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1. INTRODUCTION

The Canadian federal, provincial and territorial jurisdictions signed the Canada Wide Standard (CWS) agreement for ozone and particulate matter in 2000 (Canadian Council of Ministers of the Environment, 2000). This agreement establishes numerical targets and timeframes for achieving the Standards within each jurisdiction. The main motivation of this work is the beginning of an annual reporting on achievement of the Standards in 2011. The achievement determination of the numerical targets involves removing all the daily exceedances of the Standard that are not under the control of the jurisdiction. These are exceedances due to natural events, to high background levels and to transboundary flow of smog precursors. This work shows the application of an air quality model to determine if a given exceedance of the daily CWS metric is primarily due to transboundary flow or not.

2. CALCULATION OF THE LOCAL AND TRANSBOUNDARY CONTRIBUTION TO SMOG

In order to determine if an ozone or $PM_{2.5}$ exceedance is due to transboundary flow or to sources within the jurisdiction, the following hypothesis is made. The measured concentrations in a Reporting area can be represented as the sum of a background, a local and a transboundary contribution. The background concentration is independent of long range transport of species from anthropogenic sources, but is rather a function of the natural environment, including the emission and transport of biogenic species. To estimate the three terms, an air quality model will be used.

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Three emission scenarios will be run, one with all anthropogenic emissions (base case), a second with anthropogenic emissions set to zero within the jurisdiction and a third with anthropogenic emissions outside the jurisdiction set to zero. Let's define the following quantities :

C_i	Measured concentration at location i
C_i^m	Model concentration at location i obtained with all anthropogenic emissions
C_i^{j0}	Model concentration at location i obtained with anthropogenic emissions within the jurisdiction set to zero
C_i^{jn0}	Model concentration at location i obtained with anthropogenic emissions outside the jurisdiction set to zero
B_i	Background concentration at location i
C_i^{local}	Contribution of the jurisdiction to total concentration at location i
C_i^{trb}	Transboundary contribution to total concentration at location i

The following equation summarizes the hypothesis :

$$C_i^m = B_i + C_i^{local} + C_i^{trb} \quad (1)$$

The scenario with all anthropogenic emissions within the jurisdiction set to zero must lead to a local contribution equal to zero. Similarly, the scenario with all anthropogenic emissions outside the jurisdiction set to zero leads to a transboundary contribution equal to zero. These statements can be substituted in equation (1) and one obtains :

$$C_i^{j0} = B_i + C_i^{trb} \quad (2)$$

$$C_i^{jn0} = B_i + C_i^{local} \quad (3)$$

Equations 1-3 can now be solved for the three unknowns, the background, the local and the transboundary contributions :

$$\begin{aligned} B_i &= C_i^{j0} + C_i^{jn0} - C_i^m \\ C_i^{local} &= C_i^m - C_i^{j0} \\ C_i^{trb} &= C_i^m - C_i^{jn0} \end{aligned} \quad (4)$$

The *Guidance Document on Achievement Determination* (Canadian Council of Ministers of the Environment, 2002) gives a methodology to determine if a daily exceedance of the Standard is mainly due to transboundary flow. When air quality modelling is performed, an exceedance is qualified as transboundary influenced if the daily value at the monitor continues to be within 10% or above the Standard after removing the local contribution (as computed from equation 4) from the actual measurement. In mathematical terms, an exceedance is transboundary influenced if :

$$C_i - C_i^{local} \geq CWS_{threshold} \quad (5)$$

where $CWS_{threshold}$ is set to 10% below the daily Standard. For ozone, the daily value is the maximum of the 8-hour running average of hourly concentrations with the Standard set to 65 ppb. For $PM_{2.5}$, the daily value is the 24-hour average of the hourly concentrations with the Standard set to $30 \mu\text{g}/\text{m}^3$. As a result, the thresholds are :

$$\begin{aligned} \text{For ozone : } CWS_{threshold} &= 59 \text{ ppb} \\ \text{For } PM_{2.5} : CWS_{threshold} &= 27 \mu\text{g}/\text{m}^3 \end{aligned}$$

It has to be noted that the terms on the left hand side of equation 5 are not any more instantaneous values of the concentrations, but are the daily metrics of the Standard. To ascertain the validity of the formulation, a more stringent criteria to qualify an event as transboundary influenced is evaluated in this study. It includes the above criteria (equation 5) plus a second one. The exceedance would be qualified as transboundary influenced if the concentrations at the monitor drops below 10% of the Standard after removing the transboundary contribution (as computed from equation 4) from the actual measurement. In mathematical term, an exceedance is transboundary influenced if it satisfies equation (5) and the following :

$$C_i - C_i^{trb} < CWS_{threshold} \quad (6)$$

where the thresholds are the same as above.

