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EVALUATION OF 2005 MULTI-POLLUTANT PLATFORM: AIR TOXICS, OZONE, AND PARTICULATE MATTER

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Abstract: An annual 2005 multi-pollutant model application and evaluation study was performed for the continental United States using a multi-pollutant version of the U.S. EPA's Community Multi-scale Air Quality (CMAQ) modeling system at 36-km and 12-km grid resolutions. The CMAQ multi-pollutant v4.7 was used to predict ozone, particulate matter, mercury, and 38 other hazardous air pollutants (HAPs) within one model simulation. The focus of this effort is the evaluation of model predictions of toxics species using available 2005 measured data. Model evaluation is also conducted for ozone, $PM_{2.5}$ component species, and deposition of sulfate and nitrate. This paper will also examine model predictions to gain a better understanding of the chemical and physical interactions between concentration and deposition of ozone, $PM_{2.5}$ component species, toxic species, and precursor gases (including gaseous toxics) as well as their temporal and spatial relationships.

***Key words**: multi-pollutant, model evaluation, criteria air pollutants, hazardous air pollutants*

INTRODUCTION

Traditionally, air quality assessments have been conducted within a framework that simulates only criteria air pollutants (CAPs). In recent years, increasing attention has been given to the development and application of a multi-pollutant (MP) framework, including both CAPs and hazardous air pollutants (HAPs). An air quality modeling system with MP treatments and interactions provides an essential tool to replicate the complex atmosphere and thus to assess regulatory programs and applications. To apply and evaluate a MP modeling platform, a 2005 annual simulation was conducted by the U.S. Environmental Protection Agency's (EPA) Office of Air Quality Planning and Standards utilizing a MP version of the U.S. EPA's Community Multi-scale Air Quality (CMAQ) modeling system with horizontal grid resolutions of 12-km over the Eastern U.S. (EUS) and Western U.S. (WUS) as well as 36-km over the continental U.S. (CONUS) (Figure 1). Model evaluation for concentrations and depositions of ozone (O_3) and its precursors, fine particulate matter ($PM_{2.5}$) and its speciated components (e.g., sulfate, nitrate, elemental carbon, organic carbon, etc.), and HAPs has been performed with available surface monitoring networks in order to evaluate the predictive capabilities of CMAQ to reproduce the atmospheric processes resulting in formation and dispersion of air pollution.

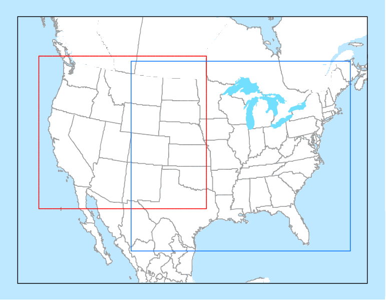

Figure 1. Map of the CMAQ modeling domain. The black outer box denotes the 36 km national modeling domain; the red inner box is the 12 km western U.S. fine grid; and the blue inner box is the 12 km eastern U.S. fine grid.

MODELING PLATFORM CONFIGURATION

CMAQ is a non-proprietary computer model that simulates the formation and fate of photochemical oxidants, primary and secondary PM concentrations, acid deposition, and air toxics, over regional and urban spatial scales for given input sets of meteorological conditions and emissions (Byun and Schere, 2006). This 2005 multi-pollutant modeling platform used the latest publicly-released CMAQ version 4.7 at the time of modeling (http://www.cmascenter.org). The model extends vertically from the surface to 100 millibars (approximately 15 km) using a sigma-pressure coordinate system. The lateral boundary and initial species concentrations for the 36 km domain are provided by a three-dimensional global atmospheric chemistry model, the GEOS-CHEM model, standard version 7-04-11 (Yantosca, 2004 and Henze *et al.*, 2008). The 36 km and both 12 km CMAQ modeling domains were modeled for the entire year of 2005 including 10 days at the end of December 2004 as a modelled "ramp up" period. These days are used to minimize the effects of initial conditions and are not considered as part of the output analyses. The conditions from the 36-km coarse grid modeling were used as the initial/boundary state for all subsequent 12-km domains. The emissions data used in the 2005 base year are based on the 2005 v4 platform (U.S EPA, 2010a). The gridded meteorological input data for the entire year of 2005 were derived from simulations of the Pennsylvania State University / National Center for Atmospheric Research Mesoscale Model version 3.7.4. This model, commonly referred to as MM5, is a limited-area, nonhydrostatic, terrain-following system that solves for the full set of physical and thermodynamic equations which govern atmospheric motions (Grell *et al.*, 1994). The MM5 simulations were run on the same map projection as CMAQ. Details on the configuration of the meteorological model runs can be found at U.S. EPA, 2010b. The meteorological outputs from all three MM5 sets were processed to create model-ready inputs for CMAQ using the Meteorology-Chemistry Interface Processor (MCIP), version 3.4, to derive the specific inputs to CMAQ.

