6B.3 EVALUATION OF A REGIONAL AIR-QUALITY MODEL (AURAMS) FOR TWO FIELD CAMPAIGN PERIODS OVER SOUTH-EASTERN CANADA AND U.S. NORTHEAST: IMPACT OF METEOROLOGY ON AIR QUALITY

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1. INTRODUCTION

The Environment Canada regional air quality modeling system, AURAMS, has been used to provide real-time forecast for several recent field campaigns. Two of the campaigns were conducted over eastern North America: the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) during the summer of 2004, mainly focused on air quality over the east coast, New England area, and the outflow to the North Atlantic (Fehsenfeld et al., 2006); and the Border Air-Quality Study - Meteorology (BAQSMet) which was conducted in the summer of 2007 over southern Ontario focused on the impact of lakebreeze meteorological conditions on air pollution in the region (Makar et al., 2009). Retrospective model runs over similar domains for the two campaign periods have since been conducted and detailed evaluation of model performance against field study data and network observations is being carried out.

The two summers (2004 and 2007) are quite different, in the sense that the summer of 2004 was characterized as been cooler and wetter than average over eastern North America (White et al., 2007) while the summer of 2007 was closer to average over the region. The ozone monitoring data from the AIRNOW network shows an average of 4 ppb difference between ICARTT period (lower) and the BAQSMet period (higher) both in terms of ozone1-hour daily maximum and daily mean. As for PM_{2.5} mass, although the average levels are comparable between the two periods, the speciated PM data from the IMPROVE network indicates a distinct difference in the composition of PM2.5 over the region, namely the sulfate fraction is significantly higher in the summer of 2004 than the summer of 2007.

* Corresponding author address: Wanmin Gong, Air Quality Research Division, Environment Canada, Toronto, Ontario, M3H 5T4, Canada; e-mail: wanmin.gong@ec.gc.ca This paper will focus on the comparison of model performance between the two periods, model's ability in capturing the impact of meteorology on air quality in the region as seen from the network monitoring data. The roles of different processes affecting ozone and PM in the region are discussed.



Figure 1 The 42- and 15-km resolution model domains for the ICARTT and BAQSMet runs.

2. SIMULATION SETUP AND EVALUATION DATA

AURAMS is a multi-pollutant, regional air-quality modeling system with size segregated and chemically speciated representation of aerosols (see Gong et al., 2006; McKeen et al., 2005; Smyth et al., 2009). The AURAMS version 1.4 was used to simulate the two field study periods in a cascading fashion, from 42- to 15- to 2.5-km resolutions, by one-way nesting. For the ICARTT study, the two coarser resolution runs (42- and 15km) were carried out for the period July 7 - August 19, 2004 and the 2.5-km resolution runs were carried out for selected flights (e.g., Gong et al., 2009). As for the BAQSMet campaign, the two coarse resolution runs were carried out for three summer months June 1 - August 31, 2007, and the high-resolution 2.5-km run was carried out for the intensive field study period of June 17 - July



Figure 2 Monitoring sites included in the model evaluation: $left - 831 (542) O_3$ sites from AIRNow; middle $- 322 (191) PM_{2.5}$ sites from AIRNow; right - 37 speciated PM sites from IMPROVE. Numbers in brackets denote the sites within the common 15-km resolution model domain.

11, 2007 over southern Ontario. This study will focus on the model simulations on the regional scale (i.e., 42- and 15-km resolution). Figure 1 shows the model domains for these simulations. Note that both the 42- and 15-km grids for the two study periods overlap one another but differ in domain size.

Being an off-line model, AURAMS is driven by a regional weather forecast model simulation (GEM; Cote et al, 1998). For this study, GEM version 3.2.2 with the additional recent parameterization for anthropogenic heat islands (Makar et al., 2006) was used. The GEM runs, in a regional configuration with a 15-km resolution in its uniform "core" centered over North America, were used to provide meteorological inputs to the AURAMS simulations at both 42- and 15-km resolutions.

