ANALYSIS OF CURRENT AND PROJECTED AIR QUALITY LEVELS IN CANADA WITH AURAMS

Véronique S. Bouchet, Environment Canada, Dorval, Canada, and S. Cousineau, L.-P. Crevier, A. Duhamel, J. Racine, M. Sassi, and M. Moran

1. INTRODUCTION^{*}

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Due to the complexity of the photochemical leading to the formation svstem and accumulation of O3 and PM2.5 at the surface, three dimensional air quality models have become the tool of choice to project the atmospheric response to air pollutants emission changes. Model are conventionally used to investigate the potential impacts on air quality of proposed emission reductions by comparing to a simulation using emissions from a reference inventory, a simulation where emission levels have been reduced from that same reference inventory. In a slight departure from this convention, the paper presented here analyzes the results of simulations performed with emission inventories from two distinct years in a first attempt to examine transient trends rather than absolute impacts.

Two simulations of current (referring to 2002) and projected (referring to 2015) levels of ozone and PM_{2.5} were performed with Environment Canada multi-pollutant model AURAMS (A Unified Regional Air quality Modelling System) as part of the latest review of smog levels in Canada, Although an actual transient analysis would also have taken into account the changes in meteorology over the years spanning the two simulations, the study only focused on the evolution of the emission and both simulations were run for the 2002 meteorological year. Expected trends in the levels of O₃ and PM_{2.5} were derived by comparing the two simulations and analyzed with respect to the projected emission changes. The seasonal response of the chemical system was also investigated to interpret the differing behaviour between O₃ and PM_{2.5}.

The ultimate objective of such an analysis would be to compare the trends projected through modelling to trends in long-term observations of O_3 and PM_{2.5}; at this time, however, the scope of the paper is restricted to understanding the tendencies that can be derived from modelling.

2. AURAMS DESCRIPTION

Designed as a "one-atmosphere" system, AURAMS allows the study of interactions between nitrogen oxides (NO_x), volatile organic compounds (VOCs), ammonia (NH₃), ozone (O₃), and primary and secondary PM, and has been used to address a variety of interconnected tropospheric air pollution problems [*Makar et al.*, 2003; *McKeen et al.*, 2005, 2007; *Moran et al.*, 2008].

The system is composed of three major components: a meteorological driver, the Canadian forecast model (GEM), the Spare-Matrix Operating Kernel Emission processor (SMOKE) and the chemical transport model itself. AURAMS treats gas-phase species and particulate-matter (PM) formation and evolution with time, as well as their interactions through gaseous, aqueous and heterogeneous reactions. Forty-two gaseous species are included and up to eight chemical components are considered to contribute to PM composition, the latter currently comprising sulphate, nitrate, ammonium, black carbon, primary organic carbon, secondary organic carbon, crustal material and sea salt. These PM chemical components are assumed to be internally mixed in each of the 12 size bins of the PM sectional size distribution, which ranges from 0.01 to 40.96 µm in diameter. For scenario applications, AURAMS is typically run on a continental domain, comprising Canada and the contiguous United-States, for an entire year using a 42 km spatial resolution and a 15 min timestep. Annual simulations are performed in 3 segments spanning periods of 4 to 5 months which overlap each other to allow for sufficient spin-up time between consecutive segments.

The version of AURAMS used in the scenario described here was evaluated for the year 2002. Results of the performance evaluation show that AURAMS reproduces well the spatial distribution of O_3 and $PM_{2.5}$ throughout the year but exhibits a tendency to underpredict PM_{2.5} levels that is exacerbated in winter (Moran et al., 2007; 2008 smoq assessment [in preparation]). А comparison with CMAQ results for a common period in 2002 showed that the performances of the two models were comparable [Smyth et al, in press]. Although a similar tendency to underpredict PM_{2.5} can be expected in projected AURAMS simulations, the trend analysis performed here focuses on the differences

^{*} *Corresponding author address*: Véronique S. Bouchet, Environment Canada, Meteorological Service of Canada, Dorval, Canada, 514-421-5020; e-mail: <u>Veronique.bouchet@ec.gc.ca</u>

between the two simulations and any influence from the low bias should be minimized.

