was possible to determine if the actual intended benefits of these changes (i.e. improved public health, recovering ecosystems) could be demonstrated, thus completing the accountability chain.

The $PM_{2.5}$ concentration decreases in Canada and the U.S. shown in Chapter 3 suggest the emission reduction policies that both countries have implemented since the 2004 Assessment have been beneficial for air quality, thus providing evidence of accountability. Table 4.4.1 below summarizes the various air quality management policies that have led to the decreases in emissions in both countries that were discussed above. The impact of the air quality management policies in the U.S. and Canada can also be seen from the emission values shown in Tables 4.4.2 and 4.4.3. These tables present the differences between emission levels projected for 2010 in the 2004 Assessment and the actual emissions reported for 2010.

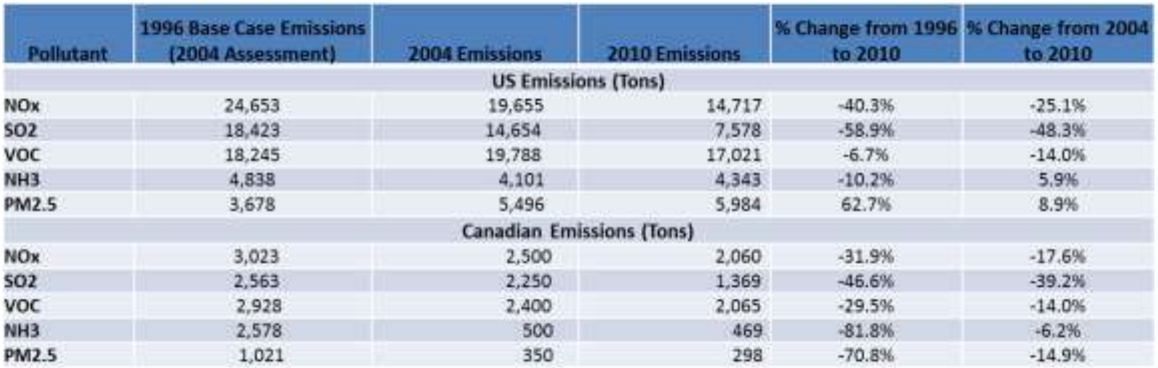
Table 4.4.1: Air quality management policies in Canada and the U.S. affecting emissions since 2004



Canada	United States
• Canada-wide Standards for PM$_{2.5}$ and O$_3$. • Clear Air Regulatory Agenda (CARA) • Provincial/Territorial Programs • Reductions of SO$_2$ emissions by 40% since 2002 are attributable to technological and process changes in base metal smelting industries, regulatory changes in the upstream oil and gas sector in Alberta, and economic slowdown and facility closures including phase-out of coal for Ontario power generation. • Lower NO$_x$ emissions are attributable to more stringent emissions regulations for the transportation and electric power generation sectors. • Improved standards for on-road and off-road vehicle engine emissions have contributed to reductions in VOC emissions	• 2006 PM NAAQS • NO$_x$ SIP Call • NO$_x$ Budget Trading Program • Acid Rain Program • Clean Air Interstate Rule (CAIR) • Tier 2 Mobile Source Standards • Prevention of Significant Deterioration (PSD) • Regional Haze Rule • U.S. vehicle, non-road engine, and fuel quality rules for VOC and NO$_x$ reductions • MACT Rules for VOC from consumer and commercial products, architectural coatings, and automobile repair coatings • NSPS for VOC and NO$_x$ reductions

* * * * *

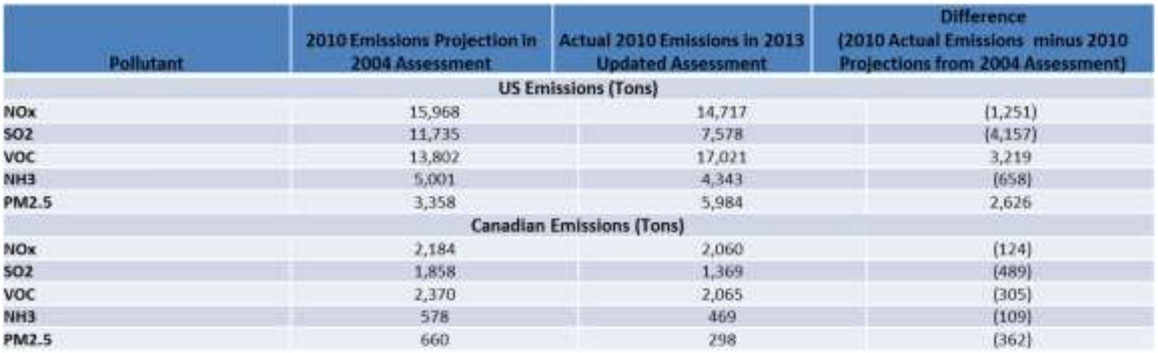
Table 4.4.2: Comparison of 2010 emissions projections in 2004 Assessment to actual 2010 emissions inventory



Pollutant	1996 Base Case Emissions (2004 Assessment)	2004 Emissions	2010 Emissions	% Change from 1996 to 2010	% Change from 2004 to 2010
		US Emissions (Tons)			
NOx	24,653	19,655	14,717	-40.3%	-25.1%
SO2	18,423	14,654	7,578	-58.9%	-48.3%
VOC	18,245	19,788	17,021	-6.7%	-14.0%
NH3	4,838	4,101	4,343	-10.2%	5.9%
PM2.5	3,678	5,496	5,984	62.7%	8.9%
		Canadian Emissions (Tons)			
NOx	3,023	2,500	2,060	-31.9%	-17.6%
SO2	2,563	2,250	1,369	-46.6%	-39.2%
VOC	2,928	2,400	2,065	-29.5%	-14.0%
NH3	2,578	500	469	-81.8%	-6.2%
PM2.5	1,021	350	298	-70.8%	-14.9%

* * * * *

Table 4.4.3: Comparison of 2010 emissions projections in 2004 Assessment to actual 2010 emissions inventory



Pollutant	2010 Emissions Projection in 2004 Assessment	Actual 2010 Emissions in 2013 Updated Assessment	Difference (2010 Actual Emissions minus 2010 Projections from 2004 Assessment)
	US Emissions (Tons)		
NOx	15,968	14,717	(1,251)
SO2	11,735	7,578	(4,157)
VOC	13,802	17,021	3,219
NH3	5,001	4,343	(658)
PM2.5	3,358	5,984	2,626
	Canadian Emissions (Tons)		
NOx	2,184	2,060	(124)
SO2	1,858	1,369	(489)
VOC	2,370	2,065	(305)
NH3	578	469	(109)
PM2.5	660	298	(362)

Table 4.4.2 shows that in both the U.S. and Canada, there were significant reductions in all pollutant emissions from 1996 to 2004. Table 4.4.3 shows the difference between projected 2010 emissions in the 2004 Assessment and actual 2010 emissions in both the U.S. and Canada. As can be seen, the actual emissions in 2010 were lower in all cases in Canada and for NO$_x$, SO$_2$ and NH$_3$ in the

U.S., while actual emissions of direct $PM_{2.5}$ and VOCs were slightly higher in the U.S. This table demonstrates that projections made in the 2004 Assessment for the next six years were reasonably reliable and that the emission reduction policies in both the U.S. and Canada were effective in achieving these anticipated outcomes. It also provides a basis for some confidence in the emission projections presented in the next section. However, the farther into the future the projections are made, the greater the uncertainty, given the difficulty in anticipating all the factors influencing air pollutant emissions decades into the future.

4.5 Projected $PM_{2.5}$ and Precursor Emissions in Canada and the United States to 2020 and 2030

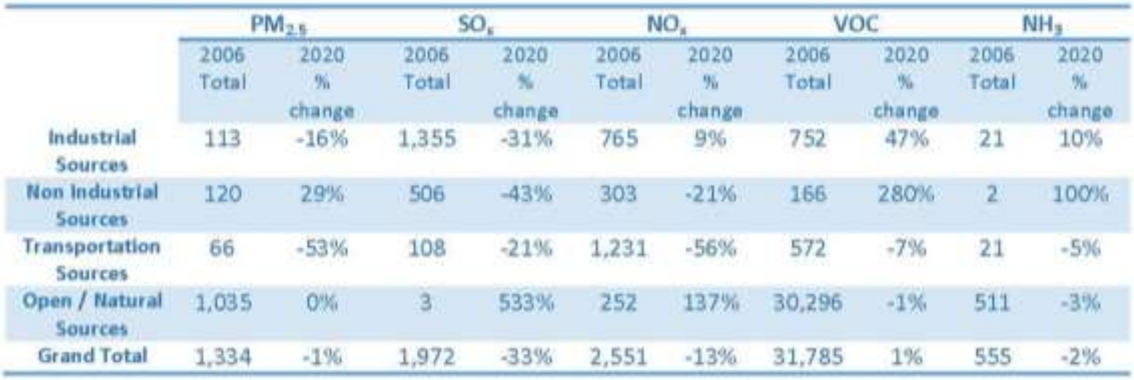
4.5.1 Canada

The emission projections are developed by Environment and Climate Change Canada using the Energy, Emissions and Economy Model for Canada (E3MC) and are based on the 2010 NEI and historical trends. Growth rates for key drivers such as economic and population growth, energy market trends and productivity are aligned with official forecasts from Finance Canada, Statistics Canada, the National Energy Board and Natural Resources Canada.

A Business-as-Usual (BAU) case was developed for the E3MC model that incorporated emission control policies and regulations in place as of summer 2012. Major federal and provincial policies that were considered include, but are not limited to, the Ozone Annex of the Agreement, the Canada-Wide Acid Rain Strategy, the Canada-Wide Standard for Mercury Emissions from coal-fired electric power generation plants, and on-road vehicle and engine emission regulations. The Base Level Industrial Emission Requirements proposals, which are being developed as part of the Air Quality Management System adopted by the CCME in October 2012, were not included in the BAU.

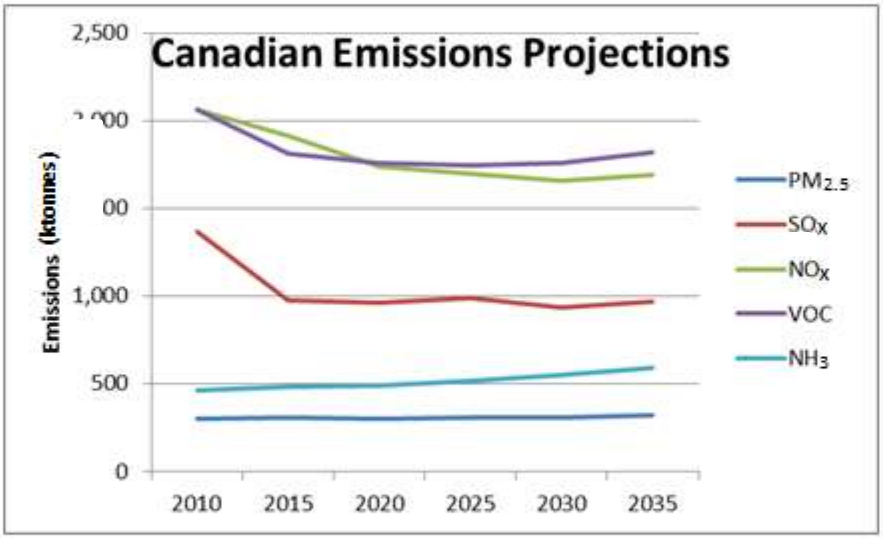
The national BAU emission projections for all sources are summarized by major pollutant in Table 4.5.1. Generally, the model projects minor increases in $PM_{2.5}$ and NH_3, and decreases in VOCs and SO_2 and NO_x emissions (Figure 4.5.1), excluding open and natural sources but including agriculture.

Table 4.5.1: Candian 2006 base-case emissions (ktonnes) and 2020 projected percent change business-as-usual (BAU) emissions by major catergories



	$PM_{2.5}$		SO_x		NO_x		VOC		NH_3	
	2006 Total	2020 % change	2006 Total	2020 % change	2006 Total	2020 % change	2006 Total	2020 % change	2006 Total	2020 % change
Industrial Sources	113	-16%	1,355	-31%	765	9%	752	47%	21	10%
Non Industrial Sources	120	29%	506	-43%	303	-21%	166	280%	2	100%
Transportation Sources	66	-53%	108	-21%	1,231	-56%	572	-7%	21	-5%
Open / Natural Sources	1,035	0%	3	533%	252	137%	30,296	-1%	511	-3%
Grand Total	1,334	-1%	1,972	-33%	2,551	-13%	31,785	1%	555	-2%

Figure 4.5.1: Projected emissions of major pollutants in Canada from 2010 to 2035, excluding open and natural sources



Note: SO_x = sulphur oxides, NO_X = nitrogen oxides, VOC = volatile organic compounds, NH_3 = ammonia

Emissions of primary $PM_{2.5}$ across Canada are projected to be slightly larger, increasing by 1% to 2020. By sector, reductions in transportation and industrial sector emissions are forecast, but these are offset by increases in non-industrial sources such as agriculture and residential wood burning.

