Regional Air Quality Deterministic Prediction System (RAQDPS)

Technical Note

Last update
June 11, 2015
from version 012 to version 013
<table>
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<tr>
<th>Version</th>
<th>Date</th>
<th>Author</th>
<th>Modification</th>
</tr>
</thead>
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<tr>
<td>1</td>
<td>08/06/2015</td>
<td>M. Moran</td>
<td>• Création du document</td>
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<tr>
<td>2</td>
<td>29/06/2015</td>
<td>R. Pavlovic</td>
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<td>3</td>
<td>24/08/2015</td>
<td>M. Moran</td>
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<td>21/10/2015</td>
<td>M. Moran</td>
<td>• Revisions to Section 5 and cover-to-cover editing</td>
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<td>5</td>
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<td>10/11/2015</td>
<td>M. Moran</td>
<td>• Final edits</td>
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Acronyms

- AEP  Alberta Environment and Parks
- ALM  aircraft, locomotive, and marine
- APEI  Air Pollutant Emissions Inventory (Canada)
- AQ  air quality
- AQMAS  Air Quality Modelling Applications Section (EC)
- CAC  criteria air contaminant
- CMA  Census Metropolitan Area (Statistics Canada)
- CMC  Canadian Meteorological Centre (EC)
- CSI  critical success index
- DA  dissemination area (Statistics Canada)
- DST  Daylight Saving Time
- EC  elemental carbon
- EC  Environment Canada
- En-VAR  ensemble-variational (data assimilation)
- EPA  Environmental Protection Agency (U.S.)
- EPS  emissions processing system
- FAR  false alarm ratio
- GEM  Global Environmental Multiscale model
- GEM-MACH  Global Environmental Multiscale model – Modelling of Air quality and Chemistry
- GIS  Geographic Information System
- G-M  short form of “GEM-MACH”
- HES  Households and the Environment Survey (Statistics Canada)
- LAM  limited area model
- LTO  landings and takeoffs
- MB  mean bias
- NAPS  National Air Pollution Surveillance program
- NEI  National Emissions Inventory (U.S.)
- NPRI  National Pollutant Release Inventory (Canada)
- OA-AQ  objective analysis – air quality
- OS  oil sands
- PC  percent correct
- PIRD  Pollutant Inventories and Reporting Division (EC)
- PM  particulate matter
- PM$_{2.5}$  particulate matter with aerodynamic diameter smaller than 2.5 μm
- PM$_{10}$  particulate matter with aerodynamic diameter smaller than 10 μm
- POD  probability of detection
- POM  primary organic matter
- RAQDPS  Regional Air Quality Deterministic Prediction System
- RDAQA  Regional Deterministic Air Quality Analysis
- RDPS  Regional Deterministic Prediction System
- SCC  Source Classification Code (U.S. EPA)
- SMOKE Sparse Matrix Operator Kernel Emissions
- TF transportable fraction
- UMOS-AQ Updateable Model Output Statistics – Air Quality
- URMSE unbiased root mean square error
- VOC volatile organic compound
RAQDPS Version 013: Upgrades to the CMC Operational Regional Air Quality Deterministic Prediction System Released in June 2015

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Summary

This technical note describes a new, hourly, gridded emissions input data set that was implemented in the CMC operational air quality forecast model RAQDPS (Regional Air Quality Deterministic Prediction System) on 11 June 2015. The objective of this change was to update and improve the emissions data set being used by the RAQDPS. The new emissions data set is based on more recent Canadian and U.S. national emissions inventories and has also been impacted by improvements made to the emissions processing system that is used to transform emissions inventories into model-ready emissions files. No other changes were made to the RAQDPS for this new operational version 013.

The previous RAQDPS emissions input data set (named SET0) was based on the 2006 Canadian national emissions inventory (version 1) and a projected 2012 U.S. emissions inventory projected from the 2005 U.S. national inventory (version 4). The new emissions input data set (named SET2.1.1) is based on the 2010 Canadian national emissions inventory (version 1) and the 2011 U.S. national emissions inventory (version 1). For the combined new Canadian and U.S. inventories versus the older inventories, SO\textsubscript{2} emissions decreased by 47\%, NO\textsubscript{x} emissions increased by 5\%, VOC emissions decreased by 1\%, NH\textsubscript{3} emissions decreased by 5\%, CO emissions decreased by 6\%, PM\textsubscript{2.5} emissions were nearly unchanged, and PM\textsubscript{10} emissions decreased by 2\%.

Methodological improvements to the emissions processing performed to generate the SET2.1.1 emissions files include the use of (a) 25 new or updated Canadian and five new U.S. spatial surrogate fields, (b) a land-use-dependent transportable fraction field in place of simple 0.25 factor to scale Canadian fugitive dust emissions to account for near-source capture, (c) new temporal profiles for all 3,384 Canadian major point sources and a number of area, on-road, and off-road sources, and (d) a new library of PM speciation profiles and additional VOC speciation profiles as well as (e) the removal of emissions from five major Canadian facilities that either closed or drastically reduced their emissions after 2010 and the addition of emissions from one new oil sands mine in northeastern Alberta that became operational after 2010.
The RAQDPS was tested with the new emissions data set for 2014 winter and summer periods and a 2015 spring period. The results showed an improvement to wintertime forecasts for PM$_{2.5}$, NO$_2$, and O$_3$. In some urban centres like Vancouver, wintertime PM$_{2.5}$ forecasts were greatly improved due to reductions in episodic overpredictions. Summertime forecasts showed a very slight improvement and/or neutral impact overall with the new emissions data set. However, the summertime results showed a substantial improvement to the PM$_{2.5}$ forecasts in eastern Canada and the U.S., where episodic overpredictions had been a problem. The effect on other pollutant fields was mixed. The spring 2015 forecasts also showed an overall improvement, including a large reduction in URMSE values for PM$_{2.5}$.

1. Introduction

The Regional Air Quality Deterministic Prediction System (RAQDPS) has been run operationally by Environment Canada (EC) since summer 2001. Beginning in November 2009 the regional air quality (AQ) model at the core of the RAQDPS was changed to a limited-area configuration of the GEM-MACH model, which consists of an on-line chemical transport model embedded within the GEM model, EC’s multi-scale operational weather forecast model (e.g., Moran et al., 2009, 2012; Im et al., 2015; Coté et al., 1998a,b; Charron et al., 2012). The acronym GEM stands for “Global Environmental Multiscale” while the acronym MACH stands for “Modelling Air quality and Chemistry”. The RAQDPS is run twice daily to produce 48-hour forecasts of hourly ozone (O$_3$), nitrogen dioxide (NO$_2$), and particulate matter with aerodynamic diameter smaller than 2.5 μm (PM$_{2.5}$) fields on a North American grid. Over the 5-year-plus period since 2009 there have been 12 upgrades of differing breadth and complexity made to the RAQDPS. For example, in 2009 the model grid had 15-km horizontal grid spacing and 58 vertical levels extending from the surface to 0.1 hPa, whereas since 2012 the grid has had 10-km horizontal grid spacing and 80 vertical levels extending over the same atmospheric depth.

RAQDPS forecasts are used as guidance by operational AQ forecasters at EC and elsewhere and are also made available to the public through EC’s public-domain weather website (see http://www.weather.gc.ca/aqfm/index_e.html). In addition, as described in Moran et al. (2012), these forecasts are combined with hourly AQ surface measurements to generate both point-specific hourly statistical AQ forecasts for Canadian urban centres using the Updateable Model Output Statistics for Air Quality (UMOS-AQ) post-processing package (Wilson and Vallée, 2002, 2003; Antonopoulos et al., 2010; Moran et al., 2012) and hourly North American AQ surface objective analyses (OA) using a combination of AQ measurements and gridded model fields with the Regional Deterministic Air Quality Analysis (RDAQA) post-processing package (Robichaud and Ménard, 2014).
Table 1 summarizes the 12 upgrades that have been made to the RAQDPS since GEM-MACH became Environment Canada’s operational air quality forecast model in November 2009. The last technical note related to the RAQDPS described the changes that were made up to February 2013 for RAQDPS versions 007, 008, and 009 (Moran et al., 2013). The subsequent changes for RAQDPS versions 010, 011, and 012 that were made up to March 2015 were relatively minor and consisted of updates to the GEM model. Note that these changes to GEM affected both the Regional Deterministic Prediction System (RDPS), the operational regional meteorological model that pilots RAQDPS runs, and the RAQDPS itself.

This technical note describes the upgrades and improvements that were made to generate a new set of hourly, gridded, anthropogenic input emissions fields, named SET2.1.1, which was implemented on 11 June 2015 in version 013 of the RAQDPS. This new emissions input data set is based on more recent Canadian and U.S. national criteria-air-contaminant (CAC) emissions inventories and it has also been impacted by improvements made to the emissions processing system (EPS) that is used to transform emissions inventories into model-ready emissions files. Differences between this new set of emissions and the previous operational emissions file set, named SET0, which was used by the RAQDPS during the nearly four-year period from October 2011 to June 2015, are also characterized in this technical note as are the improvements in forecast performance due to the use of this new emissions file set. Note that no other changes were made to the RAQDPS in the upgrade from version 012 to 013, including to the calculation of natural emissions from biogenic and oceanic sources. Note too that natural emissions from wildfires, lightning, and Aeolian processes are not considered by the RAQDPS at this time.

Section 2 of this technical note provides more details about the changeover to newer input emissions inventories, Section 3 summarizes the methodological changes to emissions processing made since 2011, and Section 4 characterizes the differences between the old SET0 and new SET2.1.1 emissions file sets. Section 5 then presents the results of recent objective performance evaluations that characterized the impacts on AQ forecasts resulting from the change in the input emissions file set used by the RAQDPS, from SET0 to SET2.1.1. Finally, Section 6 provides a summary and an indication of future work.

