

A COMPARATIVE DYNAMIC EVALUATION OF THE AURAMS AND CMAQ AIR QUALITY MODELING SYSTEMS

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1. INTRODUCTION

A comparative air-quality (AQ) model performance evaluation involves a side-by-side comparison of the predictions of two or more AQ models that employed either comparable or harmonized inputs and grids (e.g., McKeen et al., 2005, 2007; Smyth et al., 2007, Stern et al., 2008). A dynamic AQ model performance evaluation examines an AQ model's ability to simulate changes in the concentrations or deposition of air pollutants in response to changes in either emissions or meteorological conditions (e.g., Gilliland et al., 2008; Pun et al., 2008). In this study, these two evaluation approaches are combined to perform a simple harmonized comparative dynamic evaluation of the AURAMS and CMAQ AQ modeling systems.

Both modeling systems were previously applied to the same one-month period (July 2002) on identical North American grids using the same raw emissions, emissions processor, and input meteorology (Smyth et al., 2008). Building on this base case, the same systems have now been run again for the same period, map projection and grid, and meteorological inputs but for four additional sets of emission inputs.

Following the methodology of Smyth et al. (2008), the changes of surface hourly O₃, daily maximum and daily minimum O₃, and hourly PM_{2.5} total-mass and major-species concentrations predicted by the two models in response to the four sets of emissions changes were then compared to the base case at (a) measurement sites only and (b) over all grid cells in the modeling domain. This procedure allowed the signs and the magnitudes of the models's responses for each of these emissions sensitivity cases to be assessed for a number of species and metrics. The benefit

of considering multiple models in this way is that information is also available on the variability and hence on the uncertainty of model predictions, which may be useful additional guidance to users of model predictions such as policymakers.

2. METHODOLOGY

2.1 Base Case

AURAMS version 1.3.1b and CMAQ version 4.6 were both run on the same grid (polar-stereographic map projection, 42-km horizontal grid spacing) that spans North America from northern Mexico to northern Canada (see Figure 1). The models were run for the 696-hour, July 1-29, 2002 simulation period. Both models used meteorological fields generated by version 3.2.0 of GEM, Environment Canada's (EnvCan) operational weather forecasting model.

AURAMS used the ADOM-II gas-phase chemistry mechanism while the SAPRC-99 mechanism was used with CMAQ.

The same three national emission inventories were used for both models:

- 2000 Canadian emission inventories for point, area, non-road mobile, and on-road mobile sources released by EnvCan in January 2005;
- U.S. EPA 2001 Clean Air Interstate Rule (CAIR) emission inventories for point, area, non-road mobile, and on-road mobile sources released in July 2004 (available from the U.S. EPA emissions modeling clearinghouse at <http://www.epa.gov/ttn/chief/emch/index.html>);
- 1999 Mexican emissions inventories for point, area and mobile sources released with the 2001 EPA CAIR data.

All anthropogenic emissions were processed using SMOKE version 2.2. Biogenic emissions were generated using BEISv3.09 algorithms and the BELD3 land-use data set. For more details on the model set ups, see Smyth et al. (2007, 2008).

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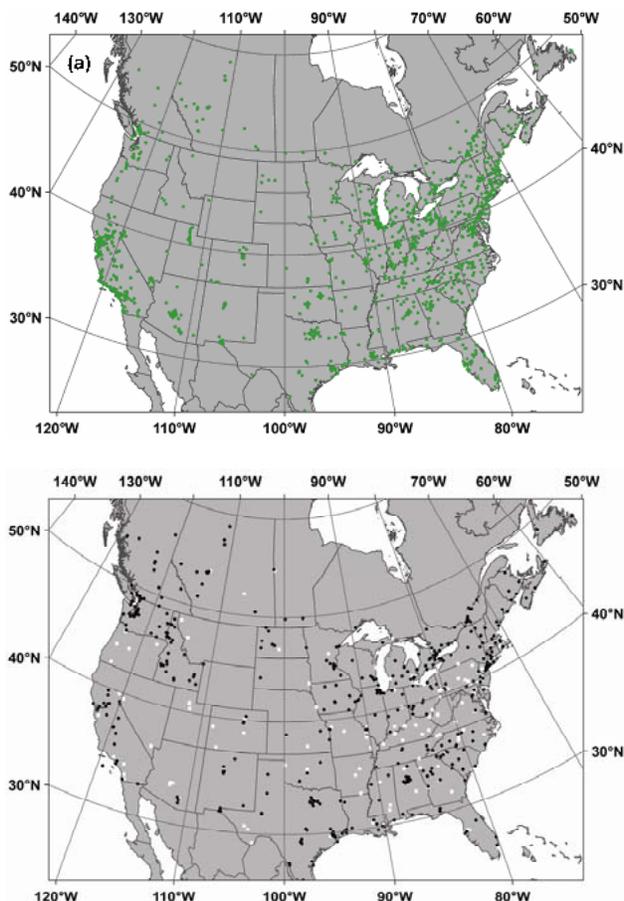