VOC emissions are predicted to decrease by 15% between 2010 and 2020, largely due to reductions in the transportation and upstream oil and gas sectors, offset by increases in residential wood combustion and solvents.

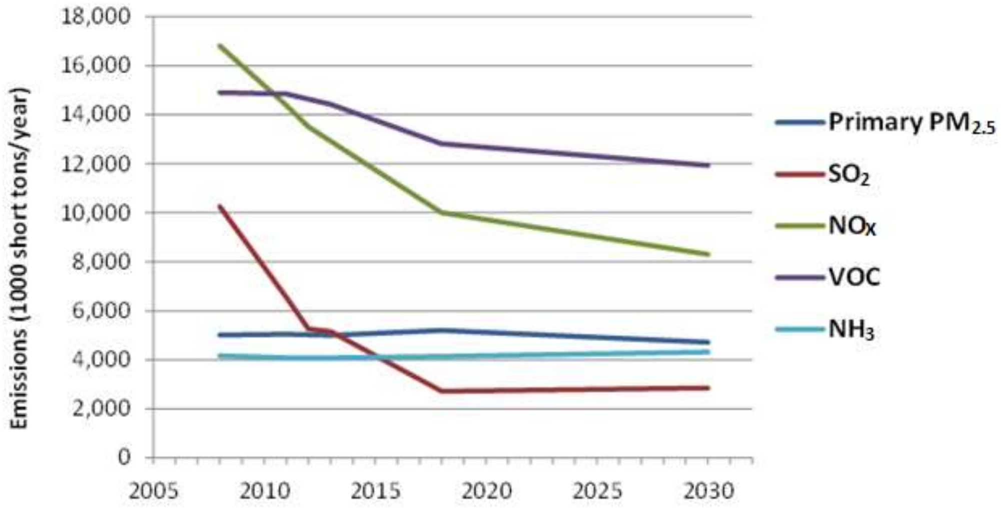
The largest reductions are projected for SO_x and NO_x due to sector-specific emission controls and facility closures. SO_2 emissions are expected to decrease by 30% between 2010 and 2020 because of closures in the non-ferrous smelting and refining sector and emission decreases from fossil fuel-fired electricity generation. These SO_x reductions are predicted to be partially offset by increases from the upstream oil and gas sectors and oil sands activities in Alberta.

Nitrogen dioxide emissions are projected to decline by 16% between 2010 and 2020. Reductions in the transportation sector and fossil fuel-fired electricity generation are offset by anticipated increases in the upstream oil and gas sectors and oil sands activities in Alberta.

4.5.2 United States

Figure 4.5.2 and Table 4.5.2 show projections of U.S. emissions for $PM_{2.5}$ and its major precursors from 2008 through 2030. The emissions in Figure 4.5.2 include all anthropogenic emission sources available from the U.S. inventory data, excluding emissions from Puerto Rico, the U.S. Virgin Islands, tribal lands, and federal waters. In addition, the wildfires and biogenic emissions are excluded entirely, to allow anthropogenic trends to be observed. Wildfire emission trends from 2003 through 2011 are available separately from the 2008 NEI Report (Rao et al, 2013). The categories used in Table 4.5.2 are derived from the Tier 1 categories used traditionally to summarize the U.S. emissions inventory, excluding wildfires, plant transpiration, and soil emission sources. These categories are groups of source category codes (SCCs) used in EPA's Emissions Inventory System (EIS). The SCCs are available on the "EIS Code Tables (including SCCs)" link provided at the EPA's main emissions inventory webpage, www.epa.gov/ttn/chief/eiinformation.html.

Figure 4.5.2: U.S. emission projections, 2008–2030



Sources: 2008: 2008 National Emissions Inventory (NEI), version 3 (US EPA, 2012);
2011-2013: NEI Emisisons Trends Data (http://www.epa.gov/ttn/chief/trends/index.html), December 2013, which includes 2011 NEI version 1;
2018: 2018 emissions projections (case 2018ed), released January 2014, as published for public comment through the Federal Register Vol. 79, No. 9, pg 2437 (1/14/2014); (Mason et al., 2014)
2030: 2018 carried forward, except for 2030 projections used for onroad mobile, nonroad equipment, aircraft, locomotives, commercial marine vessels, and animal waste. 2030 onroad emissions are consistent with baseline emissions used for the Proposed Tier3 rule (http://www.epa.gov/otaq/tier3.htm) and do not include the impact of Tier 3 vehicle and fuel standards.

The Tier 1 categories are grouped into the categories above as follows.

"Fuel Combustion" includes combustion from electricity generation, industrial, commercial, and institutional combustion, residential heating, and residential wood combustion. "Other Industrial Sources" includes Tier 1 categories for chemical and allied products manufacturing, metals processing, petroleum and related industries, and other industrial processes excluding solvents. "Miscellaneous" excludes wildfires, but includes prescribed burning, agricultural burning, solvents, storage and transport (e.g. gas stations, tank farms, materials transfer), waste disposal, road dust, fertilizer application, livestock waste, and others. "Transportation" sources include on-road vehicles, non-road equipment, planes, locomotives, and commercial marine. Notable exclusions from the inventory are geogenic sources, emissions from oceans, lightning NO_X, and windblown dust.

Table 4.5.2: U.S. 2008 emissions (1000 short tons) by major categories and 2018 projected percentage change, including on-the-books controls

	PM2.5		SO2		NOx		VOC		NH3	
	2008 Total	2018 % change	2008 Total	2018 % change	2008 Total	2018 % change	2008 Total	2018 % change	2008 Total	2018 % change
Fuel Combustion	835	0%	9,152	-78%	5,127	-34%	557	15%	104	19%
Other Industrial Sources	446	-16%	764	-32%	981	32%	2,232	46%	78	2%
Miscellaneous, no Wildfires	3,244	15%	97	41%	279	23%	6,524	-4%	3,836	0%
Transportation Sources	489	-48%	240	-83%	10,426	-52%	5,598	-52%	141	-36%
Grand Total	5,014	4%	10,254	-74%	16,813	-40%	14,911	-14%	4,160	0%

(Source: 2008 National Emissions Inventory, version 3 and 2018 baseline emissions from EPA version 6 modelling platform, released January 2014 (Mason et al., 2014))

Jointly, Figure 4.5.2 and Table 4.5.2 show that emissions of SO_2, NO_x, and VOCs are forecast to decrease substantially from 2008 to 2030 (all cited in short tons). SO_2 emissions have already diminished from 10.3 million tons in 2008 to 6.5 million tons in 2011, and SO_2 is expected to decrease further to 2.8 million tons by 2030. NO_x emissions have declined from 16.8 million tons in 2008 to 14.4 million in 2011, and NO_x is expected to further decrease to 8.3 million tons by 2030. The expected reductions to 2030 are a result of numerous existing stationary and mobile source regulatory programs and other changes (Mason et al., 2013). For SO_2, the reductions come primarily from stationary fuel combustion, resulting from the Clean Air Interstate Rule and the Mercury and Air Toxics Standards, as well as from continued use of natural gas for electric generating units (EPA, 2013a). VOC emissions have held constant at 14.9 million tons from 2008 through 2011, but could decrease further to 11.9 million tons by 2030. The VOC trends are dominated by reductions in mobile sources, but also are impacted by oil and gas emissions.

For the oil and gas sector, the most complete estimates are provided for 2011 and 2018, while excluding possible additional future increases in 2030. Furthermore, 2008 emissions were not complete for oil and gas sources, resulting in a methods-related increase for this sector from 2008 to 2011. Emission forecasts of $PM_{2.5}$ show a 4% increase from 5 million to 5.2 million tons per year between 2008 and 2018, with industrial and mobile source reductions being offset by a 300-thousand-ton annual increase in emissions from prescribed burning from 2008 to 2011. The decrease in $PM_{2.5}$ emissions estimated for 2030 is highly uncertain and could be entirely due to emission measurement method differences. Finally, emissions of NH_3 are relatively constant (around 4.1 to 4.3 million tons), with variations being due predominantly to differences in methods used for estimating emissions from agricultural sources rather than expected changes.

CHAPTER 5: Evidence of transboundary transport of $PM_{2.5}$ and the impact of predicted changes in $PM_{2.5}$ emission levels

Chapter Summary

- Updated observations and modelling since 2004 provide ongoing evidence that transboundary transport of $PM_{2.5}$ and its precursors continues to occur across the Canada – U.S. border.
- In most Canadian cities, the dominant source of $PM_{2.5}$ is from Canadian emissions; similarly, in U.S. cities the dominant contributor to $PM_{2.5}$ is domestic emissions.
- Modelling analyses of the impact of future emission projections show significant anticipated reductions in ambient $PM_{2.5}$ concentrations between 2006 and 2020 in both Canada and the U.S.
- Overall, the extent of transboundary $PM_{2.5}$ transport relative to each country's own contributions is projected for 2020 to decrease slightly in both Canada and the U.S. (relative to 2006) due to future emission reductions.
- A modelling analysis done to support the review of the U.S. PM NAAQS shows that projected U.S. emission changes in 2020 will result in reductions in $PM_{2.5}$ concentrations along the Canada – U.S. border. In addition, this analysis predicts that all U.S. counties with air monitors along the Canada – U.S. border will be in attainment with the revised primary annual $PM_{2.5}$ NAAQS of 12 $\mu g/m^3$.
- In terms of Canadian standards, future predictions of ambient concentrations in the form of the annual average and 24-h CAAQS metrics indicate that most areas near the southern Ontario and southern Quebec borders with the U.S. will be below the 2015 CAAQS. Possible exceptions include areas with relatively large local emissions that add to the rural/regional levels, forecast to be around 6 $\mu g/m^3$ (annual average). Ongoing emission reductions from 2016 to 2020 are likely to help limit the number of Canadian locations exceeding the 2020 CAAQS. Over the border regions of western Canada and Atlantic Canada, projections of $PM_{2.5}$ suggest that levels will be below the CAAQS for both 2015 and 2020.

5.1 Evidence of Transboundary Transport

Transboundary transport of PM was documented in the 2004 Assessment. To provide a quantitative assessment of more recent (post-2010) levels of transboundary transport would require targeted analysis, including both an assessment of current observations and modelling analyses using up-to-date emissions (see Chapter 7). Given that this assessment relies on pre-existing analyses, a quantitative assessment of the magnitude or extent of current transboundary transport is not possible. However, the analyses highlighted here provide insight on the continued transboundary transport of $PM_{2.5}$ across the Canada – U.S. border. Furthermore, while Chapter 4 showed that emissions in the U.S. and Canada have declined since the analyses described in this section were undertaken, the spatial distribution of the main source areas and the meteorological patterns are expected to have been relatively invariant over the period. Thus, the features of transboundary transport discussed below are qualitatively representative and also remain consistent with the evidence presented in the 2004 Assessment.

5.1.1 Review of observational evidence

Transboundary transport occurs on multiple scales, from local to regional. Different measurement approaches (e.g. time resolution) and meteorological analyses are therefore applied. Traditionally, the focus has been on regional scale transport over central and eastern sections of the international border. In these situations, 24-h average observations and multi-day trajectories are generally sufficient. For cases of local transport associated with complex meteorology and terrain, much more time-resolved $PM_{2.5}$ and precursor gas measurements and more detailed meteorological analyses are involved.