3. EMISSION ANALYSIS

For the purpose of this study, Canadian and American national emission inventories corresponding to 2002 for the present day simulation (which will be referred to as the "current case" in the rest of the document), and 2015 for the future day simulations ("future case"), were used. The 2015 emission levels are projections to 2015 of the 2000/2001 Canadian and U.S. inventories assuming that emission levels will evolve in response to changes in economic activity and population growth within the bounds of existing and/or already proclaimed emission legislations. More specifically, the 2015 U.S. projected emission levels include the reduction imposed by the NOx SIP call and the Clean Air Interstate Rule (CAIR), while the 2015 Canadian emission levels do not include the emission reductions proposed under the Clean Air Regulatory Agenda.

Table 1: Emissions (kTonnes/Year) and overall changes in NO_{x_1} , SO_2 , VOC and $PM_{2.5}$ emissions between the 2002, 2015 Canadian and 2015 combined Canada+United States inventories

Emissions ktonnes/yr	Year	NO _x	SO ₂	VOC	PM _{2.5}
Canada	2002	2,821	2,393	2,676	532
	2015	2,130	1,867	2,300	636
Relative to					
Can. inv Relative to	Δ	-24.5%	-22.0%	-14.0%	+19.6%
Can+U.S	Δ	-3.2%	-3.3%	-1.7%	+1.3%
US CAIR					
States	2002	14,195	11,768	12,289	4,420
	2015	8,502	7,861	8,370	2,727
Relative to					
Can+US	Δ	-26.0%	-24.8%	-18.0%	-21.1%
US					
Non-CAIR	2002	4,907	1,619	6,831	3,064
States	2015	3,528	1,551	2,925	1,162
Relative to					
Can+US	Δ	-6.3%	-0.4%	-17.9%	-23.7%
US (total)	2002	19,103	13,387	19,120	7,484
	2015	12,031	9,413	11,296	3,889
Relative to					
Can+US	Δ	-32.3%	-25.2%	-35.9%	-44.8%

Table 1 provides a summary of national and regional emission totals for the two simulations, for NO_x, SO₂, VOC and primary PM_{2.5}. Subtotals are also presented for the U.S. states covered by CAIR, e.g. 28 eastern states plus Texas (see http://www.epa.gov/interstateairquality/), as well as for the remainder of the states. From a national perspective, Canadian NOx, SO2 and VOCs emissions are projected to decrease by 24.5%, 22% and 14% respectively from their 2002 reference levels. Although these reductions are relatively small in the North American context (3.2%, 3.3% and 1.7% of the combined Canadian and U.S. inventories), these changes represent substantial decreases in Canada. Primary PM_{2.5} emissions however are projected to increase in Canada by more than 19%. In the U.S., all precursor and primary emissions are projected to

decrease significantly due to the implementation of a couple of emission reduction programs. The projected reductions are particularly marked for the eastern U.S. states covered by CAIR where emission reductions would decrease the total North American NO_x emission by 26%, SO_2 by 24.8%, VOC by 18% and primary PM_{2.5} by 21.1%. In the west, the largest decreases would be associated with primary PM2.5 reduction (23.7% of the North American inventory total) and VOC reduction (17.9%). With the exception of primary PM_{2.5} emissions in Canada, the projected emission levels are much lower than the 2002 reference ones; as a result, ozone and PM_{2.5} ambient levels would be expected to decrease to varying degrees across North America, with local and/or regional departures from the general trend due to the actual spatial distribution of the emission reductions.

4. DISCUSSION

Figure 1 presents a comparison between the 2002 current case ozone levels and the 2015 future case ones for the summer season defined here as June-July-August. As expected, a marked improvement in the average summertime 8hr daily maximum is predicted for the eastern half of North America, including eastern Canada, south of the 50^{th} parallel. The average summertime 8hr daily maximum is generally expected to decrease by 3 to 10 ppb (10 to 30%) in most of the populated area in southern Ontario, southern Quebec and the Atlantic provinces, areas that are downwind of eastern north American states. In addition, a large fraction of the NO_x and VOC emission reductions



Figure 1: Absolute difference in the average O3 summertime (june-july-august) 8hr daily maximum between the future and current cases.

in Canada are also projected to be concentrated in the Windsor-Quebec City corridor, as outlined in Figure 2 and Figure 3; the projected ozone improvements hence reflect the cumulative effect of the Canadian reductions in the east and the decrease in the long-range transport of precursors and pollutants from the U.S. There are a few exceptions to the general improvement in Eastern Canada, namely the close vicinities of Toronto and Montréal. Although NO_x and VOC emissions do decrease in average, Figure 2 and Figure 3 show that there are some local increases which generally coincide with urban centres. These local increases of both precursors are especially strong in Montréal and Toronto, and likely explain, given that the transboundary transport of pollution from the U.S. is also lower, the local deterioration of the ozone concentration seen in the projections.