Figure 1. AURAMS and CMAQ modelling domain with locations of (a) O₃ measurement sites and (b) PM_{2.5} measurement sites with usable July 2002 data indicated (black circles denote total PM_{2.5} sites and white squares denote speciated PM_{2.5} sites).

2.2 Emission Scenarios

Four simple “roll-back”-type NO_x-VOC emission scenarios were considered relative to the base-case emissions data set described in Section 2.1:

- Increase NO_x emissions by 50% ($1.5NO_x$);
- Decrease NO_x emissions by 50% ($0.5NO_x$);
- Decrease VOC emissions by 50% ($0.5VOC$);
- Decrease NO_x and VOC emissions by 50% ($0.5NO_x+0.5VOC$).

In these emission scenarios, emissions from *all* sources were modified by the same percentage, including biogenic sources. AURAMS and CMAQ were then re-run for the same July 2002 period using the same meteorological inputs as in the base case.

2.3 Statistical Metrics

The statistical metrics used in this study are defined in Table 1. The spatial comparisons were generated by calculating the minimum, maximum, and average absolute and relative differences between the base case and each emissions sensitivity case for each grid cell. Statistics were then calculated from these various difference fields either for (a) the set of grid cells containing at least one measurement station or (b) all grid cells. In the second case, the base-case values were treated as “observed” values.

3. RESULTS

3.1 Base-Case Results

As reported by Smyth et al. (2008), a comparison of AURAMS- and CMAQ-predicted hourly O₃ concentrations against hourly surface measurement data from both Canadian and U.S. stations (see Figure 1a) for the July 2002 period showed a lower normalized mean bias (NMB) of 20.7% for AURAMS versus 46.4% for CMAQ. However, AURAMS and CMAQ had more similar normalized mean errors (NMEs) of 46.9% and 54.2%, respectively. Both models did similarly well in predicting daily 1-hour O₃ maximums; however, AURAMS performed better in calculating daily minimums. CMAQ’s poorer performance in predicting hourly O₃ is thus partly due to its inability to correctly predict night-time lows.

Total PM_{2.5} hourly surface concentration was under-predicted by both AURAMS and CMAQ with NMBs of -10.4% and -65.2%, respectively (station locations are shown in Figure 1b). However, as with O₃, both models had similar NMEs of 68.0% and 70.6%, respectively. In general, AURAMS performance was better than CMAQ for all PM_{2.5} major species except nitrate (NO₃) and elemental carbon (EC). Both models significantly under-predicted total organic aerosol (TOA), although the mean AURAMS concentration was over four times larger than CMAQ’s. The under-prediction of TOA was partly due to the exclusion of forest-fire emissions. Sea-salt aerosol made up 50.2% of the AURAMS total PM_{2.5} surface concentration versus only 6.2% in CMAQ when averaged over all grid cells. When averaged over land cells only, sea-salt still contributed 13.9% to the total PM_{2.5} mass in AURAMS versus 2.0% in CMAQ.

3.2 Emission-Scenario Results

Table 2 summarizes the AURAMS results for the base case and the four emission scenarios for surface hourly O₃, daily maximum and daily minimum O₃, and hourly total PM_{2.5} concentrations for grid cells containing O₃ and PM_{2.5} measurement stations. Table 3 lists the corresponding CMAQ results.