3. DESCRIPTION OF THE ATMOSPHERIC MODELS AND THE SIMULATED EPISODES

The meteorological model selected to drive the air quality model is the Global Environmental Multiscale model (GEM) of the Meteorological Service of Canada (Côté, 1998). GEM is based on the fully compressible Euler equations solved by an implicit and semi-lagrangian method. The integration domain covers all North America at a resolution of 24 km. The grid has 28 levels along the vertical axis and the model top is set at 10 mb. The bottom 7 levels are in the planetary boundary layer.

The air quality model used in this study is AURAMS recently developed by the Meteorological Service of Canada (Moran, 1998; Makar, 2003a; Bouchet, 2003). AURAMS is a source based eulerian model linking emissions to the atmosphere and ambient air concentrations and deposition. The treatment of transport and advection is based on the CHRONOS model (Pudykiewicz, 1997). The gas phase chemistry is taken from the Acid Deposition and Oxidant Model (ADOM) (MacDonald, 1993) with 114 reactions and 47 species. The model includes aqueous phase chemistry as well as heterogeneous phase chemistry (Makar, 2003b). The aerosol module is based on the Canadian Aerosol Module (Gong, 2003). This aerosol module is size-resolved with 12 bins covering particle sizes from 0.01 to 40.96 microns and includes 8 chemical components : sulphate, nitrate, ammonium, sea salt, black carbon, organic carbon, crustal material and water.

AURAMS was run on a domain covering Northeastern North America (figure 1) on a grid with 125 x 110 points and a resolution of 21 km. The meteorological fields needed by the model were obtained from the GEM model outputs by interpolation. The input anthropogenic emission rates were based on the 1995-1996 national inventories processed with the Canadian Emission Processing System (CEPS1.0) (Moran, 1997). The biogenic emissions were computed with the Biogenic Emission Inventory System version 2 (BEIS-2) (Geron, 1994)

This study focusses on ozone and $PM_{2.5}$ exceedances in the Quebec portion of the Windsor-Quebec city corridor. Air quality measurements of 1999 to 2001 were analysed and a limited number of episodes were selected so as to include exceedances due to local sources and to transboundary flow. Three episodes with exceedances of the Standard for ozone and/or $PM_{2.5}$ in 1999 and 2001 were simulated with the models. They are :

July 11-18, 1999
 June 12-21, 2001
 July 29-August 4, 2001

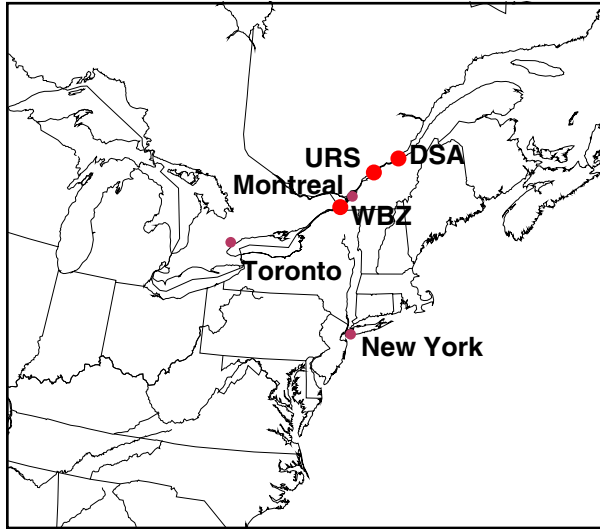


Figure 1 : AURAMS modelling domain. The sites referred to in this study are also shown. They are St. Anicet (WBZ), Trois-Rivieres (URS) and Quebec City (DSA).

4. EVALUATION OF THE MODEL

Since AURAMS has not yet been evaluated over Southern Quebec, an evaluation of the $PM_{2.5}$ concentrations was performed at three sites along the St. Lawrence valley. An evaluation of the ozone concentrations is not performed in this work since this module is identical to the CHRONOS module which has been evaluated many times with very satisfactory results.

The location of the sites for the $PM_{2.5}$ evaluation are shown on figure 1. St. Anicet is located southwest of Montreal which is a major source of smog precursors. Trois-Rivieres and Quebec City are located northeast of Montreal and, on days with smog, are usually downwind of Montreal whereas St. Anicet is upwind. Since this study focusses on the Canada Wide Standards, the comparison between the model values and measurements will be limited to the daily average of the $PM_{2.5}$ concentrations.