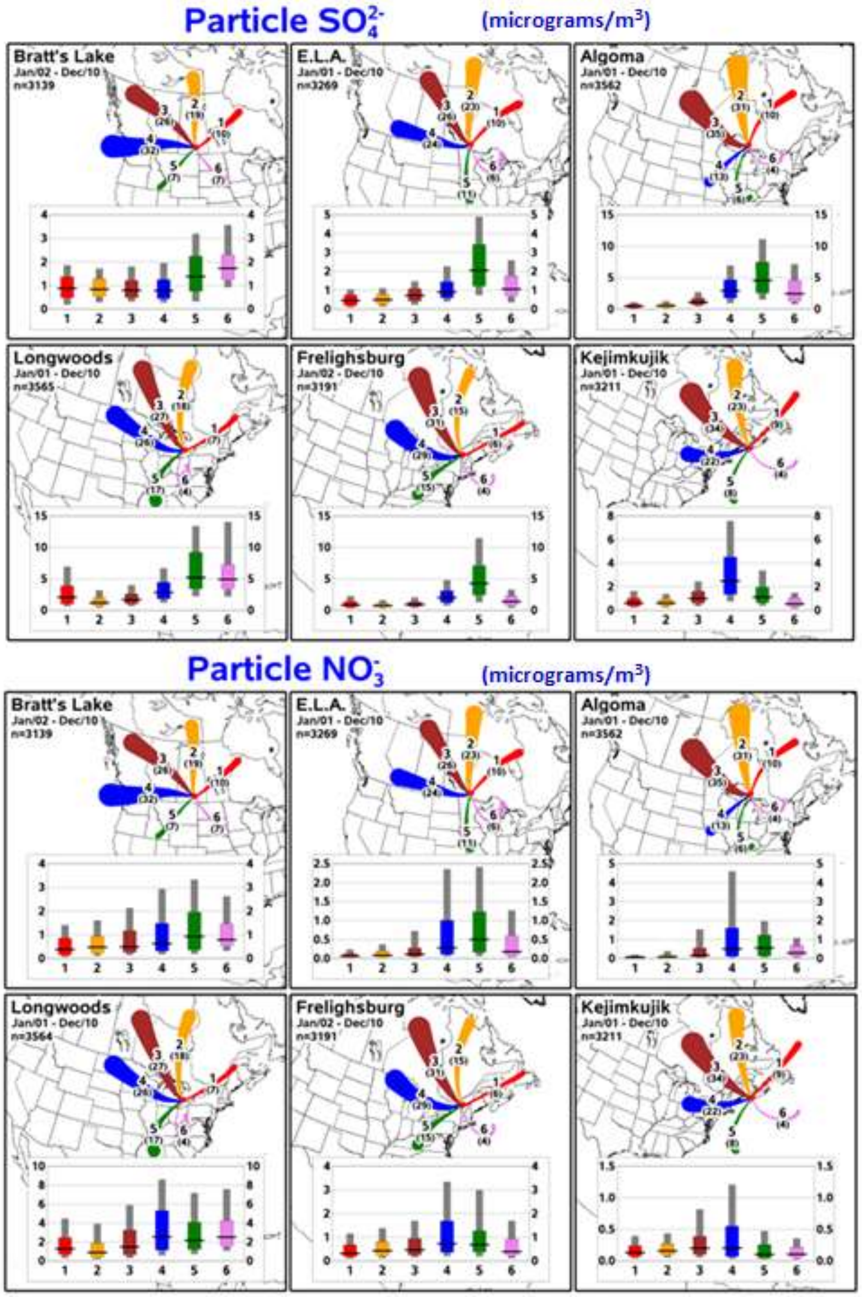
The results of back-trajectory analyses for particle SO_4^{2-} and particle NO_3^-, two major components of $PM_{2.5}$ in Canada, are shown in Figure 5.1.1. Back-trajectory “clusters” delineating common air mass transport pathways were created and matched with 24-h integrated concentrations measured at six CAPMoN monitoring sites in Canada. Nine or 10 years (January 2001 or 2002 to December 2010) of particle SO_4^{2-} and NO_3^- observations were used and are shown in the figure. Although these particle species measurements were not size-selective for $PM_{2.5}$, historical studies have shown that SO_4^{2-} and NO_3^- particles in these areas are predominantly in the $PM_{2.5}$ range, except for NO_3^- at Kejimkujik, NS, which is highly affected by coarse particle NO_3^- (Zhang et al., 2008). The trajectory plots show the percentage of time that the monitoring sites were influenced by the different transport pathways during the 9–10 years and demonstrate that each site was affected by air masses originating in the U.S.

Average SO_4^{2-} concentrations associated with each corridor (Figure 5.1.1(a)) indicate that higher concentrations were associated with transboundary flow from the U.S. (and, at eastern sites, with flow over southern Ontario/Quebec), and lower concentrations were associated with northerly flow from Canada toward the U.S. The particle NO_3^- results in Figure 5.1.1(b) show that when concentrations are higher, there was a stronger influence from the midwestern U.S. and western Canada (i.e. trajectory cluster 4 for Bratt’s Lake and ELA, and cluster 3 for Algoma). Because SO_2 and NO_x emissions changed significantly during the analysis period, the concentrations presented in the figures are likely higher, on average, than more recent concentrations. However, the relative differences in SO_4^{-2} and NO_3^- concentrations among the trajectory clusters are expected to be similar.

Jeong and colleagues (2011) also related the locations of air mass origin derived from back-trajectories to the concentration and composition of $PM_{2.5}$. Consistent with Figure 5.1.1, they showed that on days when the concentrations of the SO_4^{2-} and NO_3^- source apportionment factors derived for eastern Canadian monitoring sites were in the top 25%, there was transport from the U.S. into Canada. The researchers also observed that this transport pattern was associated with some of the other types of $PM_{2.5}$, such as particles found to be enriched in EC.

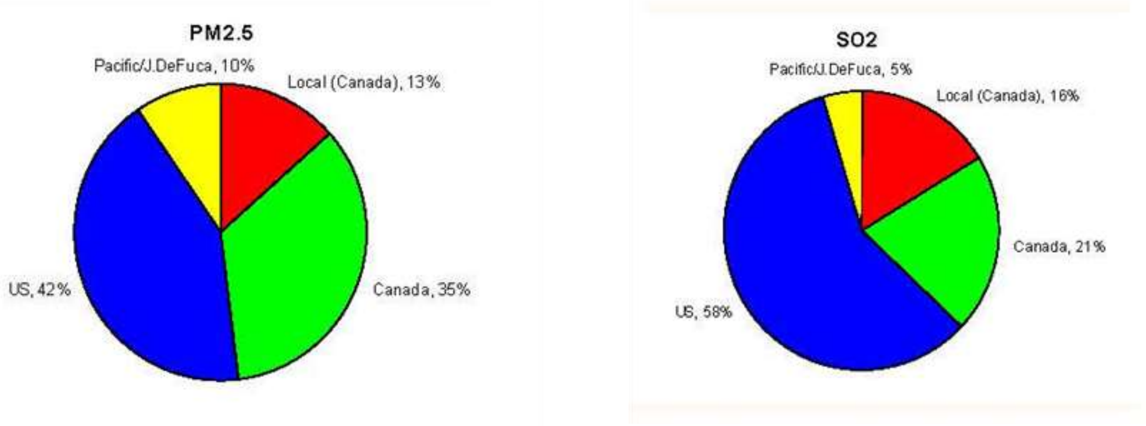
Local scale assessments of transboundary $PM_{2.5}$ transport were undertaken as part of the western and eastern pilots of the BAQS conducted from 2003 to 2006. The eastern pilot focused on southwestern Ontario and local scale transport (i.e. sources from within this region and U.S. sources near/adjacent to the border), with an interest in the influence of the meteorology associated with the Great Lakes (Brook et al., 2013). Due to this complex meteorology, it was not possible to explicitly determine what fraction of the $PM_{2.5}$ came from one country versus the other, and model scenarios were not undertaken for this purpose. However, impacts from sources in the Detroit area were observed when local sources in Ontario and the nearby states were relatively more important. In addition, during the high particle SO_4^{2-} periods, regional transport from the U.S. was implicated (McGuire et al., 2011).

Figure 5.1.1: Box-and-whisker plots of (a) particle SO_4^{2-} and (b) particle NO_3^- concentrations (µg/m^3) for the 9–10 year measurement periods identified in the headings



In southern British Columbia, where mountainous terrain and the coastal environment (i.e. Vancouver – Victoria and Puget Sound) complicate assessment of transboundary transport, three research sites were established in 2004. These were placed near the Canada – U.S. border in British Columbia—from the interior, bordering Idaho and Washington, to the coast of Vancouver Island. At each location, hourly pollutant measurements (including $PM_{2.5}$) were obtained, along with wind speed and direction, permitting an assessment of how concentrations varied by wind direction (i.e. surface winds from Canada or the U.S.). Analyses of these data showed that air pollutants, which are generally in relatively low concentrations compared with eastern North America, do move across the border at each site. The direction of transport is closely linked to weather patterns and the time of year (Meyn et al., 2007a, 2007b; Vingarzan et al., 2007). Overall, influence from each country appears to be relatively equal at these sites. For example, at the westernmost location, Christopher Point on the Strait of Juan de Fuca, elevated $PM_{2.5}$ concentrations were associated with stagnant conditions throughout the year. Hourly $PM_{2.5}$ and SO_2 observations, sorted by wind direction, indicated that emissions on both sides of the border contributed relatively equally (Figure 5.1.2). However, Canadian sources made a slightly larger percentage contribution of $PM_{2.5}$, while U.S. sources were a more important contributor to the observed SO_2. At all three points along the border, the prevailing direction of transboundary movement of air pollutants varied seasonally. The U.S. sources dominated the observed $PM_{2.5}$ during the spring and Canadian sources dominated during the winter, while generally contributions from both countries' emissions were relatively similar for $PM_{2.5}$.

Figure 5.1.2: Estimates from a wind sector analysis for percentage contributions of sources from Canada and the U.S. to pollutants measured at Christopher Point, B.C.; Canadian contributions shown as both local and transported sources

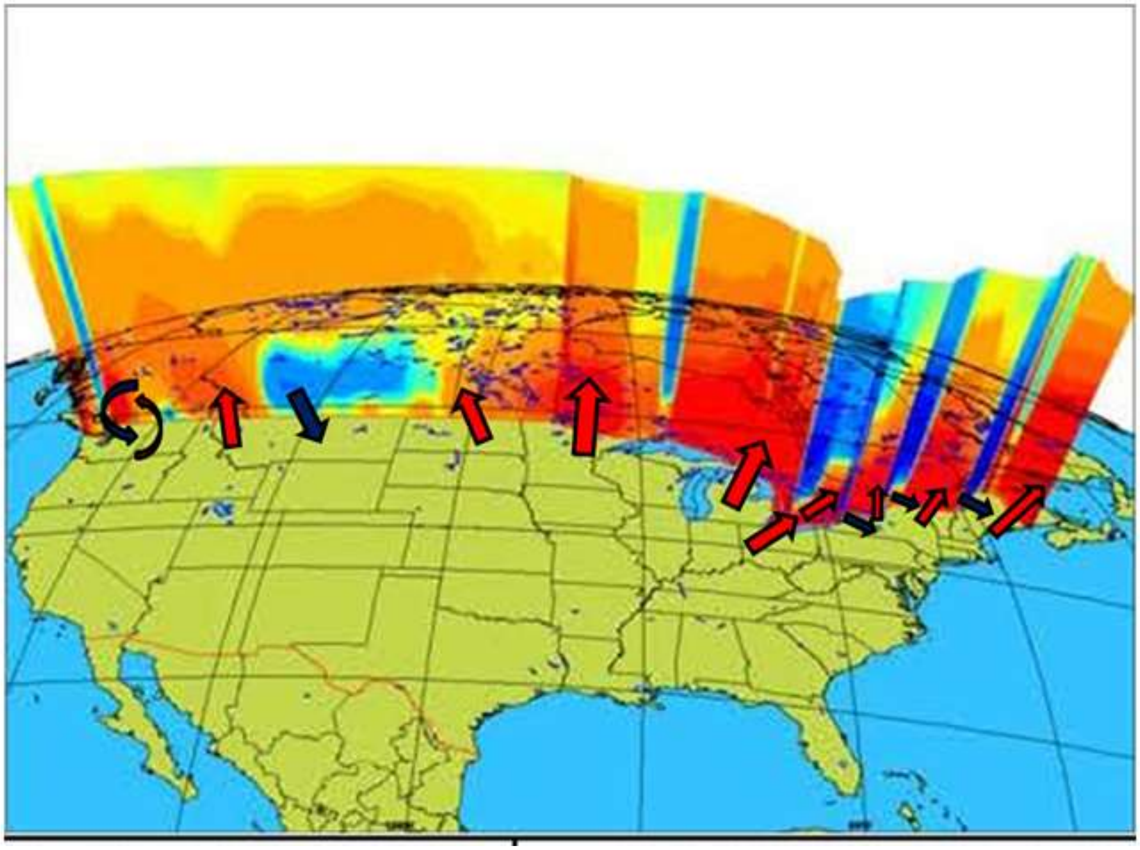


5.1.2 Applications of air quality models to assess transboundary transport

To obtain a more complete and quantitative picture of the nature of the flow across the border it is necessary to apply air quality models. For this assessment, existing output from Environment and Climate Change Canada's AURAMS model was used to estimate the extent of the impacts of emissions in Canada and the U.S. on $PM_{2.5}$ concentrations in each country. This was done by extracting the output at selected locations from a set of previously conducted annual scenarios. The base case, assumed to represent the current conditions, corresponded to the meteorology and emissions for 2006.

Clearly, a detailed discussion of AURAMS and other model uncertainties and their evaluation statistics is beyond the scope of this assessment. It should be stressed that while these available model scenarios do represent the most comprehensive estimates possible, the results should be viewed mainly as guidance helping to quantitatively understand transboundary transport, currently and in the future. However, it should be recognized that because emissions in both Canada and the U.S. have decreased significantly since 2006, these AURAMS results for the base case should be interpreted as simply adding more evidence of the presence and nature of transboundary transport along the Canada – U.S. border (Figure 5.1.3). They are not intended to indicate the current (2013) absolute levels of contributions to transboundary transport.

