

THE 2006 CMAQ RELEASE AND PLANS FOR 2007

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1. INTRODUCTION