The AURAMS model outputs $PM_{2.5}$ concentrations every hour at every grid point and several heights above ground. The first level near the ground was selected as this is the closest to the usual height where measurements are taken. The model concentrations were then interpolated linearly to the location of three sites, then the daily average concentration was computed. The following figure 2 shows the observed values (PM25 Obs) and the model values (PM25 AURAMS). The local and transboundary contributions as computed with equation (4) are also shown on the figure and will also be discussed.

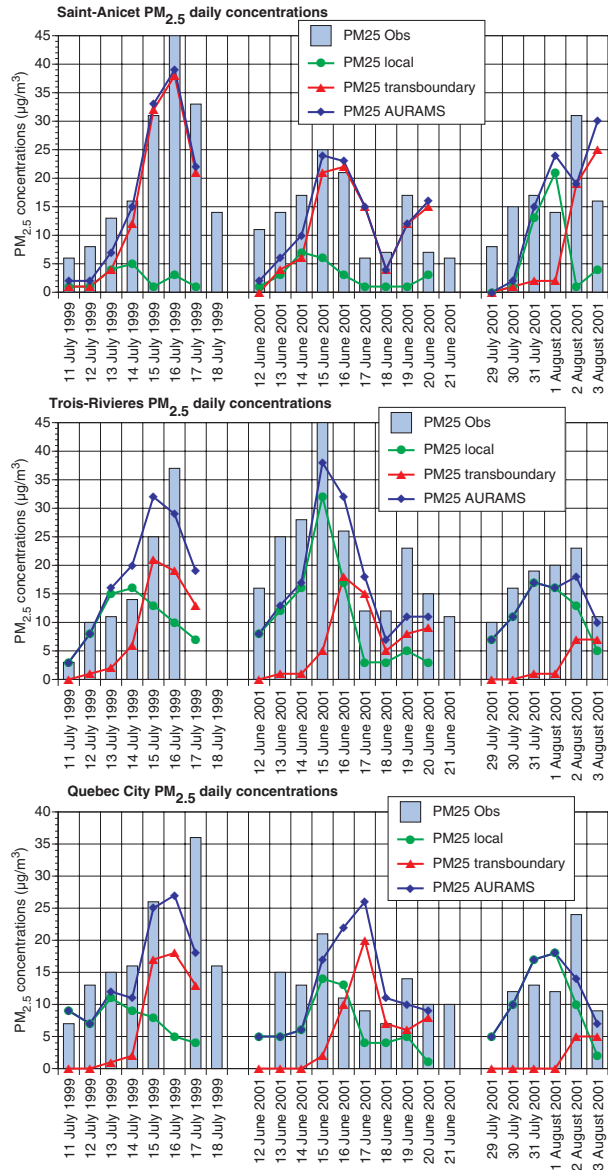


Figure 2 : Daily $PM_{2.5}$ concentrations during the three episodes. The measurements (light blue columns) and the model values (blue line) are shown. The local and transboundary contribution computed with equation (4) are also shown in green and red respectively.

At St. Anicet, the model values and the observations agree quite well with $PM_{2.5}$ peaks synchronized, except perhaps on August 2001 where the model has the peak on the 3rd while the observations are maximum on the 2nd.

The model agrees quite well with the observations at Trois-Rivieres as well. The local contribution is more important here than at St. Anicet. This is to be expected since Trois-Rivieres is downwind of Montreal which is a major local source of smog precursors. Surprisingly, during the 19 June 2001

maximum, the transboundary component is barely above the local one. The meteorological analysis reveals that there was a strong southwesterly flow during the evening of June 19 which caused an ozone exceedance that was due to transboundary flow. The reason why the transboundary signal is not so clear for $PM_{2.5}$ on that day may be that the peak concentration is somewhat underpredicted due to a lack in the transport of pollutants within the model or too strong a deposition.

Further east, in Quebec City, the agreement between the model values and the observations is not as good. The exceedance of July 17 1999 is missed; a peak concentration is predicted on 17 June 2001 while it occurred on June 15 and on June 19. The peak $PM_{2.5}$ concentrations observed on 2 August 2001 is predicted to happen one day earlier.

Modelled $PM_{2.5}$ concentrations were split into the three components according to equation (4) and the local and transboundary parts can be seen on figure 2. When the total modelled concentrations are high, the signal is quite clear and not ambiguous : one of the two components dominates. Past and present weather patterns determine which of the local sources or the remote transboundary sources dominate. A few specific cases will be examined in the next section.

