Regulatory Modeling System Performance in the Upper Midwest for 3 Annual Simulations at 36 km for PM 2.5 and 3 Summer Seasons at 12 km for Ozone

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1. INTRODUCTION

The United States Environmental Protection Agency (EPA) recently passed the Regional Haze rule to improve visibility in Class I areas by 2060. Additionally, many counties will be classified as nonattainment for the National Ambient Air Quality Standard (NAAQS) for the annual PM2.5 and 8-hr ozone standards. Existing scientific evidence shows that regional haze, fine particles, and ozone have common precursor pollutants, emission sources, and atmospheric processes (vanloon et al, 2000). EPA guidance recommends the application of 3-D Eulerian grid models for an entire calendar year to fully capture the seasonal variation in PM2.5 formation processes in various regions of the United States (EPA 2001; EPA 1999).

Model performance is examined for chemically speciated PM2.5 rather than total PM2.5 mass because both PM2.5 NAAQS and Regional Haze modeling guidance stipulates the use of relative reduction factors by PM2.5 chemical specie to express changes in future-year design values (EPA 2001; EPA 2003a). The modeling system should be able to adequately capture the seasonal differences in ambient particulate concentrations as well as the year-to-year variability (Seigneur et al, 2000).

The modeling system performance for 3 consecutive summers over the upper midwest is examined for ozone performance. The summers of 2001, 2002, and 2003 were selected since observation data from these summers was used to classify the attainment status of counties.

2. METHODS

The Comprehensive Air Quality Model with Extensions (CAMx) version 4.20 uses state of the science routines to model ozone and particulate matter formation and removal processes over a large modeling domain. The model is applied with ISORROPIA inorganic chemistry, SOAP organic chemistry, regional acid deposition model (RADM) aqueous phase chemistry, and an updated carbon-bond IV (CB4) gas phase chemistry module (ENVIRON, 2005)

Meteorological input data for the photochemical modeling runs are processed using the National Center for Atmospheric Research (NCAR) 5th generation Mesoscale Model (MM5) version 3.6 (Dudia et al, 1993; Grell et al, 1994). Important MM5 parameterizations and physics options include mixed phase (Reisner 1) microphysics, Kain-Fritsch 2 cumulus scheme, Rapid Radiative Transfer Model, Pleim-Chang planetary boundary layer, and the Pleim-Xiu land surface module. Surface and 3-D analysis nudging for temperature and moisture are only applied above the boundary layer. Analysis nudging of the wind field was applied above and below the boundary layer. These parameters and options are selected as an optimal configuration for the Upper Midwest based on multiple sensitivity simulations (Johnson, 2003; Baker, 2004).

The point and area source inventories are based on the 2002 National Emission Inventory. On-road emissions are estimated using MOBILE6. Default temporal tables are modified to represent a more complex distribution of vehicle miles traveled for the weekend. Off-road emissions are estimated with EPA's NONROAD 2004 (released April 2004) model. Biogenic emissions are estimated with EMS-2003 using

BIOME3/BEIS3 and the BELD3 land use dataset. Other inputs to the biogenic emissions model include hourly satellite photosynthetically activated radiation (PAR) and 15 m (above ground level) temperature data output from MM5. Ammonia emissions are based on the latest version of Carnegie Mellon University's (CMU) ammonia model (July 2004 version) using 2002 census of agriculture data.

Emissions data are processed using EMS-2003 (Janssen et al, 1998; Wilkinson et al, 1994). Outputs from EMS-2003 include a coordinate-based elevated point source file and gridded emissions estimates for low-point, area, mobile, and biogenic sources. The biogenic emissions are day-specific. Anthropogenic emission estimates are made for a weekday, Saturday, and Sunday for each month. The anthropogenic emissions inputs for CAMx for 2001 and 2003 are not adjusted to reflect growth and control from the 2002 inventory.

The photochemical model uses 11 land use categories to describe the surface. The land use file is based on BELD3 1 km data. The 1 km data was aggregated to 12 and 36 km grid resolution for photochemical modeling. Surface roughness varies by season and land use category, ranging from 0.0001 m for water land use and 1.30 m for forested land use.