METHODOLOGY

This evaluation principally comprises statistical assessments of model versus observed pairs that were paired in space and time on a daily or weekly basis, depending on the sampling frequency of each available 2005 surface monitoring networks (measured data). For certain time periods with missing ozone, $PM_{2.5}$ and air toxic observations we excluded the CMAQ predictions from those time periods in our calculations. In conjunction with the model performance statistics, we also provide spatial plots for individual monitors of the calculated normalized mean bias statistics (defined below). For this extended abstract, we focus model performance on eight-hour maximum daily ozone, sulfate and nitrate and particular HAPs (acetaldehyde, formaldehyde, and benzene) for the 12-km EUS and WUS, as well as five large subregions: Midwest, Northeast, Southeast, Central, and West U.S (see US EPA, 2009 for subregions). The Atmospheric Model Evaluation Tool (AMET) was used to conduct the evaluation described in this document (Gilliam *et al.*, 2005).

There are various statistical metrics available and used by the science community for model performance evaluation. For a robust evaluation, the principal evaluation statistics used to evaluate CMAQ performance were two bias metrics, normalized mean bias and fractional bias; and two error metrics, normalized mean error and fractional error (see Appel *et al.*, 2005 for equation definitions). The "acceptability" of model performance was judged by comparing our CMAQ 2005 performance results to the range of performance found in recent regional ozone, $PM_{2.5}$, and HAPs model applications (US EPA, 2005; US EPA, 2006; US EPA, 2009; Appel *et al.*, 2008; Phillips *et al.*, 2008; Strum *et al.*, 2008;"). Overall, the NMB, NME, FB, and FE statistics shown below for CMAQ predicted 2005 ozone, $PM_{2.5}$, and HAPs concentrations are within the range or close to that found in recent applications. The CMAQ model performance results give us confidence that our applications of CMAQ using this 2005 modelling platform provide a scientifically credible approach for assessing ozone and $PM_{2.5}$ concentrations.

RESULTS

Eight-hour Daily Maximum Ozone Performance: Threshold of 60 ppb

The ozone evaluation primarily focuses on observed and predicted eight-hour daily maximum ozone concentrations at a threshold of 60ppb. This ozone model performance was limited to the ozone season modelled of May, June, July, August, and September. Ozone ambient measurements for 2005 were obtained from the Air Quality System (AQS) Aerometric Information Retrieval System (AIRS). A total of 1194 ozone measurement sites were included for evaluation. Table 2 presents eight-hour daily maximum ozone model performance bias and error statistics for the range of observed and modelled concentrations at a threshold of 60 ppb and above for the 12-km EUS and WUS domain and the corresponding sub-regions defined above. Spatial plots of the NMB statistic (units of percent) for individual monitors based on the aggregate of the ozone season modelled are shown in Figure 2. CMAQ consistently under-predicts eight-hour daily maximum ozone at the higher end of the ozone distribution (≥ 60 ppb) during all the individual months in the ozone season and subsequently the seasonal aggregate. Likewise, certain areas in the WUS, e.g. Southern California, Montana, Wyoming, Utah, etc. consistently under-predict on average during the ozone season, 20-30%. Although not shown here, high winter ozone in the WUS is also under-predicted. Hence, improvements are needed to be able to simulate processes and chemistry related to albedo, snow cover, winter photochemistry, and nitrogen species. For the 12-km EUS domain, the bias statistics are within the range of approximately -6% to -16%, while the error statistics range from 10% to 18% for the aggregate of the ozone season and for most of the months modelled. For the 12-km WUS domain, the bias statistics are within the range of approximately -7% to -8%, while the error statistics range from 13% to 14% for the aggregate of the ozone season and for the individual months modelled. The five sub-regions show relatively similar eight-hour daily maximum ozone performance.

Table 2. 2005 CMAQ eight-hour daily maximum ozone model performance statistics calculated for a threshold of 60 pbb.