This simple assessment of AURAMS doesn't show all strengths and weaknesses of the model in predicting $PM_{2.5}$ and the cause of the disagreement, but is intended to make the reader aware that complex air quality models such as AURAMS are very useful tools and that they are constantly evolving.

5. EVALUATION OF THE TRANSBOUNDARY INFLUENCE ON DAYS WITH CWS EXCEEDANCES

In the province of Quebec, the main source of local smog precursors is the Montreal region with its high level of NO_x emissions. The transboundary sources come from the industrialized southern Ontario, the American Midwest and to a lesser extent the industrialized East from Washington to Boston.

The typical weather pattern leading to high ozone or $PM_{2.5}$ levels in Southern Quebec is stagnation under a high pressure system to accumulate pollutants, followed by a southwesterly flow to carry the pollutants from the Great Lakes area and Montreal along the St. Lawrence valley up to Quebec City.

Among all exceedances that occurred during the modelled episodes, two cases will be shown with more details, one where the exceedance is due to local sources and a second where it is due to transboundary flow. All other cases were also analysed and there were generally a good agreement between the backtrajectory analysis and the model results.

5.1 Case of July 31, 2001

On that day, ozone exceedances of the 8-hour running average were reported in the Montreal area including at the St. Anicet site. However, no $PM_{2.5}$ exceedances were reported. The following figure 3 shows the 48-hour backtrajectory of the air mass arriving in the Montreal area at a height of 925 mb at 1800 UTC (2:00 PM local time).

The backtrajectory clearly indicates that the air mass is of local origin and the exceedances are due to local sources. The following table 1 shows the model results and the measurements for 2 sites, St. Anicet and West Montreal, this latter site being in the western suburb of Montreal, about 40 km to the north-east of St. Anicet.

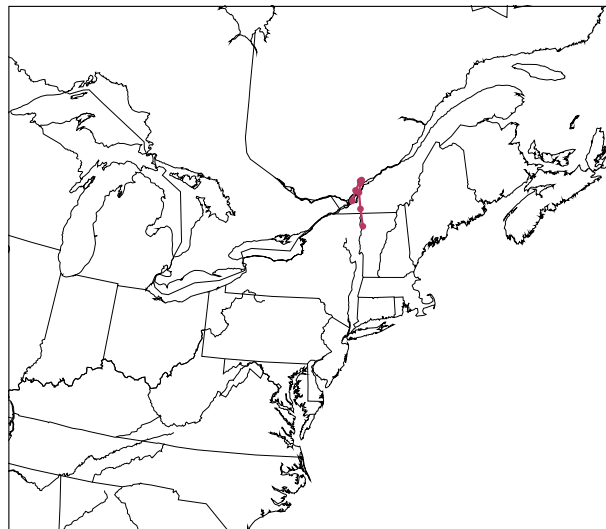


Figure 3 : 48-hour 925 mb backtrajectory (in red) for Montreal/St. Anicet ending at 1800 UTC on July 31, 2001. Each line segment between the dots represent 6 hours.

At the St. Anicet site, both equations (5) and (6) indicate that the ozone exceedance is due to local sources in agreement with the backtrajectory analysis.

TABLE 1. Model results and diagnosis of transboundary influence

August 31, 2001, ozone	St. Anicet	West Montreal
Observation	73 ppb	90 ppb
Model value	104 ppb	62 ppb
Local contribution	63 ppb	21 ppb
Transboundary contribution	4 ppb	5 ppb
Background	37 ppb	36 ppb
Transboundary (eq 5) ?	No	Yes
Transboundary (eq 6) ?	No	No

It is noted also that the model overestimates ozone concentrations by about 30 ppb. On the other hand, equation (5) indicates a transboundary influence for the West Montreal site. Model values between the 2 sites, which are about 40 km apart, have a difference of about 40 ppb. The reason is that the West Montreal site is too close in term of model grid points to the high NO_x emissions produced by the city and is affected by NO_x titration of ozone in the model but not in the real world. This is why at this site the model underestimates ozone concentrations and that the results from equation (5) are not correct.

5.2 Case of June 15, 2001

On that day, ozone exceedances were reported in the Montreal area including the St. Anicet site, and also at the Trois-Rivieres and Quebec City sites. A PM_{2.5} exceedance was also reported at the Trois-Rivieres site. Figure 4 shows the backtrajectories arriving at the 3 sites at 925 mb on June 15. The duration of the backtrajectory is again 48 hours, which is sufficient to estimate if the air mass comes from a source region or not.

The backtrajectories arriving at Trois-Rivieres and Quebec City come from a source region covering Toronto (Canada) and Buffalo (USA) regions. However, it does not appear that the backtrajectory arriving in Montreal (St. Anicet) comes from a source region, being away from the major urban centres. The model results will help to diagnose the transboundary influence in this case.