Figure 5.1.3: Three-dimensional schematic of transboundary flow of $PM_{2.5}$ across the Canada – U.S. border derived from an annual (2006) simulation using AURAMS



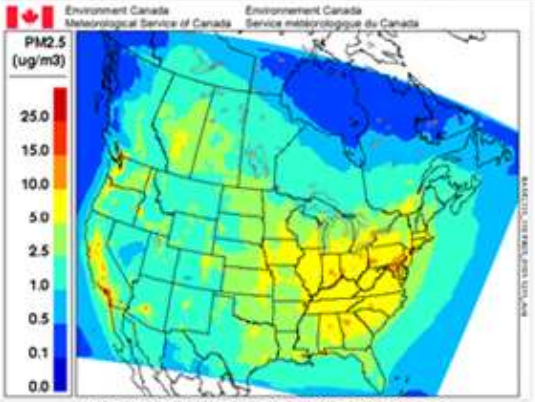
Note: Model estimates indicate that along the border the transboundary flow is complex, with differences between flow at the surface and aloft. For example, there are areas above the ground where there is significant Canada-to-U.S. transport (e.g. southern Ontario). Red colours indicate flow from the U.S. to Canada and blue from Canada to the U.S. Curved arrows highlight areas where pollutants cycle back and forth; overall contributions are relatively equal between countries. The vertical dimension has been greatly expanded to show the differences in flow with height.

Figure 5.1.3 was prepared by creating a "vertical wall" along the border and determining the overall annual average direction and magnitude of the flow (i.e. flux) of $PM_{2.5}$ through the wall under the 2006 base case conditions. This schematic of the variation in transport along the border and height above the ground highlights the overall complexity of the meteorology and its interaction with emissions and atmospheric chemistry. The model calculations suggest that in some regions there is transport in both directions that is similar in magnitude. This is the case in the Pacific Northwest (British Columbia), where flux is shown to be small. South and east of Alberta, where there are sizeable emissions and regular northerly winds, there is more flow from Canada to the U.S. From Manitoba eastward, the net transport of $PM_{2.5}$ mass at the surface travels from the U.S. to Canada. However, the estimates show (Figure 5.1.3) that there are regions aloft where this may be reversed. Furthermore, there are large Canadian emissions from southwestern Ontario to Montréal that flow across the Great Lakes, the St. Lawrence River, and along the eastern border into the U.S.; they do not, however, appear to predominate over the flow from the U.S. to Canada.

In addition to the base case 2006 model scenario used to produce Figure 5.1.3 and Figure 5.1.4(a), two additional 2006 scenarios were available. In these cases, the anthropogenic emissions in Canada or the U.S. were set to zero in the input emission files while keeping biogenic and natural emissions and other model configurations (e.g. meteorology) unchanged. The resulting hourly $PM_{2.5}$ fields for each scenario were used (with 2006 meteorology) to create the annual average contributions from each country to North American $PM_{2.5}$ (i.e. U.S. influence = Base Case – Zero Canadian Emissions; Canadian influence = Base Case – Zero U.S. emissions). These are shown in Figures 5.1.4(b) and 5.1.4(c), respectively, to provide guidance on each country's contribution in terms of possible magnitude and areas affected. The influence of emissions from the U.S. on Canadian $PM_{2.5}$ levels extends throughout all Canadian provinces, aside from the northernmost tip of British Columbia. The influence of Canadian emissions on $PM_{2.5}$ levels in the U.S. tends to be smaller; it extends to southern Oregon and Idaho, northeastern Colorado and across to southern Virginia. Note that Figure 5.1.3 differs from these figures in that it shows the net mass flow (i.e. flux through the "wall"), thereby indicating whether overall more $PM_{2.5}$ mass moves into Canada from the U.S. or vice versa.

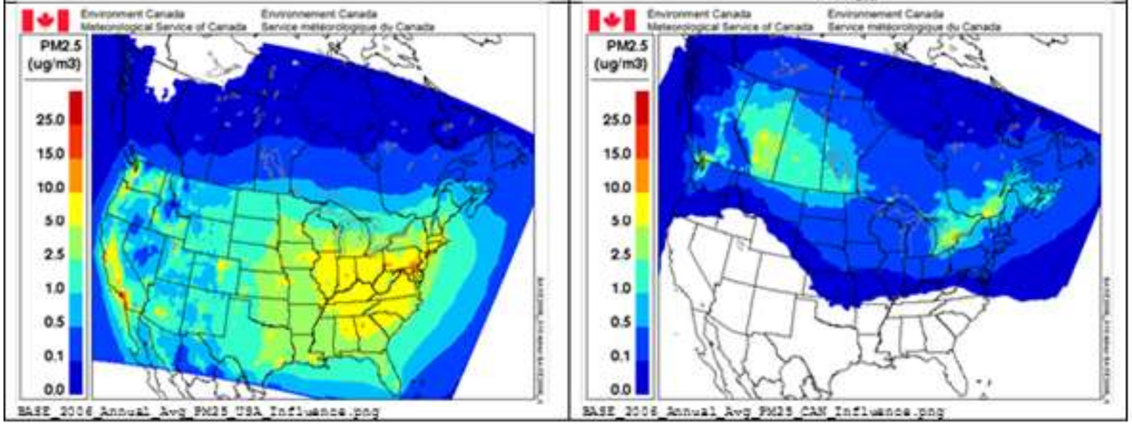
Figure 5.1.4: Predictions of ambient $PM_{2.5}$ concentrations in North America using the AURAMS model

(a) 2006 Annual Average $PM_{2.5}$



(b) 2006 USA Influence on Average $PM_{2.5}$

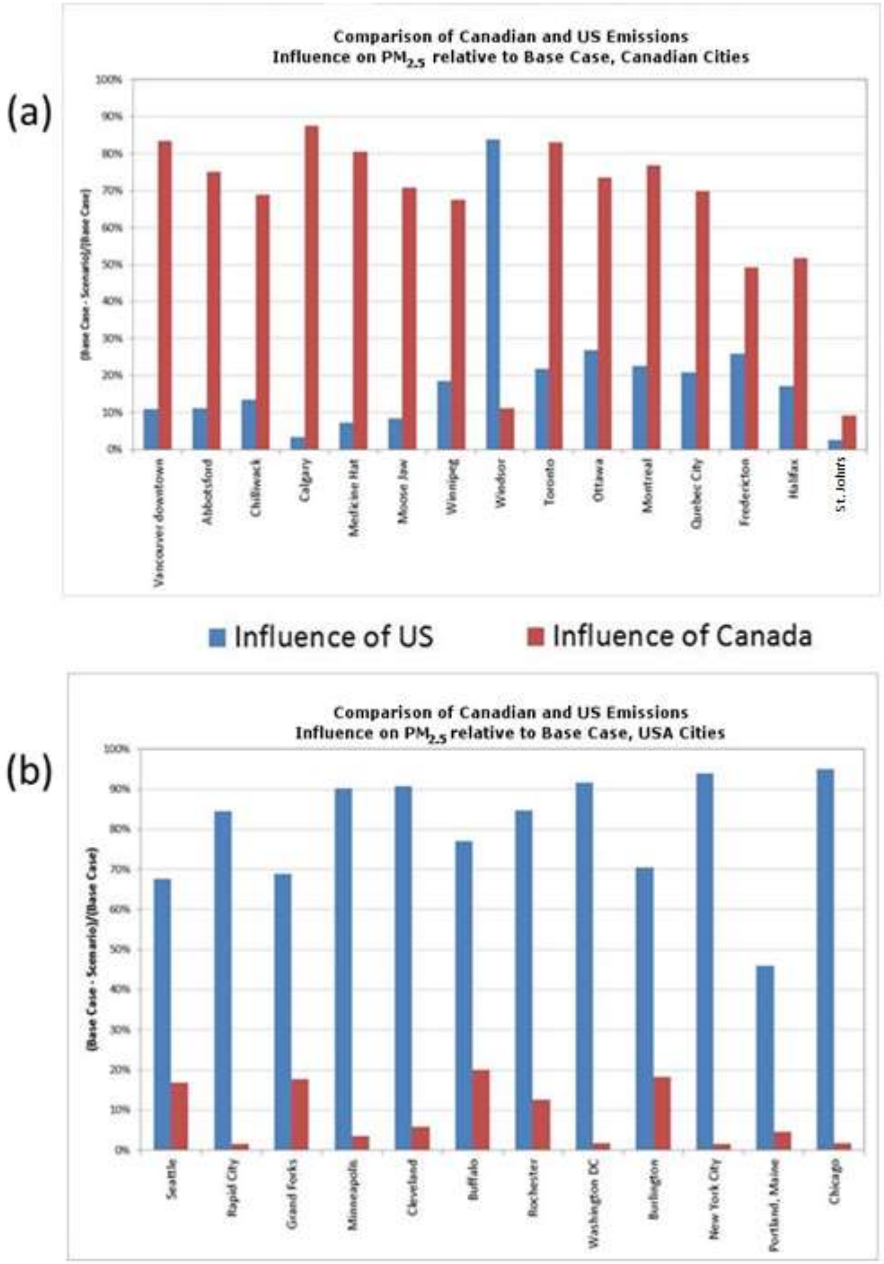
(c) 2006 Canada Influence on Average $PM_{2.5}$



Note: The base case model is presented in the top panel, and the influence of Canadian and U.S. anthropogenic emissions on North American $PM_{2.5}$ concentrations is presented in the lower panels (i.e. United States influence = base case – zero Canadian emissions; Canadian influence = base case – zero United States emissions).

* * * * *

Figure 5.1.5: Percentage contribution of emissions from Canada and the U.S. on annual average $PM_{2.5}$ levels in (a) Canadian cities and (b) U.S. cities (2006 base case)



For a range of cities located across southern Canada (Figure 5.1.5(a)), the two zeroed-out scenarios indicate that Canadian emissions are the dominant source of $PM_{2.5}$. A notable exception is Windsor, ON, where the U.S. contribution in 2006 is estimated to be 84% of the base case simulation. Toronto and Ottawa, ON, Montréal and Québec City, QC, and Fredericton, NB, all have U.S. contributions on the order of 20% of the base case. Winnipeg, MB, and Halifax, NS, also had simulated U.S. contributions of greater than 15%. For cities in the U.S. that are relatively close to the border, domestic contributions dominate. Nonetheless, Seattle, Wash., Grand Forks, N.D., and Buffalo and Burlington, N.Y., are all examples of cities where the $PM_{2.5}$ levels are influenced by Canadian emissions, showing values greater than 15% of the base case simulation (Figure 5.1.5(b)). Note that the contributions do not add to unity, due to the non-linear chemical interactions between species emitted in one country versus the other, the recirculation of pollutants, and the inclusion of natural emissions in both scenarios.

5.2 Impact of Future Emission Changes on $PM_{2.5}$ Transboundary Transport

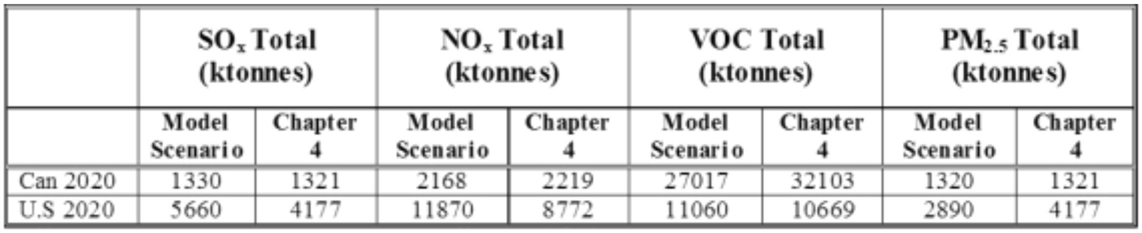
The impact of projected changes in emissions on $PM_{2.5}$ ambient concentrations and transboundary transport of $PM_{2.5}$ can only be evaluated with models. To inform this assessment, existing AURAMS runs were used to indicate how upcoming emission reduction policies may alter $PM_{2.5}$ concentrations and spatial patterns across both countries. Annual simulations at a 45-km grid resolution were carried out with 2006 meteorology and biogenic emissions, but with the 2006 emissions altered to represent future anthropogenic emissions in Canada and/or the U.S.

The 2020 emissions shown in Figure 4.4.1 were used for Canada, while for the U.S., emission projections were only available for 2016 for most sectors and for 2020 for the mobile sector at the time the AURAMS model was run. Therefore, the AURAMS future scenarios shown here, while referred to as 2020 below, actually correspond to a combination of 2016 and 2020 anthropogenic emission projections without accounting for potential future changes in meteorology and biogenic emissions. As Figure 4.4.2 shows that U.S. emissions of SO_2 and NO_x will decrease further from 2016 to 2020, the AURAMS-projected 2020 $PM_{2.5}$ levels over and downwind of the U.S. are expected to be overestimates.

The differences between the U.S. emissions considered in AURAMS and the current 2020 projections are provided in Table 5.2.1 to indicate the potential magnitude of the overestimates in U.S. influence projected for 2020. For SO_2 the AURAMS modelling scenario used 5600 ktonnes for 2020 emissions, while the most recently available U.S. projection (Table 4.4.2) is for the emissions to be 4177 ktonnes in 2020. Similarly, for NO_x the available model scenario used 11 870 ktonnes for 2020 emission from the U.S., while the recent projection is for 8772 ktonnes. For VOCs the difference is much smaller at 11 060 and 10 669 ktonnes, respectively. For $PM_{2.5}$ the AURAMS scenario used larger emissions than currently projected: 4177 ktonnes versus 2890 ktonnes.