### Table 1: Chronology of RAQDPS Versions Since Implementation of GEM-MACH AQ Model

<table>
<thead>
<tr>
<th>Date</th>
<th>RAQDPS Version</th>
<th>GEM / PHYS / GEM-MACH Versions</th>
<th>Event / Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>18 Nov 2009</td>
<td>001</td>
<td>3.3.0 / 4.5 / 1.3.0a</td>
<td>GEM-MACH15 implemented into CMC operational suite</td>
</tr>
<tr>
<td>2 Mar 2010</td>
<td>002</td>
<td>3.3.0 / 4.5 / 1.3.0a</td>
<td>New emissions files introduced with modified primary PM$_{2.5}$ emissions over some Cdn provinces</td>
</tr>
<tr>
<td>Date</td>
<td>Time</td>
<td>Version</td>
<td>Description</td>
</tr>
<tr>
<td>------------</td>
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<td>-----------</td>
<td>---------------------------------------------------------------------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>20 Oct 2010</td>
<td>003</td>
<td>3.3.0 / 4.5 / 1.3.0a</td>
<td>Meteorological piloting model changed from GEM15-global to GEM-LAM15</td>
</tr>
<tr>
<td>25 Oct 2011</td>
<td>004</td>
<td>3.3.3 / 4.7.2 / 1.4.4</td>
<td>New GEM and G-M versions and new emissions file set (named SET0)</td>
</tr>
<tr>
<td>22 Dec 2011</td>
<td>005</td>
<td>3.3.3 / 4.7.2 / 1.4.5</td>
<td>New G-M version with correction for radiation calculation at model top</td>
</tr>
<tr>
<td>2 May 2012</td>
<td>006</td>
<td>3.3.6 / 4.7.2.1 / 1.4.6</td>
<td>New versions for the new supercomputer used by operations, including bug fixes</td>
</tr>
<tr>
<td>3 Oct 2012</td>
<td>007</td>
<td>3.3.8 / 5.0.4.2 / 1.5.0</td>
<td>New GEM and G-M versions, new grid (10 km), and new emissions file set (named SET1); RDPS300 piloting</td>
</tr>
<tr>
<td>20 Nov 2012</td>
<td>008</td>
<td>3.3.8 / 5.0.4.2 / 1.5.0</td>
<td>Replacement of emissions file set introduced in Oct. 2012 (i.e., SET1) with Oct. 2011 version (SET0)</td>
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<tr>
<td>26 Feb 2013</td>
<td>009</td>
<td>3.3.8 / 5.0.4.2 / 1.5.1</td>
<td>New G-M version with 3 bug fixes</td>
</tr>
<tr>
<td>10 Apr 2014</td>
<td>010</td>
<td>3.3.8.1 / 5.0.4.3 / 1.5.2</td>
<td>Migration to GEM v3.3.8.1 and PHYS v5.0.4.3</td>
</tr>
<tr>
<td>18 Nov 2014</td>
<td>011</td>
<td>3.3.8.2 / 5.0.4.4 / 1.5.3</td>
<td>Migration to GEM v3.3.8.2, PHYS v5.0.4.4, and RDPS-EnVar</td>
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<tr>
<td>18 Mar 2015</td>
<td>012</td>
<td>3.3.8.2-isba / 5.0.4.4 / 1.5.4</td>
<td>Migration to GEM v3.3.8.2-isba</td>
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<td>11 Jun 2015</td>
<td>013</td>
<td>3.3.8.2-isba / 5.0.4.4 / 1.5.4</td>
<td>Replacement of emissions file set introduced in Oct. 2011 (SET0) with new version (SET2.1.1)</td>
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2. Upgrades to Emissions Inventories

2.1 Sets of input emissions fields used by the RAQDPS since 2009

Table 1 lists all of the changes made to the RAQDPS since November 2009. These changes have included a reduction in horizontal grid spacing (from 15 km to 10 km), an increase in the number of vertical levels (from 58 to 80), some bug fixes, migration to newer GEM versions, and updates to the file set of hourly gridded emissions fields that are input by the RAQDPS. (Note that we frequently refer to an emissions “file set” because a full set of RAQDPS input emissions files consists of 168 files, where each file corresponds to a particular month, a particular day of the
Emissions fields are a key input for any AQ model, and the emissions file set used by the RAQDPS has been updated five times since November 2009 (see Table 2). The emissions file set that was used before SET2.1.1 was called SET0. As can be seen from Table 2, SET0 was introduced into the RAQDPS in October 2011, was briefly replaced from 3 October to 20 November 2012 with another emissions file set (SET1), but was reintroduced on 20 November 2012 and was used by the RAQDPS until 11 June 2015. The reasons for the replacement of the SET0 emissions file set by the SET1 emissions file set followed a short time later by the reintroduction of the SET0 emissions file set were described by Moran et al. (2013). The focus of this technical report is the SET2.1.1 input emissions file set that replaced the SET0 emissions file set on 11 June 2015.

Table 2: Chronology of Emissions File Sets Used in Different RAQDPS Versions.

<table>
<thead>
<tr>
<th>Date</th>
<th>RAQDPS Version</th>
<th>Event / Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>18 Nov 2009</td>
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<td>004</td>
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</tr>
<tr>
<td>3 Oct 2012</td>
<td>007</td>
<td>New GEM and GEM-MACH versions and new emissions file set (named SET1); RDPS300 piloting</td>
</tr>
<tr>
<td>11 Jun 2015</td>
<td>013</td>
<td>Replacement of emissions file set introduced in Oct. 2011 (SET0) with new version (named SET2.1.1)</td>
</tr>
</tbody>
</table>

The SET0 emissions file set was based on the 2006 Canadian national CAC emissions inventory (version 1) and a projected 2012 U.S. national CAC emissions inventory projected from the 2005 U.S. inventory (version 4) (see Ménard et al. [2011]) and Moran et al. [2013] for more information). To generate the new SET2.1.1 emissions file set, these two older national inventories were replaced with two newer inventories, the 2010 Canadian national CAC emissions inventory (version 1) and the 2011 U.S. national CAC emissions inventory (version 1), which are described in the following two sections. Note that Mexican emissions must also be considered since the southern portion of the RAQDPS grid includes part of northern Mexico (e.g., Fig. 5). A 1999 Mexican anthropogenic emissions inventory that was used for generating the SET0 emissions file set (Moran et al., 2012, 2013) was also used for the SET2.1.1 emissions file set and will not be discussed further.
2.2 2010 Canadian National Emissions Inventory

Version 1 of the 2010 Canadian CAC emissions inventory, also referred to as the Canadian Air Pollutant Emissions Inventory (APEI), is a comprehensive national anthropogenic emissions inventory that includes emissions from point sources, area sources, on-road mobile sources, and off-road mobile/aircraft/locomotive/marine sources for base year 2010. It was compiled by the Pollutant Inventories and Reporting Division (PIRD) of Environment Canada using both top-down and bottom-up approaches (see https://www.ec.gc.ca/inrp-npri/default.asp?lang=En&n=B85A1846-1). For example, major-point-source emissions were compiled using a bottom-up approach based on facility-specific emissions reported to EC’s National Pollutant Release Inventory (NPRI) at the facility level, whereas area-source emissions were mainly compiled using a top-down approach based on activity levels and emission factors. Details of the procedures for the compilation of the 2010 Canadian inventory are described in Environment Canada (2013). Note that some procedures had changed since the 2006 APEI was compiled. The 2010 inventory database compiled by PIRD was then transformed by the Air Quality Modelling Applications Section (AQMAS) of Environment Canada in Oct. 2013 into a form that could be input by the Sparse Matrix Operator Kernel Emissions (SMOKE) emissions processing system (CEP, 2012; AQMAS, 2014; Sassi et al., 2014).

Table 3 shows a comparison of national-level totals of the 2006 Canadian CAC emissions (version 1) based on the SET0 emissions file set and the 2010 Canadian CAC emissions (version 1) based on the SET2.1.1 emissions file set. These values reflect the emissions that the RAQDPS “sees”, that is, the combination of both the original inventory values and SMOKE processing (see next section). In general, inventory totals are not changed by the EPS, but there are a few exceptions.

Emissions of most CAC species are lower for the 2010 base year vs. the 2006 base year, largely reflecting reductions associated with the implementation of Canadian emission control legislation targeting transportation and industry. In particular, total SO$_2$ emissions decreased by 35% during this four-year period; this large reduction was due to reductions in non-ferrous smelting and electric power generation over this period (CCME, 2013). Total NOx emissions are also lower by 14% due largely to decreases in industrial sources and on-road vehicle emissions.

Table 3: Comparison of SET0 and SET2.1.1 Canadian and U.S. national CAC emissions (tons). Note that both wildfire and prescribed-burning emissions have been excluded for both Canada and the U.S., transportable-fraction scaling has been applied to fugitive dust emissions, and selected Canadian point sources that closed during or after 2010 have been removed from the 2010 Canadian inventory and one large new Canadian point source that started operation in 2013 has been added.
The two species whose emissions increased over this period were the two particulate-matter species, PM$_{2.5}$ and PM$_{10}$. However, these apparent increases were mainly due to two methodological changes. First, as discussed by Moran et al. (2013), PM$_{2.5}$ emissions of 72 Kt y$^{-1}$ and PM$_{10}$ emissions of 363 Kt y$^{-1}$ from construction activities that were included in the 2006 Canadian national CAC inventory were not considered when preparing the SET0 emissions file set due to problems with spatial allocation and resulting overpredictions of PM$_{2.5}$ surface concentrations in some Canadian cities. However, comparable 2010 construction emissions included in the 2010 Canadian national CAC inventory were considered when preparing the SET2.1.1 emissions file set due to the availability of improved spatial surrogate fields for the spatial allocation of these emissions. And second, as discussed in Section 3.4, the transportable-fraction (TF) scaling-factor field that was applied to fugitive dust emissions fields after SMOKE processing to account for near-source capture and removal was very different for SET0 vs. SET2.1.1. Although the TF field used for the SET2.1.1 PM$_{2.5}$ and PM$_{10}$ emissions fields reduces fugitive dust emissions by a smaller amount than the TF field used for the SET0 PM$_{2.5}$ and PM$_{10}$ emissions fields, the newer TF field has a stronger physical basis and is thought to be more accurate and more representative.

### 2.3 2011 U.S. National Emissions Inventory

The U.S. National Emissions Inventory (NEI) is a comprehensive and detailed estimate of air emissions of both criteria and hazardous air pollutants from all air emissions sources, including point sources, area sources, on-road mobile sources, and off-road mobile/aircraft/locomotive/marine sources. The NEI is prepared every three years by the U.S.
Environmental Protection Agency (EPA) based primarily upon emission estimates and emission model inputs provided by state, local, and tribal air agencies for sources in their jurisdictions, and supplemented by data developed by the EPA. Recent NEI base years have included 1999, 2002, 2005, and 2008 (see http://www.epa.gov/ttn/chief/eiinformation.html). The 2011 NEI is the next inventory in this sequence. Version 1 of the 2011 inventory, which was used to generate the SET2.1.1 emissions file set, was released on 30 September 2013.

Table 3 shows a comparison of national-level totals of the projected 2012 U.S. CAC emissions inventory used in the SET0 emissions file set and the 2011 U.S. CAC emissions inventory (version 1) used in the SET2.1.1 emissions file set. Note that the one significant change to the inventory values made by the SMOKE emissions processing system was the application of transportable-fraction (TF) scaling to fugitive dust emissions; a land-use-dependent TF field was applied for both inventories (see Section 3.4). With the exception of SO\(_2\) emissions, the emissions of most species are similar for the two years in percentage terms, consistent with the closeness of the two base years (i.e., 2012 vs. 2011). The relatively small differences suggest that the assumptions that were used to project (i.e., extrapolate) 2005 emissions to 2012 were reasonably accurate since the 2012 projected emissions values are quite close to the actual 2011 emissions values.

In the case of SO\(_2\), however, the projection assumptions did not reflect the actual implementation of several SO\(_2\) emissions-control programs for large point sources that occurred in the U.S. between 2005 and 2011. As a consequence, actual 2011 SO\(_2\) emissions were 48\% smaller than projected 2012 emissions, and as shown in Section 5, this reduction had a significant impact on forecast PM concentrations since SO\(_2\) is an important precursor of atmospheric PM. It is worth noting that satellite measurements of SO\(_2\) indicate a 40\% decline in SO\(_2\) values over the largest U.S. coal-fired power plants between 2005-2007 and 2008-2010, a value that was consistent with the reported 46\% reduction in annual emissions due to the implementation of new SO\(_2\) pollution control measures at these facilities over this period (Fioletov et al., 2011).