Figure 2: Differences in annual VOC emission levels between 2015 and 2002 inventories.



Figure 3: Differences in annual NO_x emission levels between 2015 and 2002 inventories.

Western Canada is projected to experience ozone improvements of comparable magnitude to the east in southern Alberta, in the Edmonton and Fort McMurray vicinities as well as in the south-west of British Columbia at the U.S. border. Improvements throughout Saskatchewan and Manitoba are more moderate but reached 1 to 3 ppb (5 to 10%) in the southern half of the provinces. In the Prairies, areas of improvement generally correspond to regions of marked reductions in both VOC and NO_x Canadian emissions (Alberta especially) and areas downwind of these; in British Columbia, on the other hand, ozone improvements are likely due to a combination of local and U.S. reductions in the Northern part of Washington State (Figure 2 and Figure 3). Contrasting with the general trend, northern Alberta is predicted to experience significant ozone increases. Due to a reporting

error in the 2015 projected Canadian emission inventory, emissions associated with a group of major point sources are released, in the future case only, about 5 degrees west of their actual location, i.e. the Canadian oil sand area, close to the Saskatchewan border. Despite the slight displacement, elevated ozone levels in Northern Alberta are consistent with the growth in local NO_x and VOC emissions that are simulated between the two cases. New emissions associated with the projected increase in Canadian oil sands exploitation are releasing ozone precursors in a fairly isolated area, creating favorable conditions for ozone formation. Although only a single meteorological year was studied, the impact of these sources on air quality seem constrained to Northern Alberta and Saskatchewan.



Figure 4: Absolute difference in the annual $PM_{2.5}$ 24hr average between the future and current cases.

PM_{2.5} annual levels are predicted to improve in most of the U.S. with the exception of south California, a few major centers in the intermountain states and the vicinities of Chicago and New York. In Canada, however, the trends are predicted to be quite different as outlined in Figure 4. Although levels are expected to decrease by 0.2 to 3 μ g/m³ (10 to 30%) in southern Ontario, the remaining parts of the Windsor-Quebec City corridor and most of the Atlantic provinces only see marginal improvements or actual increases of PM_{2.5} levels by 0.2 to 3 µg/m³ (15 to 30%). Similarly, urban centres in Manitoba, Saskatchewan and British Columbia are predicted to suffer from a worsening of the air quality due to $PM_{2.5}$ increases by 1 to 3 μ g/m³ (20 to 50%), while the surrounding areas stay unchanged. Most notably. Alberta and southern Saskatchewan are expected to experience an overall deterioration of $PM_{2.5}$ ambient levels by 0.2 to 3 μ g/m³ with a maximum increase larger than 3 $\mu\text{g/m}^3$ in the vicinity of Edmonton. This would represent an increase of 20 to 40% of the $PM_{2.5}$ ambient levels in Alberta and Saskatchewan but is not predicted to bring average annual levels above 10 µg/m³

throughout the Prairies except in the main urban centres (not shown).



Figure 5: Differences in annual primary $\text{PM}_{2.5}$ emission levels between 2015 and 2002 inventories.



Figure 6: Differences in annual SO_x emission levels between 2015 and 2002 inventories.

Relating the ambient PM_{2.5} changes to changes in emissions is more complex for PM species as primary PM_{2.5} emissions as well as emissions from precursors of secondary PM (NO_x, SO_x and VOC) come into play. Figure 5 does show that primary PM_{2.5} emissions are projected to increase in the Winsor-Quebec city corridor, in upwind regions such as the Ohio valley as well as in Alberta and to some extend in Saskatchewan. However this does not provide a complete explanation for the projected ambient PM_{2.5} changes as, for example, emissions of NO_x, SO_x (Figure 6) and VOC are all expected to decrease substantially overall in Alberta where ambient PM_{2.5} are projected to increase. Only NH₃ emissions are projected to undergo a moderate increase in 2015 in the Prairies and the Windsor-Quebec Corridor (not shown). From the detailed PM chemical composition, it can be deduced that the changes in PM2.5 mass in Canada are primarily driven by changes in PM2.5sulphate, PM_{2.5}-nitrate and PM_{2.5}-crustal material, and to a lesser extend PM_{2.5}-ammonium (Figure 7 to Figure 10). The remaining chemical components modelled by AURAMS (elemental carbon, primary and secondary organic carbon and sea salt) are not shown here as they only present marginal differences. It is particularly interesting to see that in the Prairies, these four PM chemical components increase despite the general decrease in precursors in the same area, suggesting that chemical reactions likely play an important role in the behavior of the system.