CAMx4 is applied with day specific photolysis rate look-up tables. The Tropospheric Ultraviolet-Visible (TUV) radiation model is used to calculate photolysis rates based on solar zenith angle, height above ground, ultraviolet albedo of the ground, atmospheric turbidity, and total ozone column density. The TUV generates rates for each day as a function of 11 heights, 10 solar zenith angles, 5 ozone column values, 5 albedo values, and 3 turbidity values (ENVIRON, 2005). The ozone column data is derived from daily TOMS satellite observations. The albedo data varies by month and is based on over 10 years of TOMS satellite reflectivity observations. The actinic flux is estimated using the discrete ordinate algorithm. The two-stream delta-Eddington method is also available in the TUV model, but was not selected because the discrete ordinate approach is more accurate.

Boundary conditions represent pollution inflow into the model from the lateral edges of the grid and initial conditions provide an estimation of pollution that already exists. In the past a spin-up period of two to three days was used to eliminate initial condition effects for ozone modeling. CAMx4 ozone source apportionment runs show ozone attributed to initial concentrations does not exceed 5 ppb anywhere in the domain by the seventh day of the episode, so the first seven days of the modeling episode are not used for model performance evaluation. CAMx4 particulate source apportionment (PSAT) runs show PM2.5 sulfate ion, nitrate ion, and ammonium ion contributions from initial concentrations reduce below 0.05 $\mu g/m^3$ by the seventh day of the episode. PM2.5 elemental carbon, PM2.5 soil, and coarse mass have less than 1 ng/m^3 contribution from initial concentrations on the first day of the model episode everywhere in the modeling domain. The annual simulations have two weeks of spin-up to minimize initial condition influence. The initial and boundary conditions are based on monthly averaged species output from an annual application of the GEOS-CHEM global chemical transport model (Bey et al, 2001). Where an initial or boundary concentration is not specified for a pollutant the model will default to a near-zero concentration.

All models are applied with a Lambert projection centered at (-97, 40) and true latitudes at 33 and 45. The photochemical modeling domain consists of 97 cells in the X direction and 90 cells in the Y direction covering the central and eastern United States with 36 km grid cells (Figure 1). The 2-way nested 12 km photochemical grid is shown by the dark gray box and covers most of the upper midwest region. CAMx4 is applied with the vertical atmosphere resolved with 16 layers up to approximately 15 km above ground level. The meteorological modeling domain covers the entire continental United States with a 36 km grid, consisting of 165 cells in the X direction and 129 cells in the Y direction. MM5 is configured to use 34 vertical layers to

resolve up to approximately 15 km above ground level.

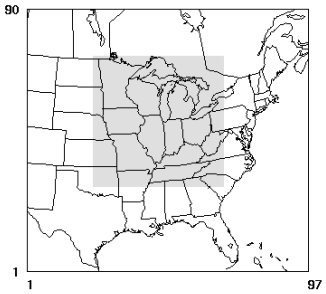

Figure 1. CAMx4 36 km (white box) modeling domain and 2-way nested 12 km domain (gray box) covering the upper midwest region

The meteorological model 12 km grid was applied as a two-way nest with no feedback to the coarse 36 km grid. The photochemical 12 km grid was applied as a two-way nest with feedback to the coarse 36 km grid. The 2-way nesting mode allows for interaction between the larger coarse grid with the smaller fine grid. This improves pollutant transport around the boundaries of the fine grid since a parcel of air may move from the fine grid, out to the coarse grid, and back into the fine grid depending on the shifting wind fields. This re-circulation is impossible in 1-way nesting applications.

Output from the chemical transport model is compared to 24 hr averaged chemically speciated PM2.5 measurements taken from a variety of networks: IMPROVE, EPA Speciation Trends, and CASTNET Visibility. PM2.5 ammonium ion is only measured at EPA Speciation Trends locations so the model performance for this chemical specie is dominated by, but not limited to, urban measurement locations.

Model performance evaluation methodology for PM2.5 and Regional Haze is described in the EPA document “Guidance for Demonstrating Attainment of Air Quality Goals for PM2.5 and Regional Haze” (US EPA, 2003). The guidelines describing good model performance for chemically speciated PM2.5 are based on a few early modeling applications that were limited in domain and episode length. For these reasons, the suggested guidelines for model performance to support regulatory applications are not included in this analysis.

Performance metrics used to describe model performance for PM2.5 species include mean bias, gross error, fractional bias, and fractional error (EPA, 2001; EPA, 1999). The bias and error metrics are used to describe performance in terms of the measured concentration units ($\mu g/m^3$). PM2.5 chemical specie concentrations are skewed toward very small values, but some species have fairly large concentrations in particular seasons, making the fractional metrics useful.