First comparing these two tables qualitatively, we see that for the base case, AURAMS mean O₃ predictions are lower than CMAQ O₃ predictions and AURAMS mean PM_{2.5} predictions are higher than CMAQ PM_{2.5} predictions. Also, with one exception, the impact of decreasing NO_x emissions ("0.5NO_x" scenario), decreasing VOC emissions ("0.5VOC" scenario), or decreasing both together ("0.5NO_x+0.5VOC" scenario) across the domain by 50% is to decrease both O₃ and PM_{2.5} mean concentrations. The exception is for total PM_{2.5} for the "0.5VOC" scenario, for which AURAMS predicted a decrease of 15.5% but CMAQ predicted an increase of 2.0%. For the "1.5NO_x" scenario, again with one exception, the impact of increasing NO_x emissions across the domain by 50% is to increase both O₃ and PM_{2.5} mean concentrations. The exception is for daily minimum hourly O₃, for which AURAMS predicted a decrease of -4.2% but CMAQ predicted an increase of 2.1%.

Turning to quantitative responses, AURAMS was less sensitive to NO_x-emissions-only changes than was CMAQ for surface-level O₃ concentrations both in terms of mean differences (MD) and normalized mean differences (NMD). On the other hand, AURAMS was more sensitive in general to VOC-emissions-only changes than was CMAQ for surface-level O₃ concentrations, especially for daily maximum O₃. The one exception was hourly O₃, for which AURAMS had a slightly larger NMD value (-15.3% vs. -14.9%) but a smaller MD value (-6.6 vs. -7.8 ppbV). AURAMS was also more sensitive overall for the "0.5NO_x+0.5VOC" scenario than was CMAQ for surface-level O₃ concentrations.

For surface-level total PM_{2.5} concentrations, AURAMS was also more sensitive than was CMAQ to both NO_x and VOC emission changes. In order to understand the differences between the models's PM responses, it is useful to examine the responses of some of the major PM chemical components to NO_x and VOC emission changes.

Tables 4 and 5 summarize the AURAMS and CMAQ responses, respectively, to the emission-change scenarios for five PM_{2.5} major species: SO₄, NO₃, NH₄, EC, and TOA. One interesting

finding from these tables is that while NO_x emissions are the precursor of PM_{2.5} NO₃ and VOC emissions are a precursor of PM_{2.5} TOA, other PM_{2.5} major species may also be affected by NO_x or VOC emission changes and the response may be "antagonistic", i.e., of opposing sign.

For example, for the "0.5NO_x" scenario, AURAMS and CMAQ both predicted that the 50% reduction in NO_x emissions would reduce atmospheric PM_{2.5} NO₃ concentrations, but they also both predicted that PM_{2.5} SO₄ and PM_{2.5} TOA would also be reduced. For the "1.5NO_x" scenario, both models predicted increases in the same species, with the exception of AURAMS's prediction of a small decrease in PM_{2.5} SO₄. Pun et al. (2008) presented CMAQ results (for the RADM2 gas-phase chemistry mechanism) from a 10-day simulation in July 1999 for a 50% NO_x-emission-reduction scenario that showed similar behavior as the "0.5NO_x" scenario here.

For the "0.5VOC" scenario, AURAMS and CMAQ both predicted that PM_{2.5} TOA would be reduced. However, as already mentioned, CMAQ, unlike AURAMS, predicted that total PM_{2.5} mass would increase, not decrease. As can be seen from Table 5, the reason is that CMAQ (using the SAPRC-99 gas-phase chemistry mechanism) predicted that both PM_{2.5} SO₄ and PM_{2.5} NO₃ would increase, more than compensating for the decrease in PM_{2.5} TOA. AURAMS, on the other hand, predicted that the reduction in VOC emissions would result in a decrease of PM_{2.5} SO₄ and no net change in PM_{2.5} NO₃ (see Table 4). By comparison, for a similar 50%-VOC-emission-reduction scenario, Pun et al. (2008) found that CMAQ (using the RADM2 gas-phase chemistry mechanism) predicted similar behavior to CMAQ in this study for the "0.5VOC" scenario.