### 2.4 Comparison of old and new national emissions inventories

Table 3 also shows the combined totals of the Canadian and U.S. inventories (after SMOKE processing) used to generate the SET0 and SET2.1.1 emissions file sets. For the combined new Canadian and U.S. inventories versus the older inventories, SO\(_2\) emissions have decreased by 47 percent, NO\(_x\) emissions have increased by 5 percent, VOC emissions have decreased by 1 percent, NH\(_3\) emissions have decreased by 5 percent, CO emissions have decreased by 6 percent, PM\(_{2.5}\) emissions are virtually unchanged, and PM\(_{10}\) emissions have decreased by 2 percent.

These are, of course, only overall changes and are not uniform geographically. For example, NO\(_x\) emissions in the newer inventories decreased in Canada but increased in the U.S. whereas
PM$_{2.5}$ and PM$_{10}$ emissions increased in Canada but decreased in the U.S. Figure 7 shows detailed spatial patterns of some of emissions difference fields.

### 3. Upgrades to Emissions Processing Tools and Methodology

Air quality models, including GEM-MACH, cannot directly use the emissions from emissions inventories as inputs. The reason is that there is a major mismatch between the contents of emissions inventories versus the level of detail about emissions needed by an AQ model. In contrast to a typical inventory, with annual or monthly emissions reported by jurisdiction (e.g., province or county) for a small number of pollutants (e.g., criteria air contaminants), GEM-MACH needs emissions fields for every hour of every day of the year for each model grid cell for the larger set of model chemical species (e.g., NO, NO$_2$, and individual or lumped VOC species).

For this reason, the Canadian, U.S., and Mexican national CAC emissions inventories described in the previous section must first be processed using a software package called an emissions processing system (EPS). The main functions of an EPS are spatial allocation, temporal allocation, and chemical speciation (Dickson and Oliver, 1991; Houyoux et al., 2000; Pouliot et al., 2012). Spatial allocation is performed to distribute non-point source emissions reported by jurisdiction to appropriate grid cells (by contrast, point-source emissions, whose locations are known exactly, can easily be assigned to the appropriate grid cell). Temporal allocation is performed to convert the inventory annual or monthly emission rates into hourly values. Chemical speciation is performed to split certain criteria pollutants (NO$_x$, VOC, PM$_{2.5}$, PM$_{10}$) into individual model species.

A number of upgrades have been made over the past four years to the EPS used to create the emissions file sets used by the RAQDPS. This section describes the more significant changes.

#### 3.1 Changes to the emissions processing system

The SMOKE (Sparse Matrix Operator Kernel Emissions) emissions processing system was used to generate both the SET0 and SET2.1.1 emission file sets. The SMOKE system is developed and distributed by the Institute for the Environment at the University of North Carolina at Chapel Hill in cooperation with the U.S. EPA (Houyoux et al., 2000; CEP, 2003; https://www.cmascenter.org/smoke/). It is a widely used EPS for preparing emissions for AQ models and was adopted by Environment Canada for emissions processing in 2004. SMOKE version 2.3 was used to generate the SET0 emissions file set while SMOKE versions 2.3, 2.6, 3.1, and 3.5.1 were used for various steps to generate the SET2.1.1 emissions file set.
3.2 Updated Canadian spatial surrogate fields

Spatial surrogate fields are used to allocate inventory emissions, which are reported by jurisdiction (i.e., Canadian provinces and territories, U.S. counties, Mexican states), to model grid cells. However, as their name suggests these surrogate fields are only proxies, that is, they are at best only a rough approximation of the actual spatial distribution of the emissions from the source sector being considered. Given this limitation, then the spatial allocation can be improved if a better surrogate can be found or constructed. Twenty-five new or updated Canadian spatial surrogate fields are now available, and 24 of these new surrogates plus another 90 Canadian surrogates were used to produce the new SET2.1.1 emissions file set (Zheng et al., 2015). The 25 new or updated spatial surrogates are listed in Table 4 and can be described in groups as follows.

3.2.1 Population and dwelling surrogates

Spatial distributions of population and dwellings are widely used spatial surrogates for many types of emissions. The Canadian population and dwellings surrogates used to generate the SET0 emissions file set were based on the 2006 Canadian census. Following the release of data from the 2011 Canadian census at the dissemination-area (DA) level in 2012, updated population and dwelling spatial surrogates have been developed (surrogates 100 to 103 in Table 4). The 56,204 DAs in the 2011 census, ranging from 50 DAs in Nunavut to 19,964 DAs in Ontario, correspond to the neighbourhood scale and each DA typically contains 400 to 700 persons. Each DA is classified as to whether it belongs to a small, medium, or large urban area or to a rural area, and the spatial area of each DA is also provided, which allows population or dwelling density to be calculated. Figure 1a shows a collation of plots of the 13 provincial and territorial population spatial surrogate fields derived from the 2011 Canadian population data set on the 15-km rotated latitude-longitude RAQDPS grid; each of these 13 spatial surrogate fields gives the fraction of the provincial or territorial population contained in each model grid cell, so that the sum of each spatial surrogate field over the model grid is unity. Note that for dwellings, the 2011 census provides two fields: TDWELL (total dwellings) and URDWELL (usual-resident-occupied dwellings). For constructing dwellings-based surrogates the latter field was used.

The three “capped” spatial surrogates 104, 107, and 108 listed in Table 4 are similar to surrogates 101, 100, and 100, respectively, except that an upper limit has been imposed on each. The reason to impose an upper limit is that a linear relationship may be expected between emissions magnitude and the magnitude of a surrogate up to a certain level but not beyond. For example, population density may be a reasonable spatial surrogate for emissions from local traffic up to the point where the two-dimensional road network has reached its physical limit even while population density continues to increases due to the presence of high-rise apartment and condominium buildings (e.g., Gately et al., 2013). For surrogate 104, dwelling density was capped at 600 dwellings km\(^{-2}\). For surrogates 107 and 108, population density was capped at
2,000 persons km\(^{-2}\) (Zheng et al., 2015). Note that the average size of a Canadian household is 2.5 persons (see http://www.statcan.gc.ca/tables-tableaux/sum-som/l01/cst01/famil53a-eng.htm).

Figure 1b shows a collation of plots of surrogate 107, capped urban population, for the 13 Canadian provinces and territories. The impact of imposing a cap on the population and dwelling spatial surrogates is to decrease the allocation of emissions to high-density urban cores while increasing the allocation to suburban and rural areas.

### 3.2.2 On-road mobile surrogates

Emissions from on-road mobile sources are very important for NO\(_x\), VOC, CO, and PM\(_{2.5}\). In constructing the SET0 emissions file set, two spatial surrogates based on road-network geometry and population density were used to allocate Canadian on-road emissions, one for trucks and one for cars, whereas a set of six spatial surrogates was used to allocate U.S. on-road emissions (Zhang et al., 2010). We have now extended the U.S. approach, in which the road network is split into three road categories (primary, secondary, and local) and into urban and rural locations, to process Canadian on-road emissions. Canadian road-network information was obtained from the 2014 version of the Canadian national road network shapefile, which considers 13 road categories with geographic locations of individual lanes correct to a few meters (see http://geogratis.gc.ca). The separation of the national road network into urban and rural subnetworks was done based on the classification of DAs across Canada in the 2011 census to either an urban or rural category (see Zheng et al., 2015). Figure 1c shows a collation of plots of one of the six new Canadian on-road spatial surrogates, number 202, the rural primary road miles surrogate. Figure 2 shows the impact of processing Canadian on-road emissions using the two older Canadian on-road surrogates and the six new surrogates. It can be seen that the use of the six new surrogates has allocated more on-road emissions to rural roads and reduced the allocation to urban areas. The spatial distribution of Canadian on-road emissions in Figure 2b based on the six new surrogates is also more like the spatial distribution of U.S. emissions. More information about the development of these surrogates may be found in Zhang et al. (2012).

Another spatial surrogate, 940, is used to allocate fugitive dust emissions from the movement of vehicles on paved roads. It is an update to an older surrogate based on the paved-road network. The same 2014 version of the Canadian national road network was used to construct this surrogate by combining multiple types of paved roads (primary and secondary highways, arterial roads). Note that local roads were not considered in the construction of this surrogate nor was any urban-rural split.

### 3.2.3 Alberta oil sands facility surrogates

Surrogates 215 to 217 are new and correspond to six large Canadian surface mines in the Athabasca oil sands region of northeastern Alberta. These mines are treated as point sources in the 2010 Canadian emissions inventory but they are each 10 to 20 km across and hence may span
two or three 10-km model grid cells in both the west-east and south-north directions. For this reason spatial surrogates were constructed for each of these mines for emissions from mine faces, from tailing ponds, and from on-site processing plants. This may be the first time that spatial surrogates have been used to allocate emissions from an industrial facility. More details about the construction of these spatial surrogates is given in Zhang et al. (2015). In addition, surrogate 946 has also been revised to account for the Athabasca oil sands mines. This surrogate is used to allocate emissions from off-road vehicles associated with the construction and mining industries. However, the 2010 Canadian inventory provides off-road emissions associated with the captive mining fleets at the six large oil sands mines separately rather than as part of a provincial total. Accordingly, surrogate 215 is used to allocate emissions from the oil sands mines and mining fleets. It was then necessary to modify surrogate 946 for the province of Alberta so that the Athabasca oil sands area is removed from consideration; otherwise, construction and mining emissions from other parts of the province would be erroneously allocated to the oil sands area.

3.2.4 Industrial sector surrogates

Surrogates 221 and 321 are based on place-of-work data from the 2006 Canadian census for the mining industry and wood-processing industry, respectively. They are used to allocate emissions from small mines and wood processing facilities that are not included in the Canadian point-source inventory to locations where there are workers in these industries. However, some workers in these industries work at corporate headquarters and in other offices rather than at the mines or plants themselves. In order not to allocate mining or wood-processing (or heavy construction) emissions to urban or suburban locations far from the actual mines or plants, for surrogate 221 any mining-industry workers located in 12 large Canadian cities were removed from consideration, and for surrogate 321 any wood-industry workers not located in forested areas were removed from consideration. Figure 1d shows a collation of plots of surrogate 321 for the 13 provinces and territories, and Figure 3a shows the difference field for the revision of surrogate 321 vs. the original version.

Surrogate 221 was also used for the spatial allocation of some construction emissions. Construction-industry emissions are reported for three subsectors: (a) residential construction; (b) bridge, tunnel, and road construction; and (c) all other construction. A single spatial surrogate based on place-of-work data from the 2006 Canadian census for the construction industry was used to allocate construction-industry emissions from the three subsectors for the SET0 emissions. For processing the SET2.1.1 emissions, however, three spatial surrogates have been used to allocate construction emissions: urban area (surrogate 996) for the residential construction subsector; paved-road density (surrogate 940; see section 3.2.2) for the bridge, tunnel, and road construction subsector; and mining-industry place-of-work data (surrogate 221) for all other construction emissions.
3.2.5 Off-road mobile surrogates

The next six surrogates in Table 4 are associated with transportation sources. Surrogate 350 is a new surrogate to allocate emissions from marine pleasure craft. Previously, only one spatial surrogate (945) was used to allocate marine emissions, but it was constructed from a commercial-marine-vessel shapefile and was not at all representative of the spatial distribution of pleasure craft. Surrogate 350 has been constructed from two other shapefiles; it starts from the location of lakes in each province and territory, but then the distribution of lakes is modulated using the distribution of population on the assumption that if nobody lives in the vicinity of the lake, then it must be inaccessible and hence pleasure craft will not be used on that lake whereas if many people live in the vicinity of the lake, then many pleasure craft will be used on that lake.