Hourly running 8-hour averaged surface ozone observations from EPA’s AIRS database are matched to hourly running 8-hour averaged layer 1 (30 m height) model estimates for evaluation. Only monitors in the 12 km modeling domain are included in the analysis. Model performance evaluation plots and metrics are based on matching predictions and observations in time and space. EPA has suggested several statistical metrics with suggested values representing model performance good enough to support regulatory applications. These metrics and guidelines are outlined in the EPA’s “Draft Modeling Guidance for Ozone Attainment Purposes” and include mean normalized bias error (MNBE) and mean normalized gross error (MNGE) (EPA, 1999). EPA guidance suggests MNBE should fall between below 15% and MNGE below 35%.

These metrics have traditionally been calculated when the observation value exceeds a certain minimum value, often 60 ppb for 1-hour ozone evaluation (Hogrefe et al, 2001). The MNBE and MNGE are estimated using 3 different minimum 8-hour ozone thresholds: 20, 40, and 60 ppb. Since 60 ppb is fairly close to the standard of 80 ppb, the metrics using this threshold reflect the models ability to predict high concentrations of 8-hour ozone. The 20 and 40 ppb minimum thresholds are included in the analysis to get a better idea about how well the model is performing at predicting diurnal formation and removal processes and for days between high ozone episodes.

3. RESULTS & DISCUSSION

Figure 2 shows the bias by month for each annual simulation for PM2.5 sulfate ion, nitrate ion, ammonium ion, and organics. These figures allow for a comparison of performance between years for the PM2.5 chemical species of most importance in the eastern United States.

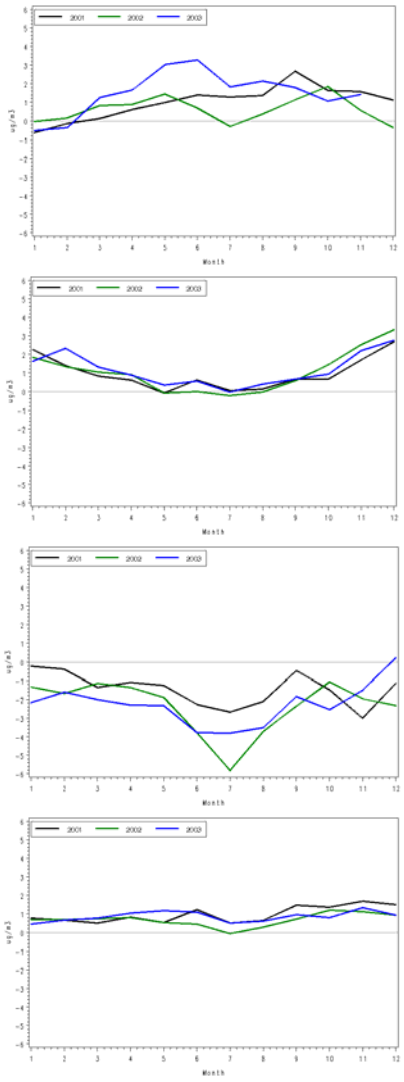

Figure 2. Monthly Bias for Each Annual Simulation by PM2.5 Chemical Specie: From top to bottom: sulate, nitrate, ammonium, and organics

The fractional bias and fractional error both express model performance on a normalized scale and are plotted against each other in Figure 3. Monthly averaged metrics from all three annual simulations are included in Figure 3, giving a total of 36 data points for each of the PM2.5 chemical species. Model performance is optimal when the data points are close to 0% fractional bias (y axis) and close to 0% fractional error (x axis).

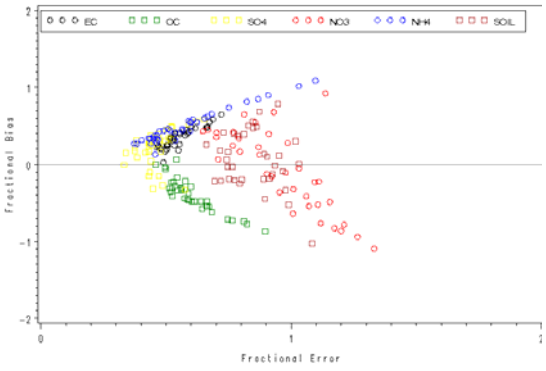

Figure 3. Fractional Bias v. Fractional Error Metrics by PM2.5 Chemical Specie for all Three Annual Simulations