These differences suggest that the model underestimated reductions in $PM_{2.5}$ concentrations between 2006 and 2020 (Figure 5.2.2), although due to non-linear relationships between precursor emissions and ambient concentrations, the actual magnitude cannot be determined. Similarly, due to the differences in the Table 5.2.1 estimates of the U.S. impact on Canadian $PM_{2.5}$, the 2020 levels shown below (e.g. Figure 5.2.1(b)) are likely to be overestimates, while the changes in the magnitude of the U.S. influence (Figure 5.2.4) are likely to be underestimates. Despite these uncertainties in modelled 2020 $PM_{2.5}$, the changes in $PM_{2.5}$ projected for 2020 do provide useful insight into future levels and the continued impact of transboundary movement on each country.

Table 5.2.1: Emissions included in the AURAMS model scenarios compared to the projected emissions shown in Tables 4.4.1 and 4.4.2



	SO_x Total (ktonnes)		NO_x Total (ktonnes)		VOC Total (ktonnes)		$PM_{2.5}$ Total (ktonnes)	
	Model Scenario	Chapter 4	Model Scenario	Chapter 4	Model Scenario	Chapter 4	Model Scenario	Chapter 4
Can 2020	1330	1321	2168	2219	27017	32103	1320	1321
U.S 2020	5660	4177	11870	8772	11060	10669	2890	4177

* * * * *

Figure 5.2.1: Model year 2020 AURAMS 45-km North American domain $PM_{2.5}$ predictions: (a) base model; (b) zero Canadian anthropogenic emissions; (c) zero U.S. anthropogenic emissions

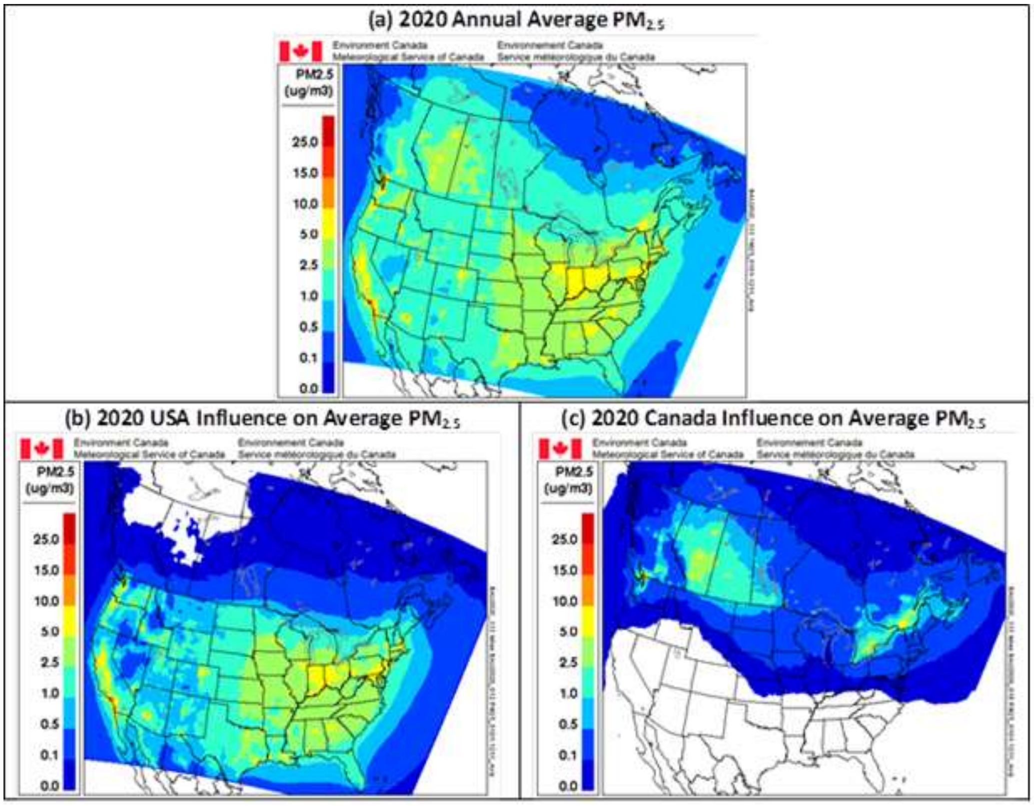


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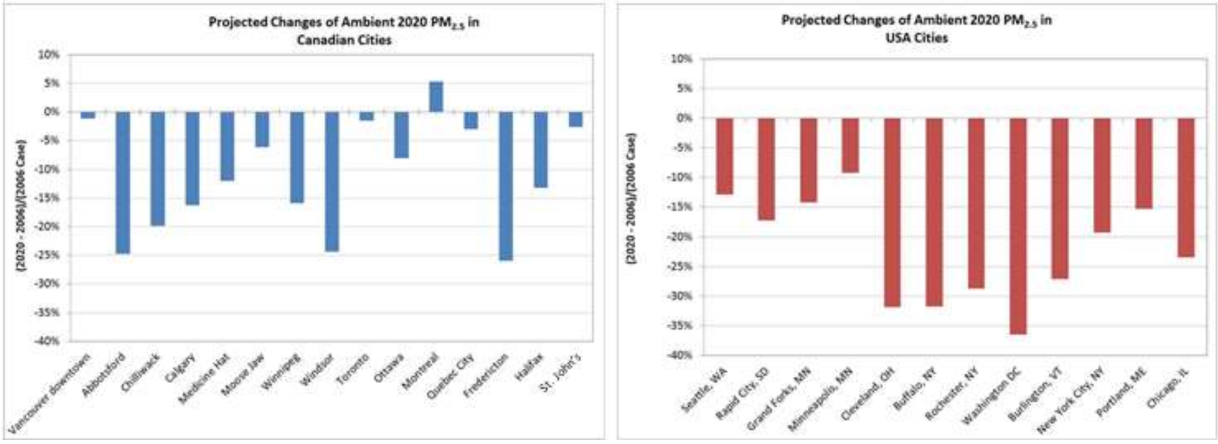
Figure 5.2.2 Projected percentage changes in annual $PM_{2.5}$ for the 2020 scenario relative to the 2006 base case levels, selected Canadian and U.S. cities



The top panel of Figure 5.2.1 shows the 2020 annual $PM_{2.5}$ concentrations calculated for the surface based on hourly model results. Compared with the 2006 base case scenario (Figure 5.1.4(a)), there are clear $PM_{2.5}$ reductions in the 2020 simulation. The projected reductions are larger in the eastern U.S. and coastal California, where annual $PM_{2.5}$ is reduced by about 5 $\mu g/m^3$. Smaller reductions, about 2 $\mu g/m^3$, are predicted for areas in Vancouver, the Calgary – Edmonton corridor, and southern Manitoba and Ontario. Despite overall decreases, a model difference plot (not shown) indicates that there are regions with small increases in $PM_{2.5}$ concentrations between 2006 and 2020. These increases are in areas of high industrial activity such as Fort McMurray, AB, Merritt, B.C., west-central Wyoming and southern Nevada.

The influence of each country on future transboundary transport of $PM_{2.5}$ was evaluated by removing anthropogenic 2020 emissions in Canada or the U.S from the input emission files of the model, while keeping biogenic emissions and other model configurations unchanged, as was done for the 2006 base case (Figure 5.1.4).

Figure 5.2.1 shows the projected 2020 $PM_{2.5}$ influences from the U.S. (panel b) and Canada (panel c), based on concentration differences from the overall projected 2020 ambient $PM_{2.5}$ concentrations (panel a). In general, the impacts on transboundary $PM_{2.5}$ transport from both the U.S. and Canada are similar to the 2006 modelling results given in Figure 5.1.4.

Future changes in $PM_{2.5}$ concentrations between 2006 and 2020 were estimated for cities near the Canada–U.S. border (Figure 5.2.2) by extracting the data for their corresponding grid squares from the available AURAMS runs. The predicted percentage reductions are generally larger for cities in the U.S. than for those in Canada. In Canada, the predicted annual $PM_{2.5}$ reductions are largest in cities downwind of major urban centres, such as Abbotsford, B.C., Windsor, ON, and Fredericton, NB.

Major urban centres with high base case $PM_{2.5}$ concentrations, such as Vancouver and Toronto, show smaller reductions (<2%) from the 2006 levels. Montréal is the only city shown with a predicted increase in $PM_{2.5}$. Analyzing regional emissions in and around Montréal indicates that this increase arises from higher emissions in 2020 from non-industrial sources, such as residential wood combustion, during winter months.

The future influence of anthropogenic emissions from Canada and the U.S. on $PM_{2.5}$ concentrations in the selected set of cities was also investigated using the AURAMS simulations for 2006 and 2020. $PM_{2.5}$ influence as result of U.S. and Canadian emissions was calculated similarly to the 2006 base case. Thus Figure 5.2.3 is the same as Figure 5.1.5, but for emissions projected to 2020.

Figure 5.2.3: Percentage contribution of emissions from Canada and the U.S. on projected 2020 annual average $PM_{2.5}$ levels in selected Canadian and U.S. cities

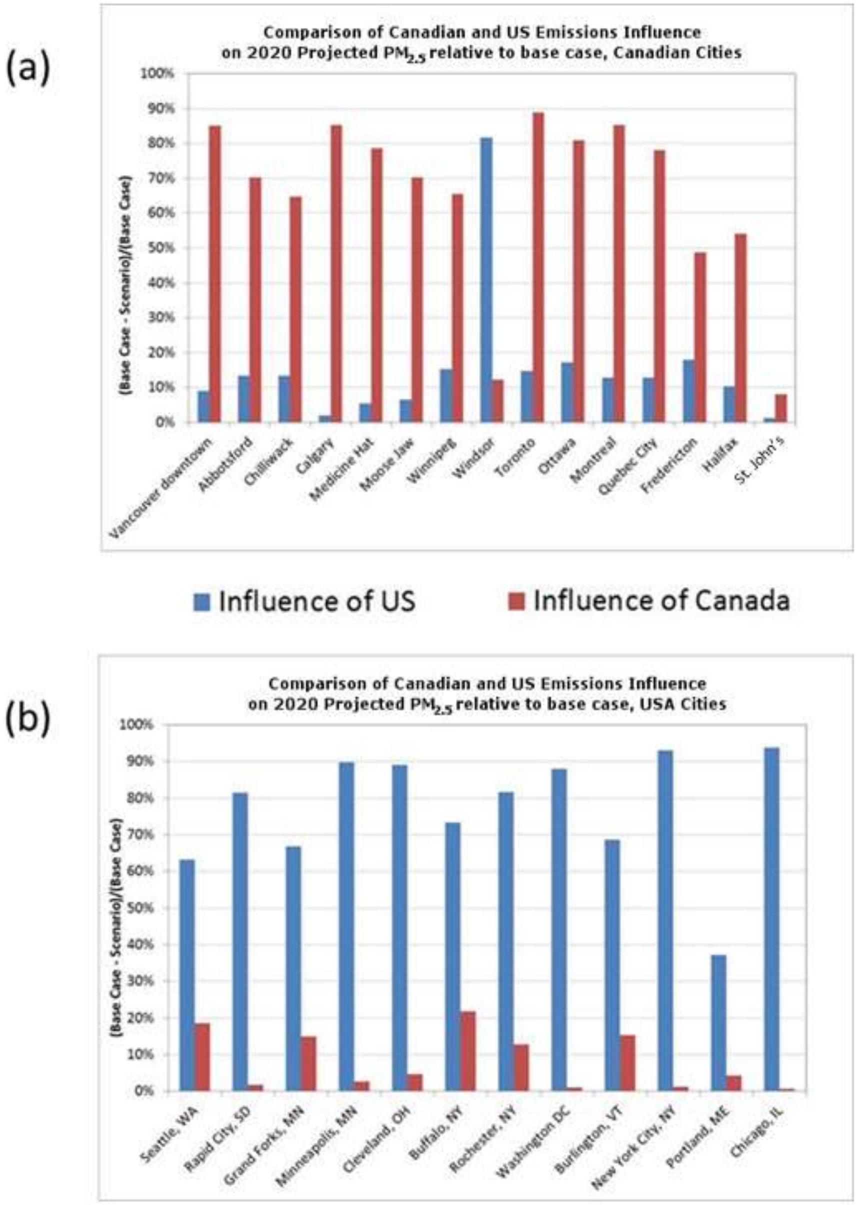
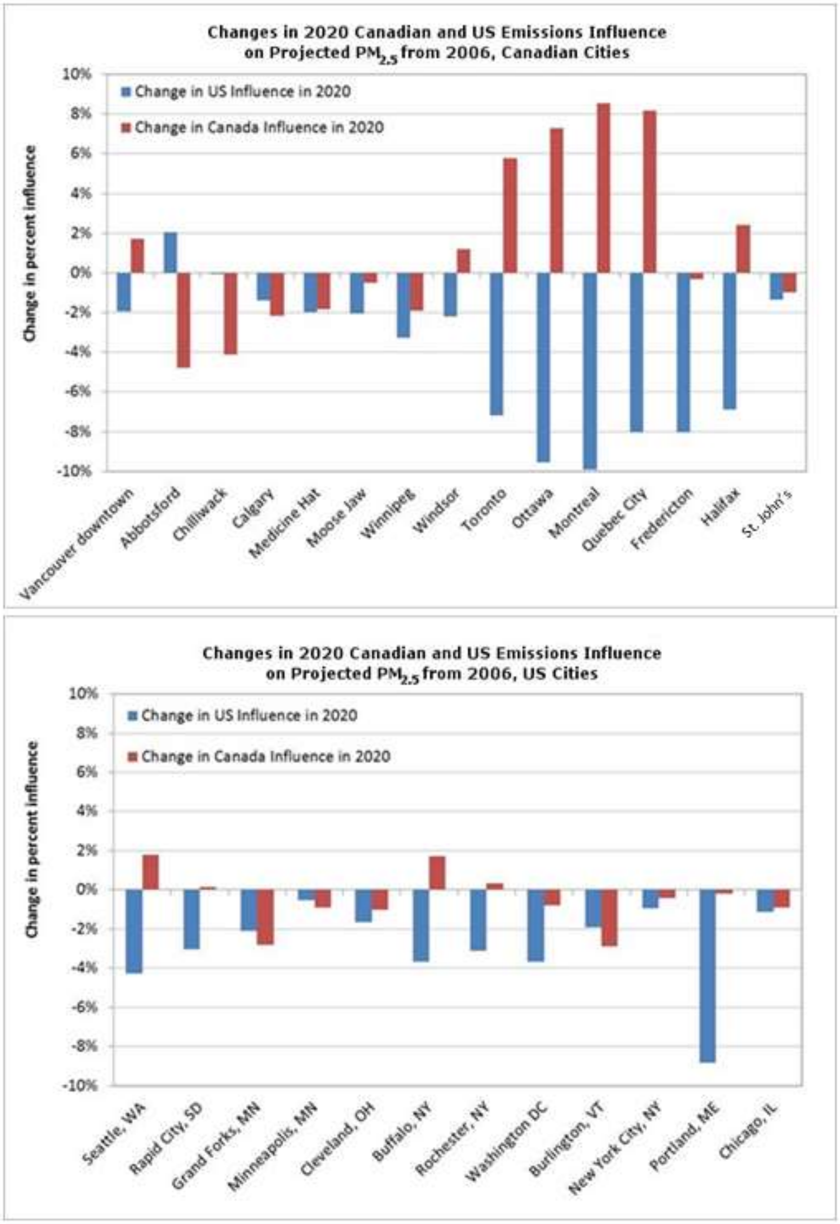