Surrogates 901 to 905 are used to allocate airport and aircraft emissions. Previously, a single surrogate comprised of circular areas 50 km in radius centered on the locations of airports in each province and territory was used for the SET0 emissions file set, but this surrogate assumed implicitly that all airports in a jurisdiction are equally busy. Another spatial surrogate (481) based on place of work for air transportation workers was used to generate the SET1 emissions file set (see Moran et al., 2013). The five new surrogates are based on annual landing and takeoff (LTO) statistics for one year, 2006, for the 211 busiest Canadian airports. The relative values of LTO for each airport are assumed to provide a better representation of the relative values of emissions at each airport. Surrogate 901 is used to allocate emissions from airport ground operations. A circular area one km in radius centred on each airport is used and ground-operation activity is assumed to be proportional to LTO activity. Surrogates 902 to 905 are used to represent aircraft emissions in the planetary boundary layer in the vicinity of an airport during either landing or takeoff. Emissions from military aircraft LTOs are a special case since military aircraft operate at only 31 of the 211 primary airports for which LTO statistics were available. For the other three aircraft categories – commercial, general aviation, and air taxis – it was assumed that they all had the same relative LTO activity for the airports in each province or territory. Figure 1e shows a collation of plots of surrogate 903 (commercial aircraft LTOs) for the 13 provinces and territories. Note that aircraft in-transit emissions between airports, which are in the free troposphere, have not been considered for the RAQDPS.

3.2.6 Residential wood combustion

The last new spatial surrogate in Table 4 is surrogate 951. It is used to allocate Canadian emissions from residential wood combustion (RWC) and it replaces the two spatial surrogates that were used when the SET0 emissions were processed: surrogate 997 for the province of Quebec and surrogate 950 for the rest of Canada. RWC is a very important source of PM$_{2.5}$ emissions in Canada in the wintertime. However, it has been an ongoing challenge to allocate these emissions spatially in a realistic manner. In particular, Canadian RWC emissions appear to have been over-allocated to urban areas, and this has led to episodic overpredictions of
wintertime PM$_{2.5}$ concentrations in some Canadian cities such as Montreal and Vancouver (Moran et al., 2013).

Surrogate 997 for the province of Quebec is based on information from several household surveys of firewood use. The national surrogate, surrogate 950, on the other hand, is a “combination” surrogate that uses the sum of two other spatial surrogates: (a) forest land-use fraction and (b) residential dwelling density. This surrogate as defined allocates RWC emissions preferentially to heavily forested areas and population centres. Note that surrogate 950 has also been defined as an “intersection” surrogate based on the product of forest coverage and dwelling density. Locations with both high dwelling density and forest coverage will be favoured by this surrogate whereas urban cores with no forest coverage will be allocated zero RWC emissions. This intersection-based definition of surrogate 950 was used in the generation of the SET0 emissions, which likely helped avoid problems with wintertime overprediction of PM$_{2.5}$ concentrations in urban downtowns due to RWC emissions. It should be noted that both of these surrogate definitions assume implicitly that urban and rural households burn the same amount of wood, which is contradicted by many surveys of fuel use. As well, neither of these versions of surrogate 950 is directly related to wood consumption statistics whereas surrogate 997 for the province of Quebec does make use of such statistics.

Recently a new data source became available: the 2011 Statistics Canada (SC) Household and the Environment Survey (HES) and its accompanying Energy Use Supplement (EUS). The 2011 HES provides data about household wood consumption at the provincial level but also has data for 28 Census Metropolitan Areas (CMAs) across Canada. A CMA must have a total population of at least 100,000 of which 50,000 or more must live in an urban core. Of these 28 CMAs, 11 are located in Ontario, 6 in Quebec, 3 in B.C., 2 in Alberta, 2 in Saskatchewan, and one in each of Newfoundland, Nova Scotia, New Brunswick, and Manitoba. For those provinces that contain one or more CMAs, these new data provide very useful sub-provincial information about the spatial distribution of RWC emissions; i.e., they give a coarse spatial distribution of provincial RWC emissions between individual CMAs and “the rest of their province” based on levels of wood consumption. The availability of these new data about sub-provincial wood consumption thus provides a much firmer information base for constructing new provincial RWC spatial surrogates.

To build a province-level spatial surrogate a second step must be performed to determine a finer distribution of wood consumption within each CMA and across “the rest of their province”, and then these spatial sub-distributions are combined and normalized by total provincial wood consumption. To describe the distribution of wood consumption within each CMA and within “the rest of their province”, use is made of residential dwelling information (URDWELL) at the DA level from the 2011 Canadian census. Note that Census Metropolitan Areas may include DAs characterized as rural. The basis for the new RWC spatial surrogate for each province and
territory was annual wood consumption by DA. The challenge was to determine this quantity from annual wood consumption by province and by CMA.

Four additional assumptions were made to describe the distribution of wood consumption within CMAs and the rest of each province.

(1) Rural households (as represented by dwellings) will burn more wood on average than urban households; rural households are much more likely to use RWC for space heating than urban households. Households in small population centres (PCs: population < 30,000) are more likely to burn wood than those located in medium or large PCs.
(2) Woodburning is unlikely to occur in high-density, multi-storey, multiple-unit buildings.
(3) RWC for space heating will be higher in colder locations.
(4) Households located within or close to forests are more likely to burn wood than households located in prairie or cleared (agricultural) locations (e.g., southwestern Ontario).

The implementation of these four assumptions required the following additional assumptions:

(1) Rural DAs were assigned a dwelling wood consumption factor of 3 while DAs in small, medium, and large population centres were assigned values of 1.5, 1.0, and 0.75, respectively.
(2) For DAs with a dwelling density between 600 and 3,000 dwellings km\(^{-2}\), the dwelling wood consumption scaling factor was set to 0.25 (this density is likely to be a mix of low-rise and high-rise dwellings) while for DAs with a dwelling density of more than 3,000 dwellings km\(^{-2}\), the factor was set to zero.
(3) A latitude-dependent scaling factor \( S \) was applied to each DA dwelling wood consumption factor to account for increased wood combustion due to colder temperatures.
(4) For each DA located in a non-forested area, the dwelling wood consumption factor was divided by 2 while for each DA located in a forested area, the factor was multiplied by 2.

By making use of all of the above assumptions, a DA-level wood combustion value can then be calculated for all provincial DAs where the sum of the DA-level wood combustion values should equal the provincial total wood combustion value. Dividing each DA-level wood combustion value in a province by the provincial wood consumption total then yields the new RWC spatial surrogate field. Figure 1f shows a collation of plots of surrogate 951 for the 13 provinces and territories and Figure 3b shows the difference field for surrogate 951 vs. surrogate 950.

Table 4: List of Canadian spatial surrogate fields that have been revised (status ‘R’) or added (status ‘N’) since the generation of the SET0 emissions file set in 2011. All were used to generate the SET2.1.1 emissions file set except surrogate 102.

<table>
<thead>
<tr>
<th>ID</th>
<th>Description</th>
<th>Status</th>
<th>Comment</th>
<th>Ref. #</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>Population</td>
<td>R</td>
<td>Based on 2011 Canadian census (was 2006 census previously)</td>
<td>2820</td>
</tr>
<tr>
<td>Code</td>
<td>Description</td>
<td>Type</td>
<td>Source</td>
<td>Notes</td>
</tr>
<tr>
<td>------</td>
<td>-------------</td>
<td>------</td>
<td>--------</td>
<td>-------</td>
</tr>
<tr>
<td>101</td>
<td>Total Occupied Dwelling</td>
<td>R</td>
<td>Based on 2011 Cdn census: usual-resident-occupied dwelling field (URDWELL)</td>
<td>2820</td>
</tr>
<tr>
<td>102</td>
<td>Urban Occupied Dwelling</td>
<td>R</td>
<td>Based on 2011 Cdn census usual-resident-occupied dwelling field (URDWELL) and DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>103</td>
<td>Rural Occupied Dwelling</td>
<td>R</td>
<td>Based on 2011 Cdn census usual-resident-occupied dwelling field (URDWELL) and DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>104</td>
<td>Capped Total Occupied Dwelling</td>
<td>N</td>
<td>Based on 2011 Cdn census usual-resident-occupied dwelling field (URDWELL) and cap of 600 dwellings km⁻²</td>
<td>2170</td>
</tr>
<tr>
<td>107</td>
<td>Capped Urban Population</td>
<td>N</td>
<td>Based on 2011 Cdn census population field, on DA urban/rural assignment, and on cap of 2,000 persons km⁻²</td>
<td>2170</td>
</tr>
<tr>
<td>108</td>
<td>Capped Rural Population</td>
<td>N</td>
<td>Based on 2011 Cdn census population field, on DA urban/rural assignment, and on cap of 2,000 persons km⁻²</td>
<td>2170</td>
</tr>
<tr>
<td>200</td>
<td>Urban Primary Road Miles</td>
<td>N</td>
<td>Based on 2014 Cdn national road network shapefile and 2011 Cdn census DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>202</td>
<td>Rural Primary Road Miles</td>
<td>N</td>
<td>Based on 2014 Cdn national road network shapefile and 2011 Cdn census DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>204</td>
<td>Urban Secondary Road Miles</td>
<td>N</td>
<td>Based on 2014 Cdn national road network shapefile and 2011 Cdn census DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>206</td>
<td>Rural Secondary Road Miles</td>
<td>N</td>
<td>Based on 2014 Cdn national road network shapefile and 2011 Cdn census DA urban/rural assignment</td>
<td>2820</td>
</tr>
<tr>
<td>215</td>
<td>Oil Sands Mines</td>
<td>N</td>
<td>Based on 2010 AEP shapefile and 2013 satellite imagery</td>
<td>1482</td>
</tr>
<tr>
<td>216</td>
<td>Oil Sands Tailings Ponds</td>
<td>N</td>
<td>Based on 2010 AEP shapefile and 2013 satellite imagery</td>
<td>1482</td>
</tr>
<tr>
<td>217</td>
<td>Oil Sands Plants</td>
<td>N</td>
<td>Based on 2010 AEP shapefile and 2013 satellite imagery</td>
<td>1482</td>
</tr>
<tr>
<td>221</td>
<td>Total Mining</td>
<td>R</td>
<td>Removed 12 urban areas from 2006 Cdn census mining place-of-work shapefile to avoid counting head-office workers</td>
<td>2263</td>
</tr>
<tr>
<td>321</td>
<td>Wood Product Manufacturing</td>
<td>R</td>
<td>Restricted worker place-of-work polygons to forested areas to avoid counting head-office workers</td>
<td>2417</td>
</tr>
<tr>
<td>350</td>
<td>Water</td>
<td>N</td>
<td>New surrogate for pleasure-craft emissions: based on intersection of near-shore water area and population</td>
<td>2198</td>
</tr>
<tr>
<td>901</td>
<td>Airport Ground Operations</td>
<td>N</td>
<td>Based on statistics of 2006 annual total LTOs for 211 primary Cdn airports</td>
<td>2245</td>
</tr>
<tr>
<td>902</td>
<td>Military Aircraft LTO</td>
<td>N</td>
<td>Based on statistics of 2006 annual military LTOs for 31 Cdn military airports</td>
<td>2245</td>
</tr>
<tr>
<td>903</td>
<td>Commercial Aircraft LTO</td>
<td>N</td>
<td>Based on statistics of 2006 annual commercial LTOs for 211 primary Cdn airports</td>
<td>2245</td>
</tr>
<tr>
<td>904</td>
<td>General Aviation LTO</td>
<td>N</td>
<td>Same as 903 but for general-aviation LTOs</td>
<td>2245</td>
</tr>
<tr>
<td>905</td>
<td>Air Taxi LTO</td>
<td>N</td>
<td>Same as 903 but for air-taxi LTOs</td>
<td>2245</td>
</tr>
<tr>
<td>940</td>
<td>Paved Roads</td>
<td>R</td>
<td>Based on new paved-road shapefile obtained from combining 4 road types in 2014 Cdn national road network shapefile</td>
<td>2216</td>
</tr>
<tr>
<td>946</td>
<td>Construction and Mining Off-road</td>
<td>R</td>
<td>Updated industrial off-road mobile surrogate for Alberta only to remove Athabasca OS facilities (these are now treated separately)</td>
<td>2305</td>
</tr>
<tr>
<td>951</td>
<td>Wood Consumption Percentage</td>
<td>R</td>
<td>Used Statistics Canada CMA wood consumption data as basis for new surrogate plus urban/rural classification, dwelling density, proximity to forest, and latitude</td>
<td>1431</td>
</tr>
</tbody>
</table>
**Figure 1**: Plots of selected new or revised Canadian spatial surrogates on the RAQDPS15 grid: (a) surrogate 100 – population; (b) surrogate 107 – capped urban population; (c) surrogate 202 – rural primary road miles; (d) surrogate 321 – wood product manufacturing; (e) surrogate 903 – commercial aircraft LTOs; and (f) surrogate 951 – residential wood combustion. Note that each image is a collation of 10 provincial and 3 territorial spatial surrogates.
**Figure 2**: Comparison of NO$_2$ emissions (g/s/grid cell) from on-road sources at 8:00 a.m. local time for a summer weekday on a 2.5-km grid that were generated using (a) previous two Canadian on-road spatial surrogates (962, 992) and (b) six new Canadian on-road spatial surrogates (107, 108, 200, 202, 204, 206). Note that the diurnal temporal profiles used for Canadian on-road emissions were also different for processing these two emissions fields, but the same U.S. spatial and temporal surrogates were used to process the U.S. on-road emissions for both panels.