CMAQ 2005 Eight-Hour Maximum Ozone: Threshold of 60 ppb		No. of Obs.	NMB (%)	NME (%)	FB (%)	FE (%)
Seasonal Aggregate (May – September)	12-km EUS	31271	-7.7	11.7	-8.5	12.4
	12-km WUS	14706	-6.8	13.2	-7.8	14.0
	Northeast	5968	-5.9	11.3	-6.4	9.7
	Midwest	7858	-7.2	10.5	-7.8	11.1
	Southeast	3577	-14.2	15.9	-16.3	17.9
	Central	6472	-12.9	14.9	-14.4	16.4
	West	12446	-6.8	13.7	-7.9	14.5

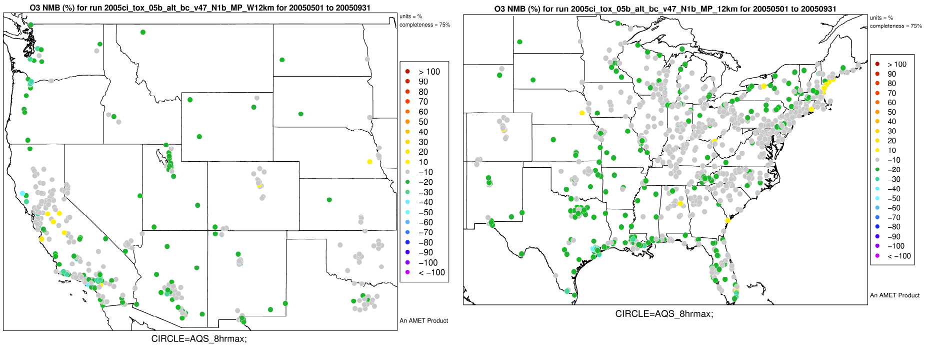

Figure 2. NMB (%) of eight-hour daily maximum ozone (60 ppb threshold) by monitor for WUS and EUS during 2005 ozone season.

Seasonal Sulfate Performance

Ambient measurements of sulfate PM for 2005 were obtained from the following networks for model evaluation: **S**peciation **T**rends **N**etwork (STN- total of 260 sites), **I**nteragency **M**onitoring of **PRO**tected **V**isual **E**nvironments (IMPROVE- total of 204), **C**lean **A**ir **S**tatus and **T**rends **Net**work (CASTNet- total of 93). Overall, CMAQ under-predicts sulfate in the 12-km EUS and WUS domains. Sulfate predictions during the summer season are moderately under-predicted in the EUS and WUS across the available monitoring data (NMB values range from -11% to -38% (Table 3). Spatial plots of the NMB statistic (units of percent) for individual monitors in the EUS and WUS during the summer season are also provided in Figure 3. The model tends to show better performance in the EUS when sulfate is the dominant $PM_{2.5}$ species during the summer season.

Table 3. CMAQ 2005 summer season (June-July-August) model performance statistics for sulfate.

CMAQ 2005 Sulfate			No. of Obs.	NMB (%)	NME (%)	FB (%)	FE (%)
Summer	STN	12-km EUS	3516	-18.6	32.5	-17.6	38.9
		12-km WUS	1075	-32.7	41.8	-26.3	43.7
		Northeast	874	-11.2	28.2	-5.0	31.9
		Midwest	621	-13.2	29.7	-2.7	31.3
		Southeast	941	-21.0	33.1	-21.9	38.7
		Central	847	-36.2	39.1	-42.1	48.4
		West	853	-33.8	45.1	-25.4	44.7
	IMPROVE	12-km EUS	2324	-22.6	35.4	-19.0	42.1
		12-km WUS	2395	-29.2	42.0	-21.1	45.6
		Northeast	590	-11.2	28.2	-5.0	31.9
		Midwest	158	-21.3	30.9	-9.4	34.8
		Southeast	427	-26.5	35.5	-27.7	43.2
		Central	601	-30.1	39.5	-26.1	45.9
		West	2121	-28.2	43.0	-20.0	45.9
	CASTNet	12-km EUS	792	-22.4	25.7	-25.3	31.7
		12-km WUS	295	-38.1	41.2	-40.2	45.8
		Northeast	192	-17.9	22.0	-14.5	24.1
		Midwest	161	-18.6	23.0	-16.8	25.1
		Southeast	270	-24.6	26.9	-31.9	35.0
		Central	75	-36.2	38.9	-42.5	48.3
		West	282	-38.2	41.6	-40.1	45.9