Figure 5.2.4 represents the difference between the 2006 and 2020 percentage contributions and shows that the U.S. influence on $PM_{2.5}$ in the selected Canadian cities is expected to decrease by up to 10% in eastern Ontario and southwest Quebec. The only exception shown is in the Fraser Valley of British Columbia (Abbotsford), where there is predicted to be a small increase in the U.S. impact on the $PM_{2.5}$ levels. However, as noted above, U.S. 2016–2020 emission reductions from sectors other than mobile were not available for the AURAMS runs generating these results. Thus, the percentage decreases in U.S. influence in Figure 5.2.4 are likely to be somewhat larger, and the U.S. impact on Fraser Valley levels may also actually decrease when the 2016–2020 emission reductions are considered. The need to update these runs with complete 2020 emission scenarios is discussed in Chapter 7.

Figure 5.2.4: Relative change in 2020 emission influences from Canada and the U.S. on $PM_{2.5}$ concentrations from 2006 in selected Canadian and U.S. cities



From 2006 to 2020, the influence of Canadian emissions on U.S. $PM_{2.5}$ (for selected cities) generally decreases by a small amount. However, also near the Fraser Valley, in Seattle, Wash., the influence of Canadian emissions rises slightly. The increases in the other country's impact on both Seattle and Abbotsford further highlight the complexity of the transboundary flow in the Pacific Northwest, as implied in Figure 5.1.3. Canadian emission influence in the Buffalo, N.Y., area is also predicted to increase slightly between 2006 and 2020. However, it is important to note that, for most cities in both countries, the dominant source of $PM_{2.5}$ in 2020 will continue to be emissions from within the respective counties, and transboundary influence should generally be less in 2020 than in 2006.

In terms of Canadian standards, future predictions of ambient concentrations in the form of the annual average and 24-h metrics are currently only available across Canada for a 2016 scenario (i.e. because U.S. 2020 emissions were not available for all sectors). At this time, the projected Canadian and U.S. emissions changes are expected to result in rural/regional $PM_{2.5}$ concentrations over areas near the southern Ontario and southern Quebec borders of 6 µg/m^3 and 19 µg/m^3, for the annual and 24-h CAAQS metrics, respectively. These modelled regional/rural $PM_{2.5}$ concentrations are affected by emission sources in both Canada and the U.S., although the relative contributions are uncertain. These levels are below the 2015 annual and 24-h CAAQS, but relatively close to the 2020 standards.

Thus while some populated areas near the border have the potential to exceed the 2015 CAAQS in 2016 due to local emissions, a larger number of such areas may exceed the 2020 CAAQS. However, ongoing emission reductions expected between 2016 and 2020, which have not yet been projected to 2020 $PM_{2.5}$ levels (as noted above), are likely to help limit the number of Canadian locations exceeding the CAAQS at that time. Over the border regions of western Canada and Atlantic Canada, predictions of $PM_{2.5}$ concentrations in 2016 suggest that levels will already be below the CAAQS for 2020.

The U.S. modelling analyses done in support of the recent review of the PM NAAQS, specifically for the Regulatory Impact Analysis, also provide insight into future transboundary transport of $PM_{2.5}$. Figure 5.2.5 shows the projected difference in $PM_{2.5}$ concentrations between a 2007 base case and a future scenario for 2020 (consistent with the emission projections described in Section 4.4). Canadian emissions were held constant, so the results in Figure 5.2.5 show the impact of projected U.S. emission changes in both Canada and the U.S., but do not reflect the impact of any projected alterations in Canadian emissions.

Thus, the changes shown are biased low over areas where Canadian emissions have an impact (see Figure 5.2.1(c) for an indication of this area). As can be seen in this figure, the most significant reductions in $PM_{2.5}$ are expected to occur in the eastern U.S. and in parts of California. Relating this analysis to U.S.– Canada transboundary transport of $PM_{2.5}$, the most significant $PM_{2.5}$ reductions are expected to occur in the Northeast and upper Midwest in the U.S. and in the southeastern part of Canada. The expected impact of U.S. emission reductions in Canada ranges from a reduction in $PM_{2.5}$ concentrations of about 0.5 µg/m^3 to a reduction of about 3 µg/m^3.

Figure 5.2.5: Modelled differences in annual $PM_{2.5}$ concentrations between 2007 and 2020

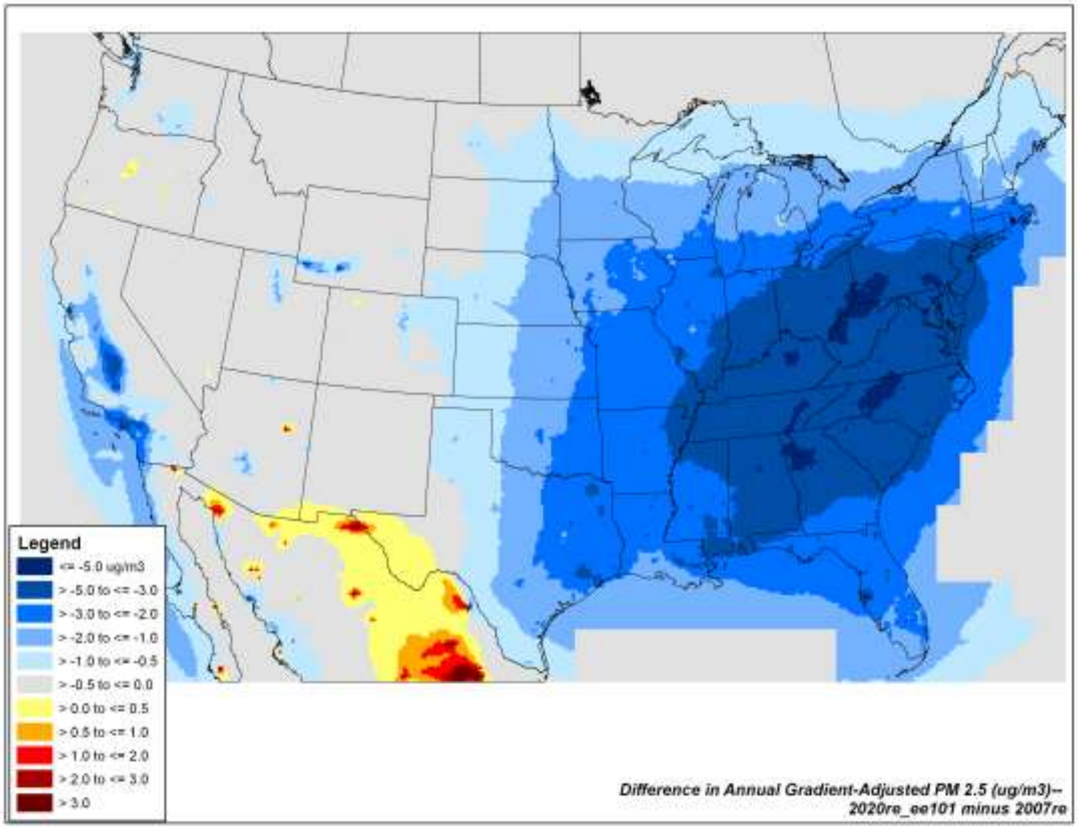


Figure 5.2.6 shows the results of projected U.S. emission changes that are expected from current federal air pollution control programs in terms of attainment with the recently revised annual PM NAAQS. Only seven counties, all in California, are predicted not to meet the current annual primary $PM_{2.5}$ NAAQS of 12 µg/m^3 in 2020.

Figure 5.2.6: EPA projections showing that 99% of U.S. counties with monitors would meet the annual fine particle health standard of 12 µg/m^3 in 2020

CHAPTER 6: EMERGING SCIENCE ISSUES

Chapter Summary

Emerging science issues related to the transboundary transport of $PM_{2.5}$ and that need to be addressed to inform air quality management actions include:

- the impacts of $PM_{2.5}$ and its components in the context of the overall air pollutant mixture
- the effects of climate change on ambient $PM_{2.5}$ concentrations and the impact of climate change mitigation activities on air quality
- the impact of a rapidly changing energy landscape on ambient $PM_{2.5}$
- the growing relative importance of the transport of natural and global $PM_{2.5}$ sources

6.1 Multi-pollutant Exposures, Effects, and Management

Air pollution exposure and health studies often investigate the potential effects of individual pollutants to support air quality management efforts, which are largely focused on one pollutant at a time. However, it is widely recognized that in real-world settings, humans and ecosystems are exposed to air pollution mixtures, and many emission reduction measures will influence the emissions of multiple pollutants (Hidy et al., 2011a). PM itself is a mixture of pollutants derived from various precursor pollutants associated with different sources; thus, reducing ambient levels of PM requires emission reductions of multiple pollutants from numerous sources. As a consequence, air quality management activities targeted at PM are by their very nature multi-pollutant strategies. Still, these strategies could potentially be optimized and strengthened with a greater understanding of the health and ecological impacts from exposure to $PM_{2.5}$ in the broader context of pollutant mixtures.

At present it is not clear, however, whether the effects of $PM_{2.5}$ alone are greater than, equal to, or less than the impact of simultaneous or sequential exposures to $PM_{2.5}$ (or one of its components) in combination with co-pollutants such as O_3, VOCs, or NO_x. Furthermore, since $PM_{2.5}$ itself is a mixture of pollutants, it is also important to better understand the relative exposure to and toxicity of PM components. Since none of these pollutants exist in isolation, these problems are only important insofar as they lead to the identification of better means to protect public health. There are many ways to limit air pollution, but estimating their benefits currently relies on relatively simple concentration–response functions; much more could be done if the roles of the gases and the variable constituents of PM were better understood (Lippmann et al., 2013). Ongoing work in these areas, as well as information on the impacts of PM types and mixtures based upon source type (e.g. traffic) will be increasingly important to consider in future management of air quality. Due to the lack of a no-effects threshold, further reductions in $PM_{2.5}$ levels can be expected to produce additional health benefits, but more precisely targeted measures may yield more effective results. Different approaches have been proposed to address these issues, including focusing on pollutant sources (Thurston et al., 2011), using adjoint models to link health impacts to emissions from real or hypothetical sources (Pappin and Hakami, 2013), studying urban exposure environments (Hubbell, 2012), or risk-based air quality management (Wesson et al., 2010).