Sulfate ion model performance appears good in the summer months, which is important since this is when concentrations are highest. Nitrate ion concentrations are highest in the winter and model performance for nitrate is best during the winter. Ammonium is over-predicted for the entire year, suggesting too much ammonia is in the modeling system. The apparent excess of free ammonia may over-state the effects of reducing nitrate ion from the reduction of nitrogen oxide emissions in nitric acid limited regions. This is hypothesized because any free nitric acid in favorable meteorological conditions (low temperatures and high humidity) will partition into the particulate phase. The excess ammonia in the modeling system may also reduce the chemical transport model responsiveness to changes in ammonia emissions in ammonia limited regions.

The modeling system does well at predicting elemental carbon. Performance for soil is poor, but given that there is little PM2.5 soil measured on filters in the eastern United States the regulatory importance of this specie is minimal compared to sulfate, nitrate, and organic carbon.

The performance for organic carbon is poor in the summer months and in many urban areas. The science of secondary organic aerosol formation is still evolving so it is not realistic for the modeling system to accurately predict organic carbon when the

formation processes are not fully understood. In addition to uncertain secondary organic aerosol formation processes, it is important to continue work to evaluate the speciation profiles used for primary PM2.5 emissions.

Average 8 hr ozone metrics over all summer days and for each minimum threshold value are shown in Table 1. A maximum of 80 days were included in the model performance analysis after the exclusion of "spin-up" days in early June. The lower minimum thresholds include all 80 days included in the model performance analysis. The 60 ppb threshold excludes a few days that do not have high 8-hour ozone concentrations.

The values in Table 1 reflect averages over all stations in the domain and all days of each summer, which illustrates the overall skill of the model predicting high and low ozone on a regional domain. The MNBE ranged from –3.1% to 19.2% over all three summers and all three minimum threshold values used for metric calculation. The MNGE ranged from 13.8% to 32.3%.

	Cutoff 20 ppb			Cutoff 40 ppb			Cutoff 60 ppb		
	2001	2002	2003	2001	2002	2003	2001	2002	2003
N	80	80	80	80	80	80	79	77	79
MNGE (%)	29.9	28.3	32.3	19.6	18.9	22.0	13.8	13.8	15.8
MNBE (%)	15.9	15.9	19.2	5.9	5.9	9.2	-3.1	-2.2	-1.0

Table 1. Episode Average Model Performance Metrics for all Three Summer Simulations using Different Minimum Thresholds for Metric Calculation: 20, 40, and 60 ppb.

The metrics in Table 1 indicate the modeling system performs consistently for all three summers. An interesting trend in the metrics that has been shown in other studies is that the gross error is lowest using the 60 ppb threshold and highest using the 20 ppb threshold (Hogrefe et al, 2001). The modeling system has a high bias at the 20 ppb cutoff, a small positive bias at the 40 ppb cutoff, and a small negative bias at the 60 ppb cutoff. This suggests the modeling system is over-predicting the lower 8-hour ozone concentrations and slightly under-predicting the higher concentrations. The overall performance for the summers of 2001 and 2002 are very consistent. The metrics for the summer of 2003 indicate additional work could be done to improve performance.

The performance metrics are also estimated by day of the week using a 60 ppb cutoff for each summer simulation. Based on the data shown in Table 2, there are no apparent trends in model performance by day of the week or even a more general trend between weekend days and weekdays. The performance by day of the week is very similar in 2001 and 2003. The performance in 2002 is very different than 2001 and 2003.

Year	Metric	MON	TUE	WED	THU	FRI	SAT	SUN
2001	Bias	-10.5	-4.5	-1.4	3.1	2.5	-0.3	-9.0
2001	Error	16.8	15.2	14.8	13.3	12.8	15.6	15.6
2001	N	12,239	11,815	10,165	7,784	4,395	5,194	8,148
2002	Bias	-4.2	-2.6	-3.6	-1.7	-5.3	-7.3	-4.1
2002	Error	14.3	12.9	12.4	12.6	15.2	14.7	13.6
2002	N	9,236	9,744	8,324	7,800	12,097	12,834	11,732
2003	Bias	-9.5	-8.4	-1.7	1.4	0.4	-0.5	-7.3
2003	Error	14.7	15.7	16.0	14.9	16.9	13.8	14.9
2003	N	9,178	7,409	3,924	5,238	5,704	6,335	8,588

Table 2. Day of Week Model Performance Metrics for all Three Summer simulations using 60 ppb minimum threshold.