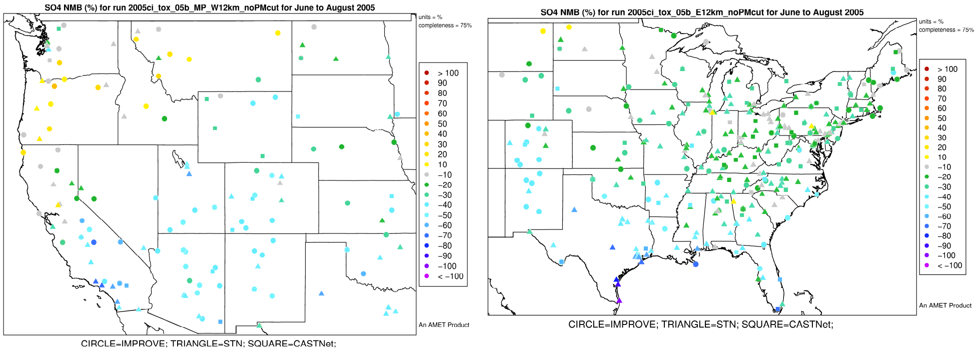

Figure 2. NMB (%) of 2005 summer sulfate by monitor for EUS and WUS.

Seasonal Nitrate Performance

Similar to sulfate PM, ambient data for nitrate PM was obtained from the STN, IMPROVE, and CASTNet networks for model evaluation. Table 4 provides the seasonal model performance statistics for nitrate and total nitrate for the 12-km EUS and WUS domains. Spatial plots of the NMB statistics for individual monitors in the EUS and WUS are provided as a complement to the tabular statistical data (Figures 4-5). The model appears to simulate the winter average of total nitrate (nitrate PM and nitric acid, HNO_3) fairly well when nitrate is most abundant, but the partitioning between nitrate PM and HNO_3 leads to modest scatter in predictions of nitrate PM. Nitrate performance at STN sites is under-predicted in the EUS (NMB ~ -10%) and WUS (NMB ~ - 40%) except in the Northeast where the model slightly over-predicted nitrate on average ~14%. Likewise, nitrate performance at IMPROVE sites in the WUS is moderately under-predicted whereas performance is mixed in the EUS (under-predicted in the Midwest and Central US and over-predicted in the Southeast and Northeast). Yu *et al.* (2005) showed that a large source of error in predicting aerosol NO_3 across the EUS stems from errors in the model predictions of NH_x (NH_4^++NH_3), SO_4^{2-}, and, to a lesser extent, TNO_3 (NO_3^-+HNO_3). Overall, total nitrate is over-predicted in the EUS and WUS (NMB -10% to -24%) except in the Midwest (NMB ~ -6%). Over-predictions of TNO_3 have been shown to occur due to overestimated NH_3 emissions and higher values of the N_2O_5 uptake coefficient (Appel *et al.*, 2008).

Table 4. CMAQ 2005 winter season (December-January-February) model performance statistics for nitrate.

CMAQ 2005 Nitrate			No. of Obs.	NMB (%)	NME (%)	FB (%)	FE (%)
Nitrate (Winter)	STN	12-km EUS	3099	-5.7	46.0	-12.6	58.0
		12-km WUS	973	-40.1	61.7	-50.9	81.7
		Northeast	829	14.1	46.1	12.6	47.7
		Midwest	598	-12.6	37.0	-9.8	42.8
		Southeast	963	-10.8	59.0	-33.3	74.3
		Central	479	-10.9	46.3	-6.4	56.1
		West	831	-44.4	63.9	-58.5	85.6
	IMPROVE	12-km EUS	2076	2.3	59.3	-25.7	83.9
		12-km WUS	2426	-30.1	61.2	-85.5	113.3
		Northeast	502	45.2	74.4	31.4	73.1
		Midwest	129	-24.4	41.5	-30.9	63.2
		Southeast	386	8.9	79.6	-37.4	85.8
		Central	539	-7.5	49.7	-16.6	71.4
		West	2176	-42.0	70.2	-89.5	119.1
Total Nitrate (Winter)	CASTNet	12-km EUS	760	10.5	27.9	16.9	31.9
		12-km WUS	267	13.1	40.4	26.4	49.7
		Northeast	193	24.5	30.9	30.1	34.2
		Midwest	142	-6.0	19.9	0.4	20.4
		Southeast	264	17.7	31.4	14.5	32.1
		Central	72	11.4	30.3	13.3	31.1
		West	255	14.4	47.5	27.2	51.4