6.2 Air Quality and Climate Change

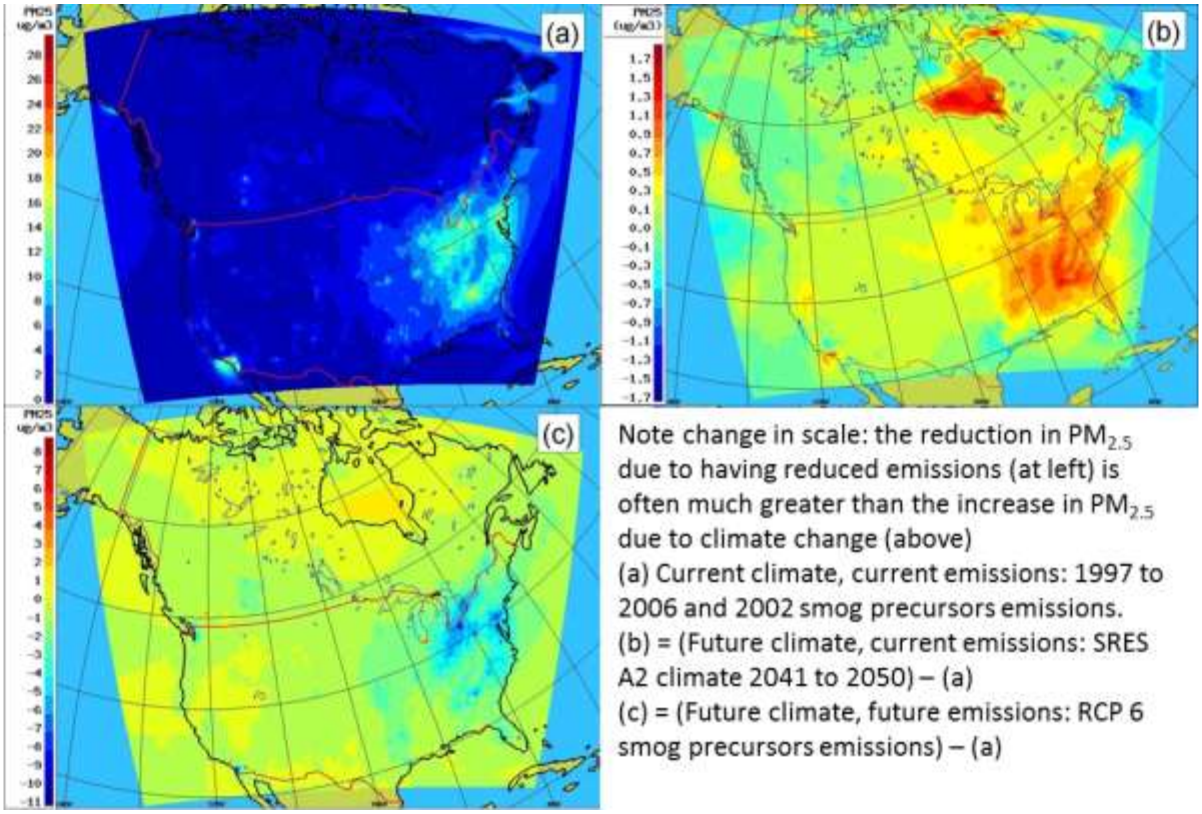
6.2.1 Impacts of climate change on $PM_{2.5}$

Air quality and climate change are linked. Weather patterns leading to smog events may change in frequency, and warmer temperatures are expected to increase O_3 concentrations and lengthen its season in some areas (EPA, 2009a). However, the overall impact of climate change on air quality (including $PM_{2.5}$) in North America is much less certain given the wide range of factors that could change in an unpredictable and interrelated manner. A warmer, drier climate is expected to increase wildfire intensity and the duration of wildfire season, leading to an increase in PM (Yue et al., 2013) and potentially regional O_3. On the other hand, warmer winters may change human behaviour (for example, less residential wood burning) in ways that would reduce PM emissions in some localities. Increased demand for cooling in the summer may also change emission patterns, potentially increasing $PM_{2.5}$ levels. Modifications to the biogenic precursor emissions for both O_3 and $PM_{2.5}$ are also likely with changes in temperature, precipitation, species distribution and growing season. Moreover, the impact of climate change on cloud properties, precipitation, humidity, and weather patterns will also affect $PM_{2.5}$; however, there is uncertainty in all of these factors and in their impact on $PM_{2.5}$, which will probably vary by location.

To obtain information on how changes in climate may affect air quality, Kelly and colleagues (2012) conducted a modelling experiment using a modified version of AURAMS driven by Regional Climate Model outputs for current and future climates. From that work, the modelled effect of climate change on future $PM_{2.5}$ concentrations is presented in Figure 6.1.

Under the Current Climate Current Emissions scenario, minor increases of $PM_{2.5}$ ($>0.2\ \mu g/m^3$) were predicted over much of North America (Figure 6.1(a)). Larger increases ($> 1.0\ \mu g/m^3$) expected to occur over Hudson Bay (Figure 6.1 (b)) would be caused by higher sea-salt aerosols released as a result of reduced ice cover, combined with increasing regional winds. Interestingly, the scenario predicted that anticipated future reductions in air pollution emissions would result in greater reductions in $PM_{2.5}$ over much of the eastern U.S. and the Ontario–Quebec corridor (Figure 6.1(c)) than the estimated increases related to climate change. However, as indicated above, understanding of how all the physical and sociological factors will change and subsequently impact air quality is relatively poor. Coupled climate, emission and air quality models currently have limited capability to predict these changes.

Figure 6.1: Impacts of future climate and emissions on $PM_{2.5}$ levels in North America



Note change in scale: the reduction in $PM_{2.5}$ due to having reduced emissions (at left) is often much greater than the increase in $PM_{2.5}$ due to climate change (above)

(a) Current climate, current emissions: 1997 to 2006 and 2002 smog precursors emissions.

(b) = (Future climate, current emissions: SRES A2 climate 2041 to 2050) – (a)

(c) = (Future climate, future emissions: RCP 6 smog precursors emissions) – (a)

6.2.2 Air quality impacts of climate change mitigation

Just as climate change will affect air quality, air pollutants can also impact climate change. While O_3 is known to have a warming impact, the effect of fine PM on climate change is much more complex. Some PM components, such as SO_4^{-2}, have a cooling effect in the atmosphere as they scatter and reflect sunlight away from the earth (Goldstein et al., 2009). Other PM components, such as black carbon (BC), have a warming effect as they absorb the sun's energy and heat the atmosphere (EPA, 2012) and contribute to positive feedback by accelerating ice/snow pack melting through decreased albedo.

As a result, it is important to understand these interactions when considering both air quality management options and reductions of greenhouse gas emissions. BC, as a short-lived climate-forcing pollutant, has been proposed as an important target for immediate action, in order to produce short-term benefits in slowing global warming. However, much remains unknown about BC as an air pollutant and as a climate forcer, and efforts must be made to better understand the air quality, human and ecosystem health benefits of reducing this $PM_{2.5}$ constituent when considering climate mitigation options (Bond et al., 2013).

6.3 The Changing Energy Landscape

The energy landscape in both Canada and the U.S. is changing rapidly. This includes development of shale oil and gas deposits in both countries and the oil sands in Canada. Factors driving the change in both the production and use of energy include the goal of energy independence, responses to climate change, and economic growth. While the shifting energy landscape includes the development and application of more renewable energy sources, the growth and expansion of more traditional sources of energy may have the biggest impact on air quality and, in particular, $PM_{2.5}$. For example, the recent increase in oil and gas production from previously untapped reserves in both the Canada and the U.S. and the corresponding decline in reliance on coal will impact both the composition and distribution of ambient $PM_{2.5}$. Its composition will change because the emissions, including direct $PM_{2.5}$ and $PM_{2.5}$ precursors, from the production and use of natural gas differ from those in the production and use of coal. The distribution of ambient $PM_{2.5}$ will be altered because the source regions for energy are changing, as well as the number and distribution of mining and production facilities.

The overall impact on $PM_{2.5}$ from these adjustments is uncertain at this time, but potential alterations in the composition and geographic distribution ambient $PM_{2.5}$ should be monitored closely in the future. As an example, the rapid growth in oil extraction from the Bakken formation, which spans both countries (in North Dakota, Montana, Alberta and Saskatchewan), is leading to greater emissions and more transboundary transport in both directions.

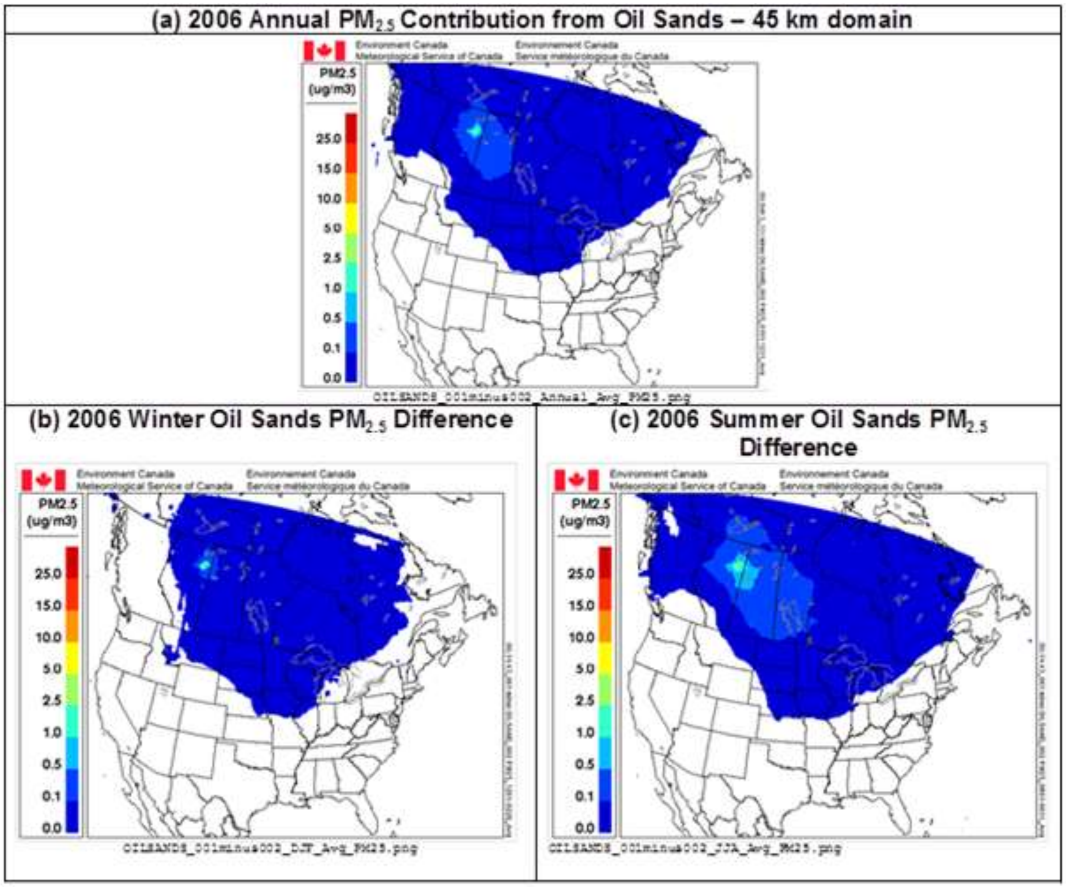
6.3.1 Case simulation – oil sands emissions

Given recent and expected future growth in emissions in western Canada arising from the development and production of synthetic crude in the oil sands, their impact on regional air quality was assessed through an additional 2006 model scenario. This involved removing oil sands-related emissions from the model input and determining the difference between the base case (Figure 5.2.2(a)) and this scenario for annual average, winter and summer $PM_{2.5}$.

The results, shown in Figure 6.2, suggest that the impact of the oil sands on average $PM_{2.5}$ transport across the southern border of the Prairie provinces is currently relatively small, with contributions to U.S. concentrations of $PM_{2.5}$ of less than 0.1 $\mu g/m^3$ on average.

This scenario may be compared with Figure 5.2.2(c), where the Canadian-influenced $PM_{2.5}$ in the same region is a maximum of 1 $\mu g/m^3$ and the U.S.-influenced $PM_{2.5}$ (Fig. 5.2.2(b)) is greater than 1 $\mu g/m^3$. The oil sands contribution is thus estimated at about 1/10th of the overall $PM_{2.5}$ concentration along the international border, based upon 2006 emissions. Continued growth in the oil sands since 2006 increases their $PM_{2.5}$ contribution both locally and in the U.S. As emissions decline elsewhere, the relative importance of oil sands emissions can be expected to increase further.

Figure 6.2: Modelled impacts of oil sands emissions on $PM_{2.5}$ concentrations, as predicted by the AURAMS 45-km North American domain model



(a) 2006 Annual $PM_{2.5}$ Contribution from Oil Sands – 45 km domain

(b) 2006 Winter Oil Sands $PM_{2.5}$ Difference

(c) 2006 Summer Oil Sands $PM_{2.5}$ Difference

Note: (a) annual average impact (base case—zero oil sands emissions); (b) average impact in winter; (c) average impact in summer

6.4 New Sources of Transboundary PM

Given the trends of emission reductions in both Canada and the U.S., anthropogenic contributions to the transboundary transport of $PM_{2.5}$ (and to the exceedance of current air quality standards) may become less significant, which leads to the emerging importance of other sources that have not been previously included in PM assessments. Natural sources and intercontinental transport of air pollutants are potentially two major contributors. Transboundary transport of natural PM components has been reported, such as dust aerosols from the U.S. (Park et al., 2007) and forest fire components from Canada (Lavoué et al., 2007). The Hemispheric Transport of Air Pollutants assessment (TF HTAP, 2010) has shown the potential contributions of intercontinental transport of O_3 and PM to ambient levels in Canada and the U.S. Increasing hemispheric background concentrations and/or an increasing frequency of significant episodes of intercontinental transport might affect the ability of Canada and the U.S. to meet more stringent standards in the future.