**Figure 3**: Plots of difference fields (new minus old) for two revised Canadian spatial surrogates on the RAQDPS15 grid: (a) surrogate 321 – wood product manufacturing; (b) surrogate 951 vs. surrogate 950 (intersection version) – residential wood combustion. Note that each image is the collation of the difference fields for 10 provinces and three territories.
### 3.3 Updated U.S. spatial surrogate fields

The same set of 55 U.S. spatial surrogates that was used in the generation of the SET0 emissions file set was also used in the generation of the SET2.1.1 emissions file set. However, the 2011 U.S. inventory that was used for the SET2.1.1 emissions file set contained emissions associated with new source classification codes (SCCs) that were not present in the 2012 projected U.S. inventory. As a consequence it was necessary to employ five additional U.S. spatial surrogates in order to allocate these new SCCs in space: 261 (Total Railroad Density); 271 (Class 1-3 Railroad Density); 680 (Oil and Gas Wells); 801 (Port Areas); and 802 (Shipping Lanes).

### 3.4 Land-use-dependent transportable fraction field

It has been known for over 15 years that the use of the PM$_{2.5}$ and PM$_{10}$ fugitive-dust emissions as given in inventories (e.g., from ploughing and harvesting on farms, construction activities such as excavation and grading, road dust emissions from paved and unpaved roads) will result in regional-scale AQ models making PM overpredictions. The reason is that regional-scale AQ models have no way to account for the fugitive dust emissions that are removed from the atmosphere within a few hundred meters of being emitted due to settling or impaction on nearby obstacles. When the SET0 emissions file set was generated, in order to account for the impact of near-source subgrid-scale settling and removal a simple uniform transportable-fraction (TF) field with a value of 0.25 was applied as a SMOKE post-processing step to scale Canadian fugitive dust emissions downwards. This calculation has the effect of reducing PM$_{2.5}$ and PM$_{10}$ emissions in the Canadian national CAC inventory from a limited number of source sectors (e.g., paved and unpaved road dust, construction, agricultural activities) by 75% and was suggested as a first-order correction by Possiel et al. (2001). To generate the SET2.1.1 emissions file set, however, this simple uniform scaling factor was replaced by a land-use-dependent TF field similar to the one used to process U.S. emissions inventories (see Pace, 2005). Land-use-dependent scaling factors range in value from 0.0 for forests to 1.0 for deserts and other barren areas and are intended to account for impaction removal on vegetation. Again, PM$_{2.5}$ and PM$_{10}$ emissions from this same limited set of source sectors were reduced, but now by differing amounts at different locations depending on the amount and type of vegetation cover.

Figure 4a shows the spatial distribution of the land-use-dependent TF field in both Canada and the U.S. Note the large difference between eastern Canada, which is largely forested, and the southern portion of the Prairie provinces, which has few trees. Figure 4b shows the difference field for the new vs. old TF fields. The upshot is that fugitive dust emissions will be reduced to lower values using the new TF field over most of southern Canada with the exception of the southern portion of the Prairie provinces. On the other hand, over North America as a whole the new land-use-dependent TF field reduces fugitive dust emissions by a smaller amount than the 75% reduction associated with the simple uniform 0.25 TF field. Pace (2005) determined for the
U.S. that the average reduction for all U.S. counties was roughly 49%. For the TF field plotted in Figure 4b, for Canada, the average TF value was 47% and for the U.S. it was 52%.

Figure 4: Plots of (a) the new land-use-dependent transportable fraction (TF) field and (b) the difference field (new minus old) between the new TF field and the previous TF field with a uniform value of 0.25 on the RAQDPS15 grid.

3.5 New temporal profiles for Canadian sources

Facilities reporting to the NPRI are required to submit a description of their operating schedule. This information from the 2010 NPRI was used to create a set of new temporal profiles for all 3,384 Canadian facilities (i.e., point sources) that reported to the NPRI.

For other source types, a number of new temporal profiles were also created for on-road sources (see Zhang et al., 2012), agriculture, construction, commercial marine vessels, fugitive dust from paved and unpaved roads, residential wood combustion, and residential and commercial meat cooking.

3.6 Updated VOC and PM speciation profiles

A total of 45 additional VOC speciation profiles were added to the profile library and then used by SMOKE. As well a new set of 84 detailed PM speciation profiles described by Reff et al. (2009) was recast in terms of the six PM chemical components considered by GEM-MACH that are emitted, and these new profiles were used for PM speciation by SMOKE. Note that Reff et al.’s detailed PM speciation profiles are also contained in the U.S. EPA SPECIATE4.3 profile library (see http://www.epa.gov/ttn/chief/software/speciate/index.html).
3.7 Post-2010 Canadian facility closures and openings

Emissions associated with the following five large industrial facilities that ceased operation during 2010 or afterwards were removed from the 2010 Canadian point-source emissions file (see Section 2.2): (a) Hudson Bay Mining & Smelting non-ferrous smelter, Flin Flon, MB; (b) Glencore Canada Corporation Kidd Creek metallurgical facility, Timmins, ON; (c) Shell Canada Products Ltd. Montreal East petrochemical refinery, Montreal, QC; (d) New Brunswick Power Grand Lake Generating Station, Grand Lake, NB; and (e) Ontario Power Generation Nanticoke Generating Station, Nanticoke, ON. At the same time, emissions for one newly opened facility, the Imperial Oil Kearl Lake oil sands mine in the Lower Athabasca region of northeastern Alberta, were estimated and inserted into the 2010 Canadian point-source emissions file (Zhang et al., 2015).

3.8 Other improvements

The spatial surrogate fields discussed in Sections 3.2 and 3.3 are constructed using Geographic Information System (GIS) boundary shapefiles and data shapefiles. In the course of developing new shapefiles for constructing these new surrogates, it was found that some of the Canadian and U.S. shapefiles that were being used had topological errors. The two main errors of concern were gaps in areal coverage and overlaps in areal coverage. Spatial gaps in shapefile coverage may cause misallocation of emissions because it is not possible to allocate emissions to the gap areas. On the other hand, overlapping polygons in the shapefiles may cause misallocation because of areal “double-counting”, that is, emissions will be over-allocated to overlap areas. The ArcGIS software package was used to identify and then correct these topological errors.

Another GIS problem concerned the use of different datums by different shapefiles. (A datum is a mathematical model that describes the size and shape of the reference ellipsoid and the origin and orientation of coordinate system; that is, it is a reference mapping surface.). For the most accurate representation of the Earth most shapefile datums use a reference ellipsoid or spheroid, since the Earth is not a true sphere but rather a “flattened” sphere at the poles with an equatorial “bulge”. However, numerical weather prediction models and AQ models approximate the Earth as a sphere. For consistency with these models and to avoid working with two or more shapefiles using different datums, tools in the Spatial Allocator package developed by the Institute for the Environment at the University of North Carolina at Chapel Hill (https://www.cmascenter.org/sa-tools/) were used to convert all shapefiles used for surrogate generation to a spherical datum.

One other problem concerned incorrect time zone assignments. A few North American jurisdictions at both the provincial and subprovincial level in Canada and at the state and county level in the U.S. do not observe Daylight Saving Time (DST). This is an issue because those times of year when DST is observed must be considered and accounted for when performing temporal allocation. Corrections for DST for two Canadian provinces, Saskatchewan and Quebec, were made to the SMOKE ancillary file “costcy.txt”.

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3.9 Mass-conserving interpolation

When the SET0 emissions file set was developed for version 4 of the RAQDPS system (see Table 1 or 2), all Canadian, U.S., and Mexican spatial surrogates were first generated for the RAQDPS15 grid and then the resulting 15-km gridded emissions fields were interpolated using mass-conserving interpolation to the 10-km RAQDPS10 grid. The extra interpolation step means that in effect some spatial smoothing was applied to the 10-km emissions fields to avoid overly sharp horizontal gradients in urban centres. This extra step was necessary because the direct use of gridded spatial surrogate fields developed for the RAQDPS 10-km grid was found to result in a deterioration in model performance relative to the use of 15-km spatial surrogates. The reason for this deterioration may be that some of the spatial surrogates that are being employed to allocate emissions to the model grid have a scale dependence at smaller scales that is not currently being represented properly.