Figure 7: Differences in annual PM2.5-ammonium component 24hr average between the future and current cases.



Figure 8: Differences in annual PM2.5-nitrate component 24hr average between the future and current cases.



Figure 9: Differences in annual PM2.5-sulphate component 24hr average between the future and current cases.

The increases in $PM_{2.5}$ -crustal material (Figure 10), a fairly non-reactive species which dominates the $PM_{2.5}$ changes in Canadian urban centres can be directly correlated to the increase in $PM_{2.5}$ primary emissions, its only source, in the future case (Figure 5). In most of the Windsor-Quebec City corridor, the $PM_{2.5}$ crustal material component, increases enough to result in local deteriorations of $PM_{2.5}$ ambient levels, despite the

general trend for inorganic components to decrease in that area. As for ozone, $PM_{2.5}$ -sulphate, $PM_{2.5}$ -nitrate and $PM_{2.5}$ -ammonium levels are decreasing in eastern Canada as a result of the lower SO_x and NO_x emissions in Canada and especially the CAIR U.S. states, but isolated increases in local emissions in western Canada only provides a partial explanation for $PM_{2.5}$ -nitrate dominated increases in Alberta and Saskatchewan.



Figure 10: Differences in annual PM2.5-crustal material component 24hr average between the future and current cases.



Figure 11: Differences in summer (June-July-August) PM2.5sulphate component 24hr average between the future and current cases.

A finer temporal analysis was necessary to obtain more insight on the predicted ambient PM_{2.5} trends. Figure 11 to Figure 14 present the summertime and wintertime changes in the average levels of PM2.5-sulphate and PM2.5nitrate between the two simulations. PM2.5sulphate levels are generally predicted to decrease across Canada, including in most of Alberta in the summer, while PM_{2.5}-nitrate levels are predicted to experience only a few local changes, either decreases (Southern Ontario) or increases (Vancouver's vicinity, Edmonton's vicinity and around Lake Ontario). Contrasting with the summer situation, the winter averages show very little change in the PM_{2.5}-sulphate levels in Canada except for an area in Central Alberta which coincides with local SO_x emissions increase in the vicinity of Edmonton and wide areas of increase of PM2.5-nitrate levels in Alberta

and Saskatchewan as well as smaller increases in the Windsor-Quebec City corridor.



Figure 12: Differences in summer (June-July-August) PM2.5nitrate component 24hr average between the future and current cases.



Figure 14: Differences in winter (January-February-March) PM2.5-sulphate component 24hr average between the future and current cases.

Under cold conditions, the formation of ammonium nitrate is be favored in the heterogeneous $SO_4^{=}$ -NO₃⁻-NH₄⁺-H₂O system, conditions that are likely encountered in Canada in the winter. At this point however, we can only speculate that the local increases of NO_x emissions at a few point sources in Alberta and Saskatchewan (Figure 3) provide the necessary precursor species to support the winter chemistry.

Correlating the seasonal analysis with the predicted annual changes of ambient $PM_{2.5}$ (Figure 4), one can see that the increases in Alberta and the limited decreases in the Windsor-Quebec City corridor in the annual plot are the results of increases under winter conditions that are not necessarily offset by the summertime changes. These results suggest that wintertime regimes could play an important role in the response of ambient PM to emission changes, and may need to be given more attention.

5. CONCLUSION

The comparison between the 2015 future case simulation and the 2002 current case suggests different trends for the evolution of O₃ and PM_{2.5} levels in Canada. While O₃ levels would generally improve as a result of combined reductions in predictor emissions in Canada and the U.S., PM_{2.5} levels would likely deteriorate, in particular in large urbanized areas, despite the large projected reductions in U.S. emissions of primary PM_{2.5} and PM precursors. The simulation results also imply that the projected increase in PM2.5 primary emissions in Canada plays a significant role in defining the future levels of ambient PM_{2.5} and possibly modulating the improvements that might results from U.S. emission reductions. Finally, wintertime chemistry appears to have a much larger role than documented so far as in the cases simulated here the changes in winter PM_{2.5} seems to be responsible for the deterioration that is predicted overall.

The conclusions of this study rely on the accuracy of the emission inventories that are used for the simulations and hence should be treated as indicative at this point given the uncertainties inherent to emission projections. Further investigations are nonetheless warranted to study the importance of wintertime chemistry on $PM_{2.5}$ formation in Canadian industrialized areas.

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