The anthropogenic emission files for both model simulation periods were prepared from the 2005 U.S. and Canadian and 1999 Mexican inventories using version 2.3 of the SMOKE emission processing system (http://www.smoke-model.org/index.cfm). For the 42-km resolution runs, time-independent chemical lateral boundary conditions were used, including gridded monthly ozone climatology of Logan (1998) for O₃ initial and boundary conditions and the monthly MOPITT data

(http://neo.sci.gsfc.nasa.gov/Search.html?group=3 5) for CO.

The observational data used for model evaluation in this study include ozone and $PM_{2.5}$ mass from the AIRNow network (<u>http://airnow.gov/index.cfm</u>) and speciated PM mass from the IMPROVE network

(http://vista.cira.colostate.edu/views/Web/Data/Dat aWizard.aspx). Figure 2 shows the measurement sites available from these networks for the study. Only the sites over eastern North America are considered as they are commonly covered by the two 42-km model domains (Fig. 1). The AIRNow O₃ and PM_{2.5} are from continuous measurements and are reported hourly; the IMPROVE speciated PM_{2.5} measurements are from 24-hour filter samples collected 1 in 3 days.



Figure 3 Averaged modelled temperature over the two study periods: left panels for the ICARTT period; right panels for the BAQSMet period; top – daily mean; bottom – daily maximum.

3. RESULTS AND DISCUSSIONS

Model simulated O_3 and $PM_{2.5}$ are evaluated against network monitoring data. The evaluation period for the ICARTT simulation is July 14 – August 18, 2004. The first 7 days of the simulation are treated as model spin-up. For the BAQSMet simulation, the evaluation is carried out here for the period of July 1 – August 31, 2007 so that the two evaluation periods cover similar time of the year. Also, for comparison purpose, only the sites located within common domains are used for the evaluation.

As mentioned in the introduction, there is a marked difference between the two summers and this is reflected in modeled temperature shown in Figure 3. The averaged modeled daily mean and daily maximum temperature are considerably higher for the 2007 BAQSMet period than the 2004 ICARTT period.

3.1 Ozone

The model predicted daily maximum ozone mixing ratios from the 15-km runs, averaged over the two study periods, are shown in Figure 4, along with krigged corresponding observations from AIRNow. As seen, over eastern U.S. and south-eastern Canada the observed ozone levels are considerably higher for the 2007 BAQSMet period than the 2004 ICARTT period. The modeled ozone



Figure 4 Ozone daily maximum averaged over (left) the ICARTT period, 14/07/2004 - 18/08/2004, and (right) the BAQSMet period, 01/07/2007 - 31/08/2007; top – AIRNow observations (krigged); bottom – model results.



Figure 5 Mean bias in modeled daily maximum ozone for the two study period: left – ICARTT; right – BAQSMet.

levels are also higher for the BAQSMet period than the ICARTT period over the same region. There is, however, a general overprediction of ozone levels for both study periods. Figure 5 shows the mean bias in the modeled daily maximum ozone as compared to the AIRNow observations. The sites with overprediction are similarly located for the two study periods, mostly in precursor source regions, with areater overprediction for the BAQSMet period. The model-observation scatter plots of mean daily maximum ozone for the two periods are compared in Fiigure 6. The model is seen to do better in correlation coefficient for the BAQSMet period than for the ICARTT period, partly due to greater range. The slope is greater than 1 for the BAQSMet period and just under 1 for the ICARTT period.



Figure 6 Mean daily maximum O₃, model vs. observation, at AIRNow sites.

In addition to looking at the usual statistical evaluation metrics it is also interesting to see whether the model is capable of reproducing the observed ozone frequency distribution for a given period. Figure 7 compares the mean daily maximum ozone frequency distributions, both observed and modeled, for the two study periods. Note that the "frequency" here is in a spatial sense within the model domain since it is for daily maximum ozone at a given AIRNow site averaged over the study period. The observed frequency distribution for the ICARTT period is relatively



Figure 7 Frequency distribution of mean daily maximum O3 at the AIRNow sites: top – observed; bottom – modeled; left – ICARTT period; right – BAQSMet period.