One example is the spatial allocation of emissions from traffic on local roads. When the SET0 emissions file set was generated, a combined population-density/road-network spatial surrogate was used. However, it is likely that at finer spatial scales the geometry of the local road network may be independent of the population distribution pattern in the case of heavily populated areas. For this reason, as noted in Section 3.2 a new spatial surrogate, capped population density, was used for SET2.1.1 to allocate Canadian on-road emissions from local traffic. However, even when those 24 new or revised Canadian spatial surrogates (Table 4) were used by SMOKE, RAQDPS sensitivity runs again determined that gridded input emission fields generated using 15-km spatial surrogates out-performed those generated using 10-km spatial surrogates. As a result, the SET2.1.1 emissions file set was also generated for a 15-km grid and then interpolated to the RAQDPS 10-km operational grid. Note that the reason for this difference in model performance associated with intrinsic emissions spatial resolution continues to be investigated.

4. Differences Between SET0 and SET2.1.1 Emissions File Sets

Table 3 summarized the total differences in emissions between the annual Canadian and U.S. national emissions inventories that the SET0 and SET2.1.1 emissions file sets were based on. Section 3 described differences in the emissions processing methodology that was used to generate the two emissions file sets. This section provides some graphical analyses to show emissions patterns and differences between the SET0 and SET2.1.1 gridded emissions.

Figures 5 and 6 present spatial patterns of mean monthly emissions of six gas-phase species and six particle-phase species, respectively, for either January or July. It should be noted that the choice of contour intervals in these plots was percentile-based. Linearly-spaced contour intervals are good for showing broad regional emissions patterns but they cannot show spatial variations in regions of very high or very low emissions. Logarithmically-spaced contour intervals are often used to plot regional emissions patterns because they are better able to show spatial variations...
across the full dynamic range of the emissions. However, different sets of contour intervals may need to be chosen for different species and time periods and it is challenging to generate these interval sets automatically.

In the percentile-based approach, an emissions distribution is calculated for each emitted model species across all grid cells and time periods of interest and then contour intervals are chosen based on the values of different percentiles. In preparing Figures 5 and 6, mean monthly emissions fields were first calculated for each of 12 months for each species. Given the RAQDPS10 grid size of 528 x 708, for 12 months the emissions distributions for each model species will then be based on 4,485,888 values. Thus, even for the 99.99th percentile there will still be nearly 450 emissions values above that percentile in the full emissions file set for each model species. Because large portions of the RAQDPS10 grid are located over ocean waters or remote northern land areas, each emissions distribution includes a very large number of either zero or extremely small emissions values. To account for this very skewed distribution, only two contour intervals were assigned below the median or 50th percentile value while 10 contour intervals were used above the median emissions value.

The spatial distributions of the SET2.1.1 emissions shown in Fig. 5 for six selected model gas-phase species – SO₂, NO, CO, higher alkanes, toluene, and NH₃ – are broadly similar overall. Emission magnitudes are higher over eastern North America than over western North America and are higher over the southern half of North America than the northern half. Emissions associated with large population centres and with the network of primary highways are visible in the panels as are off-shore emissions associated with ocean-going vessels and oil-and-gas extraction. Differences are also visible between the spatial emissions patterns for different species. For example, ocean-going vessels are relatively large emitters of SO₂ compared to their emissions of other species, and a higher number of above-median SO₂ emissions are evident off-shore in Fig. 5a as compared to the other panels. Ammonia (NH₃) emissions are dominated by the agricultural sector (i.e., emissions from animal husbandry and fertilizer application), and the NH₃ emissions pattern shown in Fig. 5f is qualitatively different from the other five panels and can be seen to have local maxima over agricultural regions in the midwestern states of the U.S.

Figure 6 shows the spatial distributions of SET2.1.1 mean July emissions of the six PM₂.₅ chemical components considered by GEM-MACH: (1) ammonium [AM]; (2) crustal material [CM]; (3) elemental carbon [EC]; (4) nitrate [NT]; (5) primary organic matter [POM]; and (6) sulphate [SU]. These patterns are not at all the same because PM₂.₅ emissions from different types of sources have different chemical compositions. For example, the two largest chemical components of PM₂.₅ emissions for on-road light duty diesel vehicles (LDDV) are EC (51%) and POM (44%), for on-road heavy duty diesel vehicles (HDDV) they are also EC (77%) and POM (22%), but for fugitive dust emissions from paved roads they are CM (85%) and POM (14%) (e.g., Reff et al., 2009).
Figure 5: Spatial distributions on the RAQDPS 10-km grid of mean January emissions (tonnes/month/grid cell) of (a) SO$_2$, (b) NO, and (c) CO and July emissions of (d) higher alkanes [A3], (e) toluene [TOL], and (f) NH$_3$ from all anthropogenic source types for the new SET2.1.1 emissions file set. Note that the contour intervals for each species are based on the percentile distribution of mean monthly emissions across all grid cells and months: for each species the upper levels of the contour intervals correspond to the 35$^{th}$, 50$^{th}$, 65$^{th}$, 75$^{th}$, 85$^{th}$, 90$^{th}$, 95$^{th}$, 97.5$^{th}$, 99.5$^{th}$, 99.75$^{th}$, 99.9$^{th}$, and 99.99$^{th}$ percentiles, respectively.
Figure 6: Same as Fig. 5 but for mean July emissions (tonnes/month/grid cell) of the
(a) ammonium [AM1], (b) crustal-material [CM1], (c) elemental-carbon [EC1], (d) nitrate [NT1],
(e) primary-organic-matter [PC1], and (f) sulphate [SU1] chemical components of PM$_{2.5}$ from all
anthropogenic source types for the new SET2.1.1 emissions file set.
Inspection of the values of the median contour interval for the six PM$_{2.5}$ species shown in Fig. 6 suggests that CM has the largest contribution overall, followed by POM, EC, and SU. AM and NT make only a minor contribution to PM$_{2.5}$ emissions, but these two species along with SU have large secondary sources whereas CM, POM, and EC have only primary sources. As in Fig. 5, emission magnitudes of these PM$_{2.5}$ components are higher over eastern North America than over western North America and are higher over the southern half of North America than the northern half, and large population centres, primary highways, and shipping lanes are visible in some of the panels. For example, highways are clearly visible in Fig. 6c, consistent with the large contribution from on-road mobile sources to EC emissions.

Figure 7 shows difference plots between SET2.1.1 and SET0 monthly total emissions for six inventory species. As shown in Table 3, emissions of SO$_2$ decreased for Canada between 2006 and 2010 and for the U.S. between the projected 2012 values and 2011, and this overall decrease is reflected in Fig. 7a by the large number of grid cells showing lower emissions for SET2.1.1 vs. SET0. Note that these decreases tend to be restricted to individual grid cells (i.e., local) because total SO$_2$ emissions are dominated by emissions from point sources. CO emissions were also somewhat lower in SET2.1.1 in most of North America (Fig. 7c), consistent with the overall decreases shown in Table 3 in both Canada and the U.S. However, Table 3 also shows NH$_3$ emissions decreasing in both countries, but the difference pattern shown in Fig. 7e is more complex, with increases in some areas and decreases in others. From Table 3, NO$_x$ emissions, on the other hand, are lower in Canada for SET2.1.1 but higher in the U.S., and Fig. 7b is consistent with this result, with decreases visible across most of Canada but increases visible across much of the U.S. According to Table 3, VOC emissions are lower in Canada for SET2.1.1 but almost unchanged in the U.S.; however, Fig. 7d shows decreases across Canada but increases as well as decreases across much of the U.S. Lastly, Fig. 7f shows the difference plot for PM$_{2.5}$ emissions. As discussed in Sections 2 and 3, PM$_{2.5}$ emissions underwent some complicated changes between the two emissions sets due to a number of factors, including new inventories and a change to the TF field. The increase in PM$_{2.5}$ emissions seen in southern Alberta is likely due to the increase in TF field values in this area discussed in Section 3.4.

One quality assurance step was to examine the temporal and spatial allocation of emissions to a set of nine large Canadian cities, since predicted concentrations for these cities are strongly influenced by local emissions and an overallocation of emissions to these cities can result in overpredictions of primary species such as NO$_2$ and PM$_{2.5}$. Because the RAQDPS system with SET0 emissions generally did not suffer from frequent large overpredictions, average and peak monthly NO$_2$ and PM$_{2.5}$ emissions for SET0 for these cities were compared with corresponding SET2.1.1 values for January and July. The two sets of values were roughly comparable (not shown).
Figure 7: Difference between January emissions (tonnes/month/grid cell) of (a) SO$_2$, (b) NO$_x$, and (c) CO and July emissions of (d) VOC, (e) NH$_3$, and (f) PM$_{2.5}$ from all anthropogenic source types for new SET2.1.1 emissions file vs. operational SET0 emissions file set. A warm colour means that SET2.1.1 emissions are larger, a cool colour means that the SET0 emissions are larger, and the white colour indicates very little or no change.
Figure 8: Plots of January emissions (tonnes/month/grid cell) for two species and six emissions source types “zoomed” over the Montreal region on an experimental 10-km “Yin” grid after processing by SMOKE using the SET2.1.1 methodology: (a) NO\textsubscript{2} area emissions; (b) NO\textsubscript{2} land off-road emissions; (c) NO\textsubscript{2} ALM off-road emissions; (d) NO\textsubscript{2} on-road emissions; (e) NO\textsubscript{2} major-point emissions; (f) NO\textsubscript{2} minor-point emissions; (g) PM\textsubscript{2.5} area emissions; (h) PM\textsubscript{2.5} land off-road emissions; (i) PM\textsubscript{2.5} ALM off-road emissions; (j) PM\textsubscript{2.5} on-road emissions; (k) PM\textsubscript{2.5} major-point emissions; and (l) PM\textsubscript{2.5} minor-point emissions. Note that the same set of contour intervals was used for all 12 panels. The order of the plots is from left to right and from top to bottom.

The spatial distribution of emissions over these same nine urban areas was also examined by plotting gridded emissions for six primary source types: area; land off-road;
aircraft/locomotive/marine (ALM) off-road; on-road; major points; and minor points. This qualitative comparison allowed the reasonableness of the spatial distributions to be examined and the relative importance of the different source types to be viewed within each city and between cities. Figure 8 shows an example for the Montreal region for emissions of two primary pollutants, NO\textsubscript{2} and PM\textsubscript{2.5}. For January NO\textsubscript{2} emissions, it appears that area sources, off-road land sources, and on-road sources are all important in the Montreal region and the locations of a small number of major point sources can be seen. For January PM\textsubscript{2.5} emissions, on the other hand, area sources are clearly dominant with off-road land sources a relatively distant second. RWC emissions are the largest contributor to the area emissions inventory, and in July when RWC emissions are expected to be small, PM\textsubscript{2.5} area emissions for the Montreal area are much lower (not shown).

5. Results of Performance Evaluations

5.1 Objective evaluation

The objective evaluation of the RAQDPS013-based AQ forecast system using the new SET2.1.1 emissions file set and comparison with RAQDPS012 using the SET0 emissions file set was performed using near-real-time hourly observations of NO\textsubscript{2}, O\textsubscript{3} and PM\textsubscript{2.5} surface concentrations for Canada and the contiguous U.S. and model-predicted NO\textsubscript{2}, O\textsubscript{3} and PM\textsubscript{2.5} surface concentrations. Three time periods were examined: winter and summer 2014 (1 January–14 February and 1 July–14 August, respectively) and spring 2015 (24 March – 4 June). Figure 9 shows the locations of the Canadian and U.S. stations measuring hourly NO\textsubscript{2}, O\textsubscript{3} and PM\textsubscript{2.5} surface concentrations and reporting those measurements in near-real time in summer 2014.