Figure 2 shows the spatial distribution of the average absolute difference fields between the base case and the "0.5VOC" scenario predicted by AURAMS and CMAQ. The two fields are very different. The large difference in the magnitude of the mean change in PM_{2.5} TOA predicted by AURAMS and CMAQ (-2.4 vs. -0.3 µg/m³) and the smaller difference in the magnitude of the mean change in the sum of PM_{2.5} SO₄ and PM_{2.5} NO₃ predicted by the two models (-0.3 vs. 0.35 µg/m³) suggests that the AURAMS average absolute difference field (Figure 2a) is dominated by the change in PM_{2.5} TOA whereas the predicted change in the two inorganic PM_{2.5} species has a much greater relative influence in the CMAQ field (Figure 2b). Interestingly, the PM_{2.5} difference field predicted by CMAQ for a similar scenario in the

Pun et al. (2008) study is more like the AURAMS field. Note that one significant difference between the two models is that the SOA scheme used by AURAMS includes isoprene contributions whereas the CMAQ SOA scheme does not.

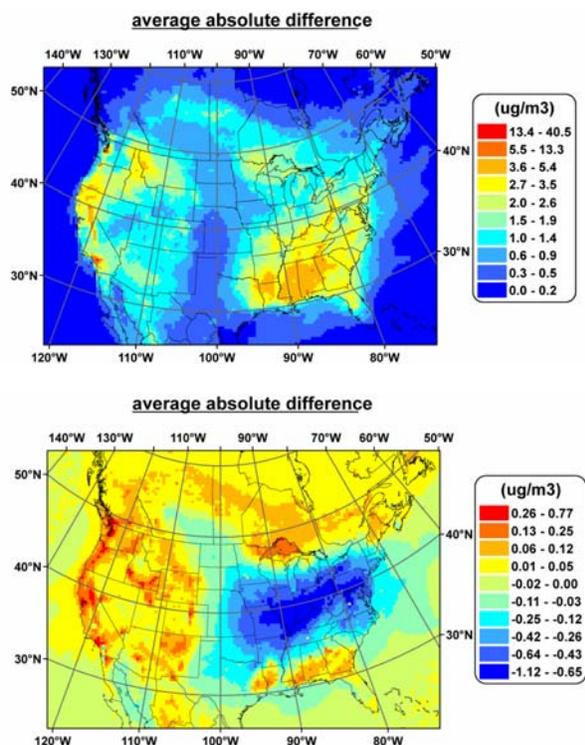


Figure 2. Average difference of surface-level PM_{2.5} (µg/m³) between the base case and “0.5VOC” scenario for (a) AURAMS and (b) CMAQ for the July 2002 period.

4. DISCUSSION

Stockwell et al. (1988) showed that both gas-phase and aqueous-phase SO₄ production rates are dependent on atmospheric NO_x and VOC concentrations. They demonstrated for the RADM2 mechanism that in low-NO_x areas (e.g., rural areas), reductions in VOC emissions lead to *increases* in gas-phase SO₄ production rate by increasing oxidant concentrations but to either increases or decreases in gas-phase SO₄ production rate in high-NO_x areas. In contrast, reductions in NO_x emissions in low-NO_x areas lead to *decreases* in oxidant concentrations and gas-phase SO₄ production rate and to either increases or decreases in gas-phase SO₄ production rate in high-NO_x areas. Pun et al. (2008) suggested that (a) the decrease in oxidant levels in low-NO_x (NO_x-limited) areas due to NO_x emission reductions will also decrease secondary organic aerosol formation and (b) the increase in oxidant levels caused by VOC emission reductions will also

increase the production of both SO₄ and HNO₃. Meng et al. (1997) reported the results of a similar analysis, but only for two-day simulations with the CIT model in the VOC-limited Los Angeles Basin.

The Stockwell et al. (1988) and Pun et al. (2008) results are consistent with the AURAMS and CMAQ PM species predictions for the “0.5NO_x” emission scenario and with the behavior of CMAQ for the “0.5VOC” scenario. For the “0.5VOC” scenario, however, AURAMS using the ADOM-II gas-phase mechanism, predicted that PM_{2.5} SO₄ mass would decrease, not increase, overall and that the domain-average PM_{2.5} NO₃ mass would remain the same.