CHAPTER 7: CONCLUSIONS

7.1 Updating the Findings from the 2004 Assessment

The 2004 Assessment provided the scientific foundation to support the future development of joint strategies under a PM annex. It concluded that there was a significant relationship between the emissions of PM and PM precursors and elevated PM levels in both Canada and the U.S., and that the transboundary transport of PM and PM precursors can be significant enough in some regions to potentially compromise the attainment of national standards. Although a PM annex has yet to be adopted under the Canada – U.S. Air Quality Agreement, as discussed above, there have been significant reductions in emissions of PM and its precursors. Therefore, it makes sense to update major findings from the 2004 Assessment in order to inform a decision on whether to pursue a PM annex at this time. The major findings from the 2004 Assessment are listed below, along with an update of the finding based upon information presented in this document.

2004 Assessment Key Finding No.1: *Transboundary transport of PM can contribute to above-average PM levels in both Canada and the U.S.*

- Ambient levels of $PM_{2.5}$ in the border regions exceed the standards set for $PM_{2.5}$ in several regions of both Canada and the U.S.
- $PM_{2.5}$ is transported across the border region between Canada and the U.S., leading to elevated concentrations of $PM_{2.5}$ in both countries.

2014 Update: Transboundary transport continues to contribute to above-natural background $PM_{2.5}$ levels in both Canada and the U.S., but absolute concentrations have decreased since the 2004 Assessment.

- Levels in most regions along the border are in attainment with current standards in the U.S. The primary annual standard in the U.S. has been strengthened to 12 $\mu g/m^3$, while the primary 24-h standard and secondary annual standard have not changed since the 2004 Assessment.
- In terms of Canadian standards, future predictions of ambient concentrations in the form of the annual average and 24-h CAAQS metrics indicate that most areas near the southern Ontario and southern Quebec border with the U.S. will be below the 10 $\mu g/m^3$ 2015 CAAQS annual standard. Possible exceptions include areas with relatively large local emissions that add to the rural/regional levels in areas close to the border, where annual average levels are predicted to be around 6 $\mu g/m^3$. Ongoing emission reductions from 2016 to 2020 are likely to help limit the number of Canadian locations exceeding the 2020 CAAQS. Over the border regions of western Canada and Atlantic Canada, projections of $PM_{2.5}$ suggest that levels will be below the CAAQS for both 2015 and 2020.

2004 Assessment Key Finding No. 2: *PM levels vary significantly over geographic regions.*

- Elevated concentrations of $PM_{2.5}$ are found more often in the following regions: northeastern U.S., the industrial Midwest, southwestern Ontario and northwestern U.S.

2014 Update: PM levels continue to vary over geographic regions.

- Concentrations of $PM_{2.5}$ have been reduced significantly in all regions along the border. The border area with the highest concentration in the U.S. is the industrial Midwest, but the levels are still in attainment for the current PM NAAQS standard. In eastern Canada, $PM_{2.5}$ levels tend to decrease from southwestern Ontario towards the Atlantic coast, with higher concentrations in southwestern Ontario and in large cities (Hamilton, Toronto and Montréal). In western Canada, the highest concentrations are seen in mountainous regions where terrain plays a role in trapping local emissions. Higher concentrations also occur in winter during calm, stable conditions, particularly near industrial development (oil sands) and in large cities in the Prairies (e.g. Edmonton). $PM_{2.5}$ mass has decreased significantly in eastern Canada from 2003 to 2011, while western sites have generally remained stable.

2004 Assessment Key Finding No. 3: *There are many sources of PM and PM precursors:*

- Local mobile and industrial sources make a constant contribution to PM levels in Toronto and dominate on relatively clean days with air flow coming from the north.
- Coal-related sources in the U.S. make a substantial contribution to PM in Toronto and elsewhere in Canada.
- High agricultural NH_3 emissions influence ammonium nitrate (NH_4NO_3) levels.
- Sulphur dioxide (SO_2) and nitrogen oxides (NO_x) are primarily regional contributors to PM, while organic or black carbon and other PM constituents tend to originate from local sources.

2014 Update: There remain many sources of $PM_{2.5}$ and $PM_{2.5}$ precursors, and they all continue to contribute to the observed $PM_{2.5}$. The following trends are noted:

- Reductions in SO_2 and NO_x emissions suggest that the relative contributions of coal-related sources and motor vehicles have decreased. Ambient concentrations of SO_4^{-2} related to coal combustion have declined in eastern North America.
- Wildfire emissions are a growing source of regional carbonaceous PM.
- Oil and gas production is an emerging source of new emissions in geographic areas that had lower emissions in 2004.

2004 Assessment Key Finding No. 4: *Air quality modelling of emission reduction scenarios for PM and PM precursors found the following:*

- Air quality modelling of emission reduction scenarios for PM and PM precursors found that
    - projected reductions vary spatially and by season
    - pdditional reductions of SO_2 and NO_x emissions should produce concomitant effects on ambient $PM_{2.5}$ levels as well as concurrent reductions in particle ammonium (NH_4^+).

2014 Update: Analyses of existing emissions and ambient concentrations confirm the emission projections and modelling analyses reported in the 2004 Assessment. In addition, updated emission projections show similar trends in both the U.S. and Canada, and analyses of existing air quality modelling predict further reductions in ambient $PM_{2.5}$ concentrations.

- National emission inventories indicate that total anthropogenic $PM_{2.5}$ and NH_3 emissions have remained fairly stable in Canada and the U.S. from 2002 to 2010, while emissions of SO_2, NO_x, and VOCs all have declined over this time period.
- Emissions of SO_2, NO_x and VOCs are projected to decrease through about 2020 in both countries, while emissions of $PM_{2.5}$ and NH_3 are expected to change much less during this period. Beyond 2020, decreases in all emissions are expected to be smaller.
- Projections of ambient concentrations in 2016 and 2020 show continued reductions in $PM_{2.5}$ in both the U.S. and Canada. In the U.S., no border areas are expected to exceed the current primary annual NAAQS for $PM_{2.5}$ of 12 $\mu g/m^3$ in 2020. The projected reductions are larger in eastern U.S. and coastal California, where annual $PM_{2.5}$ will be reduced by about 5 $\mu g/m^3$. Smaller reductions, around 2 $\mu g/m^3$, are predicted for areas in Vancouver, the Calgary–Edmonton corridor, and southern Manitoba and Ontario.
- In Canada, the predicted 2006–2020 decreases in $PM_{2.5}$ are expected to result in rural/regional background $PM_{2.5}$ concentrations near the southern Ontario and southern Quebec borders that

will be below the 2015 and 2020 annual and 24-h CAAQS. However, these levels are above natural background concentrations and close enough to the CAAQS that some populated areas with relatively large local emissions may experience $PM_{2.5}$ above the CAAQS. In the border regions of western and Atlantic Canada, 2015 and 2020 CAAQS are not projected to be exceeded.

- Additional U.S. emission reductions from 2015 to 2020 not included in the available model runs are expected to lead to lower 2020 $PM_{2.5}$ in the vicinity of the Canada – U.S. border than currently projected; however, further reductions in transboundary and domestic PM2.5 will lead to additional public health benefits.

2004 Assessment Key Finding No. 5: *There are linkages between PM and other air quality issues, including ecosystem deposition leading to acidification, ozone, and visibility reduction.*

2014 Update: Linkages between PM and other issues continue to be recognized. There are emerging issues that could impact future $PM_{2.5}$ air quality management policies including climate change, the changing energy landscape, new sources of transboundary PM, and the need to manage PM in a multi-pollutant context.

2004 Assessment Conclusion: *The results of the Assessment indicate that there is a significant relationship between the emissions of PM and PM precursors and elevated PM levels in both Canada and the U.S. The transboundary transport of PM and PM precursors can be significant enough in some regions to potentially compromise the attainment of national standards. The information presented in this Assessment provides the scientific foundation to support the future development of joint strategies under a PM Annex pursuant to the Agreement.*

2014 Update: Because of the important health and environmental effects associated with $PM_{2.5}$, it would be beneficial for both countries to track progress in and exchange information relevant to achieving $PM_{2.5}$-related emissions reductions, air quality improvement, and program implementation over time. Doing so would provide confidence to each country that future $PM_{2.5}$ concentrations in the border region will be below the NAAQS and CAAQS. It would also offer an ongoing opportunity to determine how emerging issues, such as climate change and an evolving energy landscape, may be gradually altering $PM_{2.5}$ concentrations and transboundary transport.

Given the information in this assessment and the important role of $PM_{2.5}$ in air quality management activities in both Canada and the U.S., there would be value in addressing $PM_{2.5}$ in some manner under the Canada – U.S. Air Quality Agreement.

7.2 Important Considerations for Future Analyses

7.2.1 Quantifying the magnitude of recent transboundary transport

This assessment includes ambient observations from both Canada and the U.S. up through 2011, showing recent decreases in $PM_{2.5}$ in both countries. The air quality modelling results available to quantify the impact of U.S. emissions on Canada and vice versa used 2006 as the base year for emissions and meteorology. As was mentioned previously, these results and analyses of back trajectories for selected Canadian monitoring sites from data for the years 2000 through 2010 essentially provide a relative sense of transboundary transport given that there have been changes in emissions and ambient levels since those years. Thus, in general, they are used to qualitatively illustrate the impact of key factors that impact transboundary transport, such as meteorological conditions and spatial distribution. To better inform the present-day absolute magnitude of transboundary transport, however, the results need to be updated because of the decrease in the emissions of key $PM_{2.5}$ precursors in both Canada and the U.S. during the past five years.

Quantifying the actual magnitude of transport along the Canada – U.S. border more recently (e.g. since 2010) requires new targeted analyses using more recent emission inventories and monitoring data. These analyses would likely benefit from utilizing multiple modelling tools, such as AURAMS, GEM-MACH and CMAQ, to capture meteorological and chemical transport processes and uncertainty, in conjunction with monitoring data, to ground truth the modelled ambient concentrations levels. In addition, back trajectory analyses, such as those discussed in Chapter 5, could be conducted with more recent $PM_{2.5}$ mass and speciation data and meteorology.

7.2.2 Transboundary transport and potential human health impacts

Future emissions of $PM_{2.5}$ precursors are projected to decrease significantly during the next decade in both Canada and the U.S., resulting in lower ambient $PM_{2.5}$ concentrations. However, fully validated model predictions of 2020 $PM_{2.5}$ with complete U.S. emission projections for comparison with the current and future Canadian and U.S. standards were not available for the Assessment. A combination of 2016 and 2020 scenarios was used to create an estimate of future U.S. emissions. Additional analyses of available modelling scenarios for 2020 and newer model runs are needed to more realistically determine and better understand the predicted $PM_{2.5}$ levels over the Canada – U.S. border regions. These improved predictions also need to be coupled with the most up-to-date concentration–response functions to estimate the future magnitude of human health impacts in these regions attributable to both transboundary and local $PM_{2.5}$. These estimates are important for more fully informed discussions about approaches to managing air quality, in particular $PM_{2.5}$, and protecting public health in areas along the Canada – U.S. border. Furthermore, given the increasing recognition of the importance of local emissions over parts of the border region, more detailed, high-resolution modelling can be expected to provide more insight into the causes of the higher $PM_{2.5}$ concentrations. This needs to be done for the present time and for the future, particularly over populated areas near the border where $PM_{2.5}$ can move back and forth, making net transboundary flow more difficult to determine (e.g. the Pacific Northwest).

As discussed in Chapter 6, both the changing climate and the evolving energy landscape could have a substantial impact on future ambient $PM_{2.5}$ concentrations, including alterations in the composition of particles and in the spatial and temporal distributions at regional and local scales. However, there is uncertainty about how changes in climate and in energy production and use will ultimately impact $PM_{2.5}$ concentrations and, in particular for this report, transboundary flow of $PM_{2.5}$ along the Canada – U.S. border.

Given the uncertainties about the future, it is important to continue to track $PM_{2.5}$ and precursor emissions, including additional emphasis on their location and magnitude in areas of new energy resource development, production and use. It is also important to monitor ambient $PM_{2.5}$ concentrations and to conduct modelling and data analyses to track trends. The goal of this research will be to measure progress toward anticipated reductions and/or detect new areas of concern, thereby enabling continued assessment of transboundary transport.

Finally, current assessments of the transboundary transport of $PM_{2.5}$ rely on surface monitoring networks and modelling studies, which are influenced by the spatial and temporal resolution of the networks and by uncertainties in the model inputs, such as emission inventories and model processes. As the ability of geostationary satellites to measure $PM_{2.5}$ becomes better understood (Liu et al.,2009; Paciorek and Liu, 2012) and new satellites are launched (https://directory.eoportal.org/web/eoportal/satellite-missions/t/tempo), it will become more feasible to use satellite data to assess the transboundary transport of PM. Such possibilities will be important to explore in the future.

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