The day of the week metrics by summer show that doing this type of analysis for a single summer may lead to a particular conclusion that does not translate well to other summer seasons.

4. CONCLUSION

In the near term, most PM2.5 and Regional Haze control plans are likely to target emission reductions of sulfur dioxide and nitrogen oxides, which match up well with the current strengths of the modeling system: predicting summer sulfate ion and winter nitrate ion.

The results shown for the three summer simulations show that the modeling system tends to over-predict minimum concentrations and slightly under-predict peak concentrations. The over-prediction of minimum concentrations is not of great regulatory concern since attainment tests are based on the application of relative reduction factors to daily peak concentrations. Since the regulatory applications are not focused on minimum concentrations of ozone it appears that the modeling system is doing a good job of appropriately estimating 8-hour average ozone in the upper midwest region for the summers of 2001, 2002, and 2003. This

confidence in the modeling results allows for the modeling system to be used to support the development of emissions control scenarios and State Implementation Plans to meet the 8-hour ozone and annual PM NAAQS.

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REFERENCES

Baker, K. Meteorological Modeling Protocol for Application to PM2.5/Haze/Ozone Modeling Projects, 2004. See http://www.ladco.org/tech/photo/photochemical.html

Bey, I.; Jacob, D; Yantosca, R.; Logan, J.; Field, B.; Fiore, A.;Li, Q.; Liu, H.; Mickley, L.; Schultz, M. Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res*., **106**, *23*,073-23,096, 2001.

Dudhia, J. A nonhydrostatic version of the Penn State/NCAR mesoscale model: Validation tests and simulation of an Atlantic cyclone and cold front, *Mon. Wea. Rev.,* **121,** 1493-1513, 1993.

ENVIRON International Corporation. 2005. User's Guide Comprehensive Air Quality Model with Extensions (CAMx4) Version 4.00. ENVIRON International Corporation, Novato, California. See www.camx.com (accessed July 15, 2005).

Grell, G. A.; Dudhia, J.; Stauffer, D. *A description of the Fifth Generation Penn State/NCAR Mesoscale Model (MM5),* NCAR Tech. Note, NCAR TN-398-STR, 138 pp., 1994.

Hogrefe, C.; Rao, S.T.; Kasibhatla, P.; Hao, W.; Sistla, G.; Mathur, R.; McHenry, J. Evaluating the Performance of regional-scale photochemical modeling systems: Part II—ozone predictions. *Atmospheric Environment*, 2001, **35***,* 4175-4188.

Janssen, M.; Hua., C. Emissions Modeling System-95 User's Guide. 1998. See http://www.ladco.org/emis/guide/ems95.html

Johnson, M. Meteorological Modeling Protocol: IDNR 2002 Annual MM5 Application, 2003. See http://www.iowacleanair.com/prof/progdev/files/protocol.pdf

Seigneur, C.; Pun B.; Pai, P.; Louis, J.; Solomon, P.; Emery, C.; Morris, R.; Zahniser, M.; Worsnop, D.; Koutrakis, P.; White, W.; Tombach, I. 2000. Guidance for the performance evaluation of three-dimensional air quality modeling systems for particulate matter and visibility, *J. Air Waste Manage. Assoc*., 50, 588-599.

U.S. Environmental Protection Agency, Draft Guidance on the use of Models and Other Analyses in Attainment Demonstrations for the 8-hour Ozone NAAQS, EPA-454/R-99-004, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 1999.

U. S. Environmental Protection Agency, Guidance for Demonstrating Attainment of Air Quality Goals for $PM_{2.5}$ and Regional Haze, Draft 2.1, January, 2001.

U. S. Environmental Protection Agency, Guidance for Tracking Progress Under the Regional Haze Rule, EPA-454/B-03-004, Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2003.

vanloon, G.; Duffy, S. *Environmental Chemistry: A Global Perspective*; Oxford University Press, 2000.

Wilkinson, J.; Loomis, C.; Emigh, R.; McNalley, D.; Tesche, T., 1994. *Technical Formulation Document: SARMAP/LMOS Emissions Modeling System (EMS-95)*. Final Report prepared for Lake Michigan Air Directors Consortium (Des Plaines, IL) and Valley Air Pollution Study Agency (Sacramento, CA).