narrow with a peak around the 50 - 60 ppbv range. In comparison, a wider distribution is observed for the BAQSMet period with a broader peak around the 50 - 70 ppbv range. The contrast between the two periods is qualitatively captured by the model in that the modeled frequency distribution is narrower for the ICARTT period and



Figure 8 PM_{2.5} daily mean averaged over (left) the ICARTT period, 14/07/2004 - 18/08/2004, and (right) the BAQSMet period, 01/07/2007 - 31/08/2007; top – AIRNow observations (krigged); bottom – model results.

broader for the BAQSMet period with the distribution extended to higher end. However the modeled distributions are broader than the respective observed distributions and shifted to higher end as indicated by the statistics shown.

3.2 PM_{2.5}

The averaged daily mean $PM_{2.5}$ concentrations from the observation (using krigging) and the model simulations at 15-km resolution are compared in Figure 8 between the two study periods. In contrast to ozone, the $PM_{2.5}$ concentration levels are comparable over the eastern North America between the two periods based on the observations. This is however not the case from the model simulations: the model predicted $PM_{2.5}$ concentrations are considerably higher for the BAQSMet period than the ICARTT



Figure 9 Mean bias in modeled daily mean $PM_{2.5}$ for the two study period: left – ICARTT; right – BAQSMet.

period. In general $PM_{2.5}$ concentrations are underpredicted for the ICARTT period and overpredicted for the BAQSMet period.

Figure 9 shows the mean bias in modeled daily mean $PM_{2.5}$ for the two study periods at the AIRNow sites. There are more sites where the model over-predicted $PM_{2.5}$ from the BAQSMet simulation than from the ICARTT run. Again the model-observation scatter plots of $PM_{2.5}$ daily mean are shown in Figure 10. Similar to the case of ozone, the modeled daily $PM_{2.5}$ also correlated better with the observations from the BAQSMet



Figure 10 Averaged daily mean PM_{2.5}, model vs. observation, at AIRNow sites.

run than from the ICARTT run.

Figure 11 compares the frequency distributions of

averaged daily mean PM_{2.5} over the two study periods. As seen, the observed frequency distributions are quite different with the one for the ICARTT period more skewed toward the lower end, though the mean and median of the observed distributions are comparable. The different skewness in the frequency distribution between the two periods seems to be picked up by the simulations correctly. However, model the modeled frequency distribution for the ICARTT period is shifted towards the lower end in comparison to the observed distribution corresponding to an overall under-prediction (or over-prediction of the low-concentration events). In contrast, the modeled frequency distribution for the BAQSMet period is flatter compared to the observed distribution indicating over-prediction of both the low-concentration and the hiahconcentration events with an overall overprediction in the averaged daily mean PM2.5 for this period.

3.3 Speciated PM_{2.5}

The modeled $PM_{2.5}$ components are compared to the IMPROVE speciated PM (fine) measurements. Here we focus on sulfate and organic matter components as they account for a main part of the total $PM_{2.5}$ mass regionally in the study area.



ICARTT: July 14 – Aug 18, 2004

BAQSMet: July 1 – Aug 31, 2007

Figure 11 Frequency distribution of averaged daily mean $PM_{2.5}$ at the AIRNow sites: top – observed; bottom – modeled; left – ICARTT period; right – BAQSMet period.

While sulfate is mainly secondary formed from SO₂ oxidation, both primary emission and secondary formation/production contribute significantly to the organic matter component.

Figure 12 shows the side-by side comparisons of observed and modeled sulfate_{2.5} at the IMPROVE sites over the two periods. The model values are obtained by sampling the model results for the same 24-hour sampling period and the sampling days (1 in 3) as the observations. The sites are arranged on the x-axis from west to east, or from sites located (generally) closer to the sources (of SO_2 , in particular) to the sites farther away from the sources.



Figure 12 Observed and modeled sulfate_{2.5} at the IMPROVE sites (within the common 15-km domain) averaged over the ICARTT period (top) and BAQSMet period (bottom).

Table 1 Averaged sulfate $_{2.5}$ (µg m⁻³) at all IMPROVE sites within the common 15-km model domain.