It is interesting to examine the model results for the “0.5NO_x+0.5VOC” emissions scenario in light of the above analysis. Tables 4 and 5 suggest that the impacts for the component scenarios (i.e., “0.5NO_x”, “0.5VOC”) preserve the general direction (sign) but are not additive for most PM components. For example, the NMD values for PM_{2.5} TOA for AURAMS for the “0.5NO_x” and “0.5VOC” scenarios are -6% and -50%, respectively, but the corresponding value for the “0.5NO_x+0.5VOC” scenario remains at -50%. For CMAQ, the corresponding values are -2%, -29%, and -28%.

5. SUMMARY AND CONCLUSIONS

In this sensitivity study, O₃ and PM_{2.5} concentration changes resulting from four sets of arbitrary percentage changes to bulk NO_x and VOC emissions were analysed for July 2002 for paired simulations with harmonized set-ups of the AURAMS and CMAQ AQ modeling systems. Such a comparative dynamic evaluation of the AURAMS and CMAQ AQ modeling systems provides a measure of the uncertainty in the predictions of two important pollutants for policy applications of these two AQ modeling systems. Given the considerable harmonization that was achieved in this study, this comparison also yields insights into the influence of differences in model science on resulting model predictions.

Hourly O₃ and daily maximum and minimum O₃ concentrations were evaluated at measurement sites for each model and each emissions scenario. For NO_x emission changes alone, CMAQ was in general more sensitive than was AURAMS for all O₃ metrics. For VOC emission reductions alone, AURAMS daily maximum and minimum O₃ was more sensitive, while the models responded similarly for hourly O₃. And for combined NO_x and VOC emission reductions, the two models responded similarly for all O₃ metrics.

Table 1. Statistical metrics used for evaluation and comparison of sensitivity cases to the base case.

mean concentration	$(1/n) (\sum_i m_i)^a$
mean bias (MB); mean difference (MD)	$(1/n) \sum_i (m_i - o_i)$
normalized mean bias (NMB); normalized mean difference (NMD)	$[\sum_i (m_i - o_i) / \sum_i o_i] \times 100\%$
normalized mean error (NME); normalized mean absolute difference (NMAD)	$[\sum_i m_i - o_i / \sum_i o_i] \times 100\%$

^a n is the total number of data pairs and $i = 1, 2, \dots, n$; o is the measured concentration, m is the modelled concentration; for sensitivity case comparison, o is the base case and m is the sensitivity case.

Table 2. Comparison of AURAMS mean concentrations of surface-level O₃, daily maximum O₃, daily minimum O₃, all in units of ppbV, and total PM_{2.5} (μg m⁻³) for base case and sensitivity cases at measurement site locations only. NMD values are percentages.

Species	base	1.5NO _x			0.5NO _x			0.5VOC			0.5NO _x + 0.5VOC		
	MC	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD
hourly O ₃	43.0	45.3	2.3	5.3	36.4	-6.6	-15.3	36.4	-6.6	-15.3	33.1	-9.9	-23.0
max. O ₃	68.4	74.2	5.8	8.4	55.5	-12.9	-18.8	57.3	-11.1	-16.2	49.6	-18.8	-27.5
min. O ₃	16.5	15.8	-0.7	-4.2	15.8	-0.7	-4.2	14.2	-2.3	-13.9	14.8	-1.7	-10.3
total PM _{2.5}	12.9	13.6	0.7	5.4	11.8	-1.1	-8.5	10.9	-2.0	-15.5	10.1	-2.8	-21.7

Table 3. Same as Table 2 but for CMAQ.

Species	base	1.5NO _x			0.5NO _x			0.5VOC			0.5NO _x + 0.5VOC		
	MC	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD
hourly O ₃	52.2	57.2	5.0	9.6	43.1	-9.1	-17.4	49.4	-7.8	-14.9	43.2	-9.0	-17.2
max. O ₃	71.6	81.4	9.8	13.7	55.7	-15.9	-22.2	66.7	-4.9	-6.8	55.0	-16.6	-23.2
min. O ₃	32.6	33.3	0.7	2.1	29.8	-2.8	-8.6	31.6	-1.0	-3.1	30.4	-2.2	-6.7
total PM _{2.5}	5.0	5.2	0.2	4.0	4.8	-0.2	-4.0	5.1	0.1	2.0	4.9	-0.1	-2.0

Table 4. Comparison of AURAMS mean concentrations of surface-level total PM_{2.5} and various major PM_{2.5} species (all in units of μg m⁻³) for base case and sensitivity cases at measurement site locations only. NMD values are percentages.