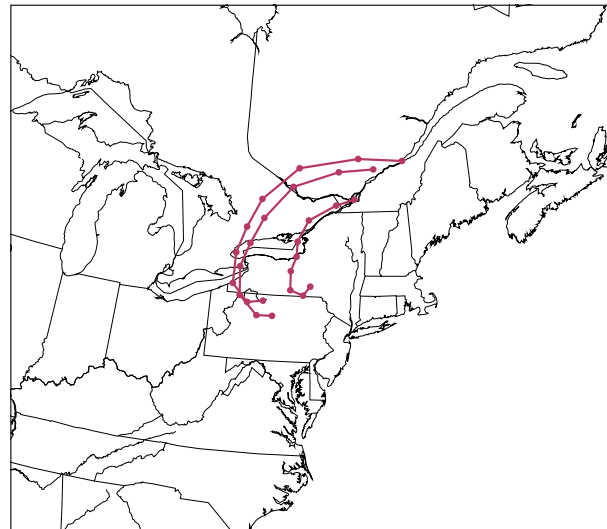


Figure 4 : 48-hour 925 mb backtrajectory for the 3 sites ending at 1800 UTC on June 15, 2001. Each line segment between the dots represent 6 hours.

Table 2 presents the model results for ozone. Despite the fact that the ozone concentrations are again slightly overestimated, the model gives a clear signal that the local component is dominant for Trois-Rivieres and Quebec City and that the transboundary component dominates for St. Anicet. As a result, the transboundary influence is confirmed by equations (5) and (6) at St. Anicet. It should be concluded that the two other sites are not mostly transboundary influenced since both equation lead to different results and that the local component clearly dominates.

TABLE 2. Model results and diagnosis of transboundary influence

June 15, 2001, ozone	St. Anicet	Trois-Rivieres	Quebec City
Observation	72 ppb	67 ppb	67 ppb
Model value	83 ppb	81 ppb	81 ppb
Local contribution	4 ppb	36 ppb	34 ppb
Transboundary contribution	36 ppb	11 ppb	9 ppb
Background	43 ppb	34 ppb	38 ppb
Transboundary (eq 5) ?	Yes	No	No
Transboundary (eq 6) ?	Yes	Yes	Yes

Table 3 below presents the model results for the daily PM_{2.5} exceedance at the Trois-Rivieres site. Similarly to the ozone results above, the local component is dominant and the elevated concentrations value of 45 µg/m³ make both

equations (5) and (6) agree with a non influence of transboundary flow. This conclusion is reinforced by the fact that no other PM monitor reported exceedances of the Standards in the vicinity.

TABLE 3. Model results and diagnosis of transboundary influence

June 15, 2001, PM _{2.5}	Trois-Rivieres
Observation	45 µg/m ³
Model value	38 µg/m ³
Local contribution	32 µg/m ³
Transboundary contribution	5 µg/m ³
Background	1 µg/m ³
Transboundary (eq 5) ?	No
Transboundary (eq 6) ?	No

6. CONCLUSION

This study focussed on the daily exceedances of the Canada Wide Standard for ozone and PM_{2.5} over Southern Quebec during summertime. Three episodes were simulated with the AURAMS model for a total of 25 days. The PM module of the model was briefly evaluated and it appeared that the model can reproduce the PM_{2.5} concentrations quite well in the Montreal and Trois-Rivieres regions. The model was not as good in the Quebec City area which is farther from major sources of PM_{2.5} and its precursors. There might be a problem with advection and/or deposition of PM_{2.5} in the model. Further analysis is needed.

The modelled ozone and PM_{2.5} concentrations were split into three components, the local, transboundary and background using model runs obtained by setting successively to zero anthropogenic emissions within and outside the jurisdiction. The local and transboundary components were used to assess the transboundary influence during CWS exceedances. By subtracting in turn the local and the transboundary component from the actual measurement and comparing against a threshold, the usefulness of this two-criteria approach was demonstrated. In all cases, the model simulations have to be compared to field measurements to verify if it can reproduce adequately the current situation, then the results must be compared with other techniques such as a backtrajectory analysis or the use of an upwind monitor as described in the Guidance Document on Achievement Determination.

The study highlighted the fact that the model can predict the wrong result if the grid cannot represent adequately a given site. The

measurement station must be representative of an area similar to model resolution. In this study, the model resolution was set to 21 km and it was seen that the West Montreal site was not correctly represented by the model.

7. REFERENCES

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