Tables 5 to 7 present a set of objective scores for the hourly forecasts for these three periods for the RAQDPS013 and RAQDPS012 simulations. Values of three statistics – mean bias (MB), Pearson correlation coefficient (R), and unbiased root mean square error (URMSE) – were calculated for each period for the full model domain plus six different regions of the model domain (see Fig. 10). The relative model performance of RAQDPS013 with SET2.1.1 emissions vs. RAQDPS012 with SET0 emissions is indicated in these tables by colour coding, where the statistical significance of the differences in each statistic was determined by a bootstrap calculation for a confidence interval of 95%.

The statistics for the RAQDPS013 system vs. the RAQDPS012 system show an overall improvement in the wintertime and springtime forecasts for both Canada and the contiguous U.S. (Tables 5 and 7). The statistical comparison for the summertime forecasts (Table 6), on the other hand, shows mixed results with neither model version outperforming the other. Interestingly, while there is a stronger negative mean bias for the RAQDPS013 system for predicted PM\textsubscript{2.5} (i.e., increased underprediction) for most regions for all three seasons, the URMSE scores for PM\textsubscript{2.5}
Figure 9: Locations of air quality measurement stations whose observations were used to calculate objective scores for summer 2014: (a) NO₂ stations (161 in Canada, 102 in U.S.); (b) O₃ stations (204 in Canada, 1,097 in U.S.); and (c) PM₂.₅ stations (200 in Canada, 579 in U.S.).

for the new system are a marked improvement in both countries in all three seasons and especially in the summer.

Tables 8 to 10 present comparable statistics to Tables 5 to 7, but this time for the daily maximum concentrations for the same periods. Note that tests of statistical significance by means of bootstrapping were not performed for these results. A significant improvement in forecast maxima is observed for PM₂.₅, O₃, and NO₂ for the winter period (Table 8). For the two other
seasons (Tables 9 and 10), the results are more mixed, with slightly better scores for the RAQDPS013 forecast.

It is also of interest to examine model skill in forecasting extreme values, that is, values greater than specified thresholds. Figures 11 to 13 show categorical scores over Canada (left side) and the contiguous U.S (right side) for NO$_2$, O$_3$, and PM$_{2.5}$. The four categorical statistics presented are the percent correct (PC), the probability of detection (POD), the false alarm ratio (FAR), and the critical success index (CSI) (e.g., Mason, 2003). The three concentration thresholds considered were 30 ppbv for NO$_2$, 65 ppbv for O$_3$, and 30 μg m$^{-3}$ for PM$_{2.5}$. In general, we note that the categorical scores for both systems are similar.

The UMOS-AQ post-processing package uses the RAQDPS forecasts together with hourly AQ surface measurements to generate point-specific statistical AQ forecasts for larger Canadian urban centers (e.g., Moran et al., 2012). As part of the objective evaluation of the new system, UMOS-AQ was tested with RAQDPS013 for both cold and warm seasons. It was found that the results were very similar to those obtained with RAQDPS012, and that overall the two systems are statistically equivalent. Certain pollutants at certain stations did produce lower scores with the new system; however, further improvements may be expected as the UMOS-AQ statistics become more robust with the accumulation of more cases from RAQDPS013.

5.2 Discussion

In order to understand some of the reasons for the differences in the objective scores for the forecasts made by the two model versions, it is useful to examine plots of the spatial distributions of mean seasonal differences in surface concentrations for the three AQHI species, NO$_2$, O$_3$, and PM$_{2.5}$. Figures 14, 15, and 16 show mean concentration patterns for the RAQDPS012 simulations for winter 2014, summer 2014, and spring 2015, respectively, together with the corresponding mean concentration difference fields between the two model versions (calculated as RAQDPS013 minus RAQDPS012). It is also useful to compare the spatial distributions of changes in pollutant emissions shown in Fig. 7 to the spatial distributions of differences in NO$_2$, O$_3$, and PM$_{2.5}$ concentrations shown in the right columns of Figs. 14, 15, and 16 because the only difference between the two model versions are the differences in emissions.

Given that NO$_2$ is a primary pollutant (i.e., it is directly emitted), NO$_2$ concentrations would be expected to be directly impacted by changes in NO$_x$ emissions. From Table 3 we know that NO$_x$ emissions are 5% higher overall in the SET2.1.1 emissions file set, but this net increase is due to the combination of lower NO$_x$ emissions in Canada and higher NO$_x$ emissions in the U.S. Figure 7b shows the difference in the annual NO$_x$ emissions fields between the SET2.1.1 emissions (RAQDPS013) and the SET0 emissions (RAQDPS012). The difference pattern is complicated. While NO$_x$ emissions are lower in general over Canada, over the U.S. increases and decreases in NO$_x$ emissions are intermingled. This intermingling can be explained by a more
Table 5: Hourly performance statistics for NO$_2$, O$_3$, and PM$_{2.5}$ for winter 2014 (1 January – 14 February; 45 days) for RAQDPS013 and RAQDPS012 48-hour forecasts. The three statistics considered for each of the three species and seven analysis regions were mean bias (MB), Pearson correlation coefficient (R), and unbiased root mean square error (URMSE). Pink-coloured boxes indicate a better forecast with the RAQDPS013, blue boxes indicate a better forecast with RAQDPS012, and yellow boxes indicate no statistically significant difference between the two model versions. 60% of the boxes are pink, 25% are blue, and 11% are yellow.

Table 6: Same as Table 5 but for summer 2014 (1 July – 14 August; 45 days). 41% of the boxes are pink, 40% are blue, and 19% are yellow.
Table 7: Same as Table 5 but for spring 2015 (24 March – 4 June; 73 days). 48% of the boxes are pink, 35% are blue, and 17% are yellow.

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Table 8: Same as Table 5 but for daily maximum error statistics for winter 2014 (1 January – 14 February; 45 days) for RAQDPS013 and RAQDPS012 forecasts. Pink-coloured boxes again indicate a better forecast with the RAQDPS013, blue boxes indicate a better forecast with RAQDPS012; no estimates of statistical significance have been made for this comparison. 70% of the boxes are pink and 30% are blue.

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Table 9: Same as Table 8 but for summer 2014 (1 July – 14 August; 45 days). 56% of the boxes are pink and 44% are blue.

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<td>4.44</td>
<td>5.11</td>
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<tr>
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<td>0.04</td>
<td>0.09</td>
<td>0.01</td>
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<td>17.53</td>
<td>25.46</td>
<td>23.39</td>
<td>28.60</td>
<td>24.10</td>
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</table>

Table 10: Same as Table 8 but for spring 2015 (24 March – 4 June; 73 days). 52% of the boxes are pink and 48% are blue.

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<th>Pollutant</th>
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<th>ECAN</th>
<th>USA</th>
<th>WUSA</th>
<th>EUSA</th>
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<td>0.58</td>
<td>0.59</td>
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<tr>
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<td>10.60</td>
<td>10.69</td>
<td>10.16</td>
<td>10.76</td>
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<tr>
<td>O₃</td>
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<td>-7.06</td>
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<tr>
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Figure 10: Locations of four of the analysis regions considered in Tables 5 to 10: (1) western Canada; (2) eastern Canada; (3) western U.S.; and (4) eastern U.S. The other three analysis regions considered are (5) all of Canada, (6) all of U.S., and (7) North America. The filled green circles denote the locations of National Air Pollution Surveillance (NAPS) network NO$_2$ measurement stations in Canada.

Detailed analysis by source type (not shown) of the differences in U.S. NO$_x$ emissions, which revealed that NO$_x$ emissions from point sources such as power plants and large industrial facilities decreased whereas NO$_x$ emissions from on-road mobile sources, off-road sources, and area sources all increased. One interesting feature is the general increase in off-shore NO$_x$ emissions over the waters of the northern Gulf of Mexico.

The NO$_2$ concentration difference plots shown in Figs. 14b, 15b, and 16b are complex but are clearly linked to the NO$_x$ emissions changes. Over Canada NO$_2$ concentrations have generally decreased for RAQDPS013, with the notable exception of the industrial Athabasca oil sands region of northeastern Alberta where NO$_x$ emissions increased between 2006 and 2010. Note
Figure 11: Categorical scores for winter 2014 (1 Jan.–14 Feb.) for NO$_2$ (upper row) and PM$_{2.5}$ (lower row), respectively, for RAQDPS012 forecasts (blue bars) and RAQDPS013 forecasts (red bars). The left-hand column contains scores for Canada while the right-hand column contains scores for the contiguous U.S. Note that categorical scores are not shown for O$_3$ because wintertime O$_3$ levels are typically below the selected threshold.

that a positive trend in NO$_x$ emissions in the Athabasca region has also been detected by satellite (McLinden et al., 2014). Over the U.S., increases and decreases in NO$_2$ concentration are intermingled, similar to the NOx emissions. Over the Ohio Valley region of the eastern U.S., reductions in NO$_x$ emissions from power plants have resulted in decreases in NO$_2$ concentrations, whereas over the northern Gulf of Mexico, increases in NO$_x$ emissions have resulted in increases in NO$_2$ concentrations.
Figure 12: Categorical scores for summer 2014 (1 July–14 Aug.) for NO$_2$ (upper row), O$_3$ (middle row), and PM$_{2.5}$ (lower row). The left-hand column contains scores for Canada while the right-hand column contains scores for the contiguous U.S.
Figure 13: Same as Figure 12 but for spring 2015 (24 Mar. – 4 June).
Figure 14: Left column (panels a, c, e) shows mean NO₂, O₃, and PM₂.₅ RAQDPS012 forecast fields for the winter 2014 period (1 Jan. – 14 Feb.; 45 days). The right column (panels b, d, f) shows the corresponding difference fields between RAQDPS013 and RAQDPS012 (i.e., version 013 minus version 012) for the same period. The units for NO₂ and O₃ are ppbv while the units for PM₂.₅ are μg m⁻³.
Figure 15: Same as Fig. 14 but for a summer 2014 period (1 July – 14 August; 45 days).
As a secondary pollutant, O₃ depends upon atmospheric chemistry and meteorology as well as emissions, and from Figs. 14d, 15d, and 16d its response can be seen to be seasonally dependent. In winter O₃ concentrations have increased on average over much of the domain whereas in spring and summer the opposite is true. The wintertime increases in O₃ are likely associated with
the decreases in NO\textsubscript{2} (and NO) concentrations due to reduced titration by NO. On the other hand, in the spring and summer when photochemical production of O\textsubscript{3} associated with NO\textsubscript{x} emissions will be more important, those areas with reduced NO\textsubscript{2} (and NO) concentrations such as Canada and the eastern and western U.S. will have less photochemical production of O\textsubscript{3} and hence lower O\textsubscript{3} concentrations whereas those areas with increased NO\textsubscript{2} and NO concentrations such as the central U.S. will have higher O\textsubscript{3} concentrations.