	ICARTT		BAQSMet	
	Obs.	Mod.	Obs.	Mod.
mean	5.3	4.6	4.8	5.7
median	5.4	4.4	5.4	5.9
std	2.1	2.9	2.3	3.5

The model seems to over-predict sulfate at sites close to the sources. For example at Quaker City and Frostburg sites, both close to major point sources, sulfate is significantly over-predicted for both periods indicating possible problem with emission input or the treatment of emission from the sources. The over-prediction is much more significant (in magnitudes and at more sites) for the BAQSMet period. For the sites on the east, farther away from the sources, sulfate is considerably under-predicted for the ICARTT period in contrast to the BAQSMet period. Table 1 presents the mean, median, and standard deviation for the two periods for the observed and modeled sulate_{2.5}. It is seen, on average, the observed sulfate is higher for the ICARTT period than the BAQSMet period, while the opposite is predicted by the model.

Similar bar charts comparing observed and modeled organic matter component $(OM_{2.5})$ are presented in Figure 13. One thing stands out is that the model under-predicted $OM_{2.5}$ at all sites for the ICARTT period while over-predicted at all sites for the BAQSMet period. In general the predicted $OM_{2.5}$ concentrations are much higher for the BAQSMet period, while the differences between the two periods are small from the observations, (as in mean, median, and standard deviation shown in Table 2). This will be examined further below in 3.5.



Figure 13 Observed and modeled $OM_{2.5}$ at the IMPROVE sites (within the common 15-km domain) averaged over the ICARTT period (top) and BAQSMet period (bottom).

Table 2 Averaged $OM_{2.5}$ (µg m⁻³) at all IMPROVE sites within the common 15-km model domain.

	ICARTT		BAQSMet	
	Obs.	Mod.	Obs.	Mod.
mean	2.8	1.9	2.6	4.1
median	2.8	1.8	2.5	3.6
std	0.7	0.7	0.9	2.1

3.4 Sensitivity to Changes in Emission under the NOx Budget Trading Program



Figure 14 Adjustment ratios used for state-wide adjustment to 2005 NOx emission from major point sources for the BAQSMet sensitivity run.

When comparing the model performance between the two summer periods in 2004 and 2007, one of the things to be considered is the changes in emission between the years. As mentioned in section 2, the 2005 Canada and U.S. inventories are used for generating the anthropogenic emission input for simulating both summer periods. A sensitivity test was carried out to assess the impact from some "known" changes in NOx emission from large energy generating utilities (EGUs) in the U.S. northeast due to the U.S. EPA NOx Budget Trading Program (NBP) on



Figure 15 Difference in modeled ozone with vs. without the adjustment to account for the emission change due to NBP between 2007 and 2005 in averaged daily maximum (left) and in averaged daily mean (right).

model performance, ozone particularly. A more elaborated way of implementing the changes would be to make use of the Continuous Emission Monitoring System (CEMS) measurements (e.g., Frost et al., 2006). In this study, a much simpler approach was used. State-wide ratios of ozone season NOx emission from all NBP EGUs between 2007 and 2005 were computed based on the published 2007 EPA report on program compliance and environmental results (http://www.epa.gov/airmarkt/progress/nbp07.html). These ratios (as shown in Figure 14) were used to adjust NOx emissions from major point sources in the NBP participating states.



Figure 16 Contribution to sulfate_{2.5} from aqueous-phase oxidation: left – averaged sulfate2.5 over the study periods; middle – difference in averaged sulfate_{2.5} ("with" – "without" aqueous oxidation); right – relative difference in averaged sulfate_{2.5}; top - ICARTT; bottom – BAQSMet.

Figure 15 shows the differences in the model prediction of ozone (in averaged daily maximum and daily mean) for the BAQSMet period, with and without the adjustment to NOx emission from major point sources in the NBP participating state from 2005 to 2007. The differences are small, mostly within ± 1 ppbv in averaged daily maximum and ± 0.5 ppbv in averaged daily mean regionally, with some localized larger differences in immediate vicinity of the point sources. This result seems to suggest that meteorology is the main factor for the observed difference in ozone between the two summer periods over the region, as the model is shown to qualitatively reproduce the contrast in observed ozone between the two periods (e.g. Fig. 7) with only the difference in meteorology.