PM Species	base	1.5NO _x			0.5NO _x			0.5VOC			0.5NO _x + 0.5VOC		
	MC	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD
total PM _{2.5}	12.9	13.6	0.7	5	11.8	-1.1	-9	10.9	-2.0	-16	10.1	-2.8	-22
PM _{2.5} SO ₄	5.5	5.4	-0.1	-2	5.3	-0.2	-4	5.2	-0.3	-5	5.2	-0.3	-5
PM _{2.5} NO ₃	1.9	2.6	0.7	36	0.92	-0.98	-52	1.9	0.0	0	1.1	-0.8	-42
PM _{2.5} NH ₄	1.6	1.8	0.2	12	1.4	-0.2	-12	1.6	0.0	0	1.4	-0.2	-12
PM _{2.5} EC	0.28	0.28	0.0	0	0.28	0.0	0	0.28	0.0	0	0.28	0.0	0
PM _{2.5} TOA	4.8	4.8	0.0	0	4.5	-0.3	-6	2.4	-2.4	-50	2.4	-2.4	-50

Table 5. Same as Table 4 but for CMAQ.

PM Species	base	1.5NO _x			0.5NO _x			0.5VOC			0.5NO _x + 0.5VOC		
	MC	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD	MC	MD	NMD
total PM _{2.5}	5.0	5.2	0.20	4	4.8	-0.20	-4	5.1	0.10	2	4.9	-0.10	-2
PM _{2.5} SO ₄	2.4	2.5	0.10	4	2.2	-0.20	-8	2.7	0.30	12	2.5	0.10	4
PM _{2.5} NO ₃	0.23	0.35	0.12	52	0.09	-0.14	-61	0.28	0.05	22	0.13	-0.10	-43
PM _{2.5} NH ₄	0.80	0.85	0.05	6	0.71	-0.09	-11	0.90	0.10	11	0.81	0.01	1
PM _{2.5} EC	0.32	0.31	-0.01	-3	0.33	0.01	3	0.33	0.01	3	0.34	0.02	6
PM _{2.5} TOA	1.0	0.98	-0.02	-2	0.98	-0.02	-2	0.71	-0.29	-29	0.72	-0.28	-28

The direction or sign of the change predicted for the three O₃ metrics was the same for all four emission scenarios with the exception of daily minimum O₃ for the “1.5NO_x” case, for which AURAMS predicted a decrease on average and CMAQ predicted an increase.

For surface-level PM_{2.5}, AURAMS was more sensitive to both NO_x and VOC emission changes. For the “1.5NO_x”, “0.5NO_x”, and “0.5NO_x+0.5VOC” emission scenarios, the responses of the models were in the same direction. However, for the “0.5VOC” scenario, AURAMS predicted a 15.5% reduction in total PM_{2.5} concentration whereas CMAQ predicted a 2.0% increase in total PM_{2.5} concentration.

This difference arose due to differences in the predicted response of individual PM_{2.5} major species. For the “0.5VOC” scenario, CMAQ predicted a much smaller decrease in PM_{2.5} TOA mass than did AURAMS and also increases in both PM_{2.5} SO₄ and PM_{2.5} NO₃ mass whereas AURAMS predicted a decrease in PM_{2.5} SO₄ mass and no net change in PM_{2.5} NO₃ mass.

These differences in PM_{2.5} response for the two models for a reduction in VOC emissions illustrate the significant coupling between gas-phase and particle chemistry, the significant influence of gas-phase oxidants, and the impact of differences in the parameterization of atmospheric organic chemistry. Results from several previous studies agree with the CMAQ-predicted changes in PM_{2.5} SO₄ and PM_{2.5} NO₃ mass for the “0.5VOC” case, but the CMAQ prediction of the change in PM_{2.5} TOA mass is likely too low, illustrating the uncertainty in total PM_{2.5} predictions due to the combined uncertainties of multiple PM formation pathways.

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