Because PM\textsubscript{2.5} is both a primary and secondary pollutant, its response to emissions changes can be very complex. It can be seen from Figs. 14f, 15f, and 16f that PM\textsubscript{2.5} concentrations for RAQDPS013 have decreased over eastern North America in all three seasons but have increased over the Canadian provinces of Alberta and Saskatchewan. Referring to Table 3 and Fig. 7f, we know that primary PM\textsubscript{2.5} emissions increased over Canada, in particular the province of Alberta, but decreased in the U.S. On the other hand, emissions of SO\textsubscript{2} decreased by 35% in Canada and by 48% in the contiguous U.S., which is relevant because SO\textsubscript{2} is an important PM\textsubscript{2.5} precursor due to its conversion in the atmosphere to particle sulphate by both gas-phase and aqueous-phase oxidation. The combination of these emissions changes can explain the predicted PM\textsubscript{2.5} concentration changes. The increase in emissions of primary PM\textsubscript{2.5} and of ammonia, another PM\textsubscript{2.5} precursor, in Alberta (see Figs. 7f and 7e, respectively) would increase PM\textsubscript{2.5} concentrations in this region while the reductions in SO\textsubscript{2} emissions over eastern North America would reduce both particle sulphate and PM\textsubscript{2.5} concentrations over this region due to both reduced sulphate production in SO\textsubscript{2} source regions and reduced transport to surrounding regions such as eastern Canada.

It is worth noting one special case, the reduction in PM\textsubscript{2.5} concentrations seen over the San Joaquin Valley of California in winter 2014 (see Fig. 14f). This reduction is likely driven at least in part by reductions in NO\textsubscript{x} emissions in this area (cf. Figs. 7b and 14b), since NO\textsubscript{x} is another PM\textsubscript{2.5} precursor due to its atmospheric conversion to particle nitrate, and particle nitrate is known to be an important component of PM\textsubscript{2.5} in this area in the wintertime (e.g., Pun et al., 2009).

It is also possible to look in more detail at the impact of the regional changes in PM\textsubscript{2.5} concentrations shown in Figs. 14f, 15f, and 16f on the regional objective scores presented in Tables 5 to 7. Figure 17a shows a plot of PM\textsubscript{2.5} mean bias values at individual PM\textsubscript{2.5} measurement stations for the summer 2014 period for the RAQDPS012 simulation. It is clear from this plot that the RAQDPS012 tends to underpredict PM\textsubscript{2.5} concentrations in western North America and to overpredict them in eastern North America. From Table 6 we see that the MB values for PM\textsubscript{2.5} for western Canada and the western U.S. were -6.3 and -2.7 ug m\textsuperscript{-3} while for eastern Canada and the eastern U.S. the corresponding values were 0.5 and 4.3 ug m\textsuperscript{-3}. Figure 17b shows the corresponding differences in MB values at the same stations between RAQDPS013 and RAQDPS012. In western North America the changes are small whereas in eastern North America the differences are generally negative; a negative difference value means
that the MB value has decreased for the RAQDPS013 simulation, which is welcome for eastern North America since the RAQDPS012 had positive MB values in this region. This is consistent with Table 6, since for the RAQDPS013 simulation, the MB values for PM$_{2.5}$ for western Canada and the western U.S. were -5.9 and -3.0 ug m$^{-3}$ while for eastern Canada and the eastern U.S. the corresponding values decreased to -1.6 and -1.3 ug m$^{-3}$.

Figure 17 : Average per-station MB values for RAQDPS012 (left side) and differences in average per-station MB values for the two simulations (RAQDPS013-RAQDPS012) (right side) for summer of 2014 for PM$_{2.5}$.

It is evident from Tables 8 to 10 that MB values for daily maximum PM$_{2.5}$ concentration forecasts also decreased in general for all three seasons for the RAQDPS013 simulation. The largest decreases occurred for summer 2014, with a domain-wide decrease of 5.4 ug m$^{-3}$ and a maximum regional decrease of 10.0 ug m$^{-3}$ for the eastern U.S. To examine this reduction further, Figure 18 shows plots of the maximum hourly PM$_{2.5}$ concentration values forecast at each model grid cell during the entire 75-day summer 2014 period by the RAQDPS012 and RAQDPS013 simulations. It is clear that the PM$_{2.5}$ hourly concentration maxima are considerably smaller for the RAQDPS013 simulation. Note that the concentration range plotted in this figure goes to 150 ug m$^{-3}$ whereas in Figure 15e, which shows the PM$_{2.5}$ mean hourly concentration field for the same period, the concentration range plotted only goes to 29 ug m$^{-3}$. 
Figure 18:  Maximum hourly PM$_{2.5}$ concentration forecasted at each model grid cell for the 45-day summer 2014 period by RAQDPS012 (left) and RAQDPS013 (right).

In addition to the general reduction in the magnitudes of PM$_{2.5}$ concentrations for the RAQDPS013 simulations, it was also noted in Section 5.1 that unbiased RMSE (URMSE) values were generally much improved (i.e., reduced) for the RAQDPS013 for all three seasons. Figure 19 shows how this reduction varied day by day by 48-hour forecast for the spring 2015 period. The RAQDPS013 simulation has consistently lower domain-average URMSE values, but the magnitude of the difference varies by day and period. There are weeks when there is little difference and weeks when the difference is consistently large, suggesting that synoptic variations in meteorological regime may be influencing these differences.

One other important aspect of RAQDPS performance is the quality of its predictions for large urban centres, since population exposure to air pollution is generally greatest for these high-emissions (e.g., Fig. 8) and high-population-density areas. Past experience with the RAQDPS has shown that the use of standard emissions processing methodologies has resulted in the over-allocation of emissions to urban centres, with the result that urban air pollutant concentrations tend to be over-predicted (e.g., Moran et al., 2013). As a consequence it is important to examine RAQDPS predictions for large Canadian urban centres.
Figure 19: Time series of domain-average URMSE values for the RAQDPS012 (blue) and RAQDPS013 (black) simulations for the spring 2015 period. Each point represents the average 48-hour value over the RAQDPS domain for each of the two forecasts per day.

Figures 21, 22, and 23 show examples of RAQDPS012 and RAQDPS013 time series of hourly NO\textsubscript{2}, O\textsubscript{3}, and PM\textsubscript{2.5} concentrations vs. observations for spring 2015 for three Canadian cities: Montreal, Edmonton, and Halifax. Model performance displays a range of behaviour. For Montreal (Fig. 21) the RAQDPS012 and RAQDPS013 time series are quite similar, but where they differ, the RAQDPS013 time series tends to be closer to the observations. For Edmonton (Fig. 22) the same comment can be made for NO\textsubscript{2} and O\textsubscript{3} but the PM\textsubscript{2.5} time series predicted by the RAQDPS013 has more spurious peaks than the RAQDPS012 time series. And for Halifax (Fig. 22) the RAQDPS013 time series are markedly better than the RAQDPS012 time series for all three pollutants. Current practice is to monitor RAQDPS performance for 13 major Canadian cities: Vancouver, Yellowknife, Calgary, Edmonton, Saskatoon, Winnipeg, Hamilton, Toronto, Ottawa, Montreal, Quebec City, Moncton, and Halifax. For the spring 2015 period, overall RAQDPS013 performance was better than or equivalent to the RAQDPS012 performance for all of these cities except Calgary and Edmonton.
Figure 20: Comparison of time series of observed (green), RAQDPS012-predicted (blue), and RAQDPS013-predicted (red) hourly concentrations of (left) NO\textsubscript{2}, (middle) O\textsubscript{3}, and (right) PM\textsubscript{2.5} for Montreal, Quebec for the period 2 March to 1 June 2015.
Figure 21: Same as Fig. 20 but for Edmonton, Alberta.
Figure 22: Same as Fig. 20 but for Halifax, Nova Scotia.
6. Summary and Future Work

RAQDPS013, a new version of Environment Canada’s operational Regional Air Quality Deterministic Prediction System, was implemented operationally on 11 June 2015. The only difference between this version and RAQDPS012, the previous operational version that had been in service since 18 March 2015, is the update made to the hourly, gridded emissions input data set that is used to provide chemical bottom boundary conditions. The new emissions data set is based on more recent Canadian and U.S. national emissions inventories and also benefited from improvements made to the emissions processing system that is used to transform emissions inventories into model-ready emissions files.

The previous RAQDPS emissions input data set was based on the 2006 Canadian national emissions inventory (version 1) and a projected 2012 U.S. emissions inventory projected from the 2005 U.S. national inventory (version 4). The new emissions input data set is based on the 2010 Canadian national emissions inventory (version 1) and the 2011 U.S. national emissions inventory (version 1). For the combined new Canadian and U.S. inventories versus the older inventories, SO₂ emissions decreased by 47%, NOₓ emissions increased by 5%, VOC emissions decreased by 1%, NH₃ emissions decreased by 5%, CO emissions decreased by 6%, PM₂.₅ emissions were nearly unchanged, and PM₁₀ emissions decreased by 2%.

Methodological improvements to the emissions processing performed to generate the new emissions files included the use of (a) 25 new or updated Canadian and five new U.S. spatial surrogate fields, (b) a land-use-dependent transportable fraction field in place of simple 0.25 factor to scale Canadian fugitive dust emissions to account for near-source capture, (c) new temporal profiles for all 3,384 Canadian major point sources and a number of area, on-road, and off-road sources, and (d) a new library of PM speciation profiles and additional VOC speciation profiles as well as (e) the removal of emissions from five major Canadian facilities that either closed or drastically reduced their emissions after 2010 and the addition of emissions from one new oil sands mine in northeastern Alberta that became operational after 2010.

RAQDPS013 was tested with the new emissions data set for winter and summer periods in 2014 and a spring period in 2015 for a total of 163 days. The results showed an improvement to the winter forecasts for NO₂, O₃, and PM₂.₅, the three components of the Air Quality Health Index. In some urban centres like Vancouver, wintertime PM₂.₅ forecasts were greatly improved due to reductions in episodic overpredictions. The summer forecasts showed a very slight improvement and/or neutral impact overall with the new emissions data set. However, the summertime results showed a substantial improvement to the PM₂.₅ forecasts in eastern Canada and the U.S., where episodic overpredictions had been a problem. The effect on other pollutant fields was mixed. The spring 2015 forecasts also showed an overall improvement, including a large reduction in URMSE values for PM₂.₅.
Going forward, the next release of the RAQDPS will likely include a major update to the model source code. As shown in Table 1, all RAQDPS versions to date have been built on version 3 of the GEM numerical weather prediction model. However, a new generation of the GEM model, version 4, has been available for several years and a GEMv4-based version of the RAQDPS source code has been developed and is now undergoing testing.

Acknowledgments

We would like to acknowledge the contributions to this project of a number of other Environment Canada personnel. Mourad Sassi of AQMAS created a SMOKE-ready version of the new 2010 Canadian emissions inventory. Hugo Landry of AQMAS worked on code updates, execution scripts, and delivery to the operations. Sylvain Ménard of AQMAS assisted with the preparation of the operational emissions files. Samuel Gilbert of AQMAS provided objective scores for numerous model runs. Management support was provided to this project by Didier Davignon and Sophie Cousineau of AQMAS and by Heather Morrison of ARQI.

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