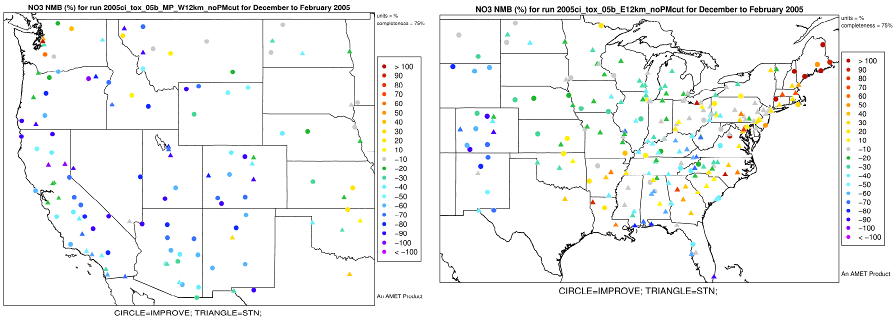

Figure 4. NMB (%) of 2005 winter nitrate by monitor for EUS and WUS.

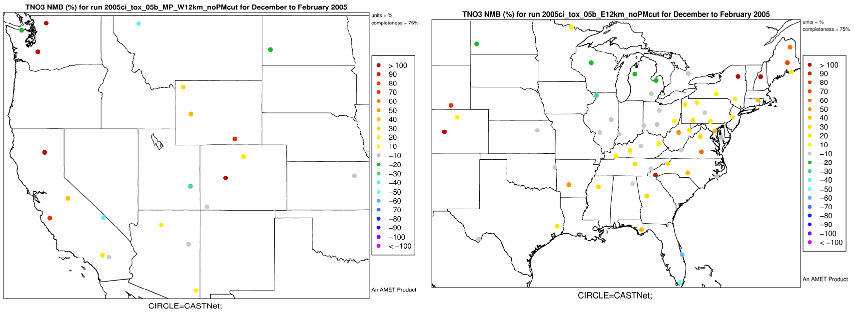

Figure 5. NMB (%) of 2005 winter total nitrate (NO_3+HNO_3) by monitor for EUS and WUS.

Annual Hazardous Air Pollutants Performance

For this paper, the air toxics evaluation focuses on the annual average of specific species, i.e., formaldehyde, acetaldehyde, and benzene. Toxic measurements for 2005 were obtained from the National Air Toxics Trends Stations (NATTS) with 471 sites in the EUS and 135 sites in the WUS. Although model performance for these non-ubiquitous HAPs is not as good as model performance for ozone and $PM_{2.5}$, model predictions of annual formaldehyde, acetaldehyde and benzene showed a general under-prediction tendency when compared to observations. Technical issues in the HAPs data consist of (1) uncertainties in monitoring methods; (2) limited measurements in time/space to characterize ambient concentrations ("local in nature"); (3) commensurability issues between measurements and model predictions; (4) emissions and science uncertainty issues may also affect model performance; and (5) limited data for estimating intercontinental transport that effects the estimation of boundary conditions (i.e., boundary estimates for some species are higher than predicted values inside the domain).

Table 5. CMAQ 2005 annual model performance statistics for air toxics.

CMAQ 2005 Annual HAPs		No. of Obs.	NMB (%)	NME (%)	FB (%)	FE (%)
Formaldehyde	12-km EUS	6365	-55.5	65.3	-39.2	65.6
	12-km WUS	1928	-28.4	52.1	-30.1	60.7
	Northeast	771	-77.1	85.4	-25.8	74.0
	Midwest	1982	-30.5	51.3	-28.5	61.6
	Southeast	1246	-66.2	72.2	-51.3	70.4
	Central	1815	-43.5	51.0	-41.4	61.5
	West	1746	-25.5	52.3	-26.0	59.8
Acetaldehyde	12-km EUS	6094	-4.2	62.0	-8.2	60.3
	12-km WUS	1892	-19.2	53.7	-19.5	59.6
	Northeast	703	-12.6	58.0	-12.1	60.0
	Midwest	1969	-9.5	62.8	-9.0	63.7
	Southeast	1231	0.4	63.5	-6.2	62.2
	Central	1640	1.8	57.0	-4.3	51.1
	West	1709	-20.4	54.1	-20.1	60.6
Benzene	12-km EUS	11615	-32.6	66.8	-13.5	62.8
	12-km WUS	3369	-38.4	60.8	-30.4	63.9
	Northeast	1425	-8.3	72.7	25.2	62.4
	Midwest	2589	21.6	53.3	18.1	46.8
	Southeast	2426	-41.1	68.6	-17.2	59.8
	Central	4737	-47.0	68.3	-32.7	69.4
	West	2333	-30.5	61.2	-19.2	63.4

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