3.5 Secondary Aerosol Production

As shown in 3.3 comparing modeled and observed speciated $PM_{2.5}$, there are some significant differences in model performance between the two periods. We will look into these differences further in this section.

Sulfate

The model is shown to under-predict sulfate_{2.5} for the ICARTT period but over-predict for the BAQSMet period. A sensitivity test was carried out to switch off the aqueous-phase (in-cloud) oxidation in order to see the relative contributions from the clear-air and aqueous-phase oxidation to the regional sulfate. The results are shown in Figure 16, including the base-case concentration (averaged over the study periods), the difference and relative difference in modeled sulfate between the base-case (with aqueous-phase oxidation) and the sensitivity test (without the aqueous-phase oxidation). As seen, the aqueous-phase oxidation played a much bigger role during the ICARTT than during the BAQSMet period over south-eastern Canada and eastern U.S., in consistent with the wetter and more cloudy summer in 2004 than in 2007. The higher sulfate predicted by the model for the BAQSMet period is more likely to be a result of enhanced clear-air oxidation during the period.

Organic matters

As seen from Figure 13 and Table 2, the modeled $OM_{2.5}$ is significantly higher on average for the BAQSMet period than the ICARTT period, while the opposite is shown by the observations. Figure



Figure 17 Model-predicted primary (top) and secondary (bottom) organic matter component averaged over the two periods: left – ICARTT (14/07/2004 – 18/08/2004); right – BAQSMET (01/07/2007 – 31/08/2007).

17 shows the modeled primary and secondary organic component for the two periods. Clearly the higher modeled OM_{2.5} for the BAQSMet period is mainly due to higher modeled secondary organic aerosol (SOA) component for the period. The comparable modeled primary organic component for the two periods is consistent with similar emission inputs (both based on the same inventory data). The main difference between the two periods, as far as the model simulations are concerned, is the meteorology. From examining modeled SOA precursor fields, it is found that isoprene in the model simulation is much higher for the BAQSMet period than the ICARTT period (not shown), and the higher isoprene regions correspond well with the higher modeled SOA regions for the BAQSMet period shown in Figure 17. This finding indicates that the model is producing more organic aerosol mass through higher biogenic SOA production for the BAQSMet period, a considerably warmer summer period in 2007 than the summer in 2004.

4. SUMMARY AND CONCLUSIONS

A comparative operational performance evaluation of the Environment Canada regional air quality model (AURAMS) is carried out for two different summer periods, corresponding to the 2004 ICARTT and the 2007 BAQSMet field campaigns over eastern North America. The two periods are characterized by marked difference in meteorological conditions. The summer of 2004 was considerably cooler and wetter than average over eastern North America whereas the summer of 2007 was closer to average over the region. The model simulations are evaluated against surface ozone and $PM_{2.5}$ monitoring data from AIRNow and PM sulfate and organic matters from IMPROVE. Preliminary findings include the following:

(1) The difference in ozone over eastern North America between the summers of 2004 and 2007 is mainly due to meteorology.

(2) The model is biased high in predicting regional ozone. The over-prediction is greater for the BAQSMet period in summer 2007 than the ICARTT period in summer 2004.

(3) Modelled sulfate is higher for the 2007 BAQSMet period than the 2004 ICARTT period, while the opposite is indicated from observations. The higher modeled sulfate for the BAQSMet period seems to be contributed by clear-air oxidation.

(4) The model under-predicted organic aerosol component for the 2004 ICARTT period but overpredicted for the 2007 BAQSMet period. The higher modeled $OM_{2.5}$ for the BAQSMet period is mainly due to secondary formation/production, specifically through biogenic SOA production in the model.

Work is underway to test improved lateral and upper ozone boundary condition which will address part of ozone bias. The SOA scheme in the model is also being updated with better representation of SVOC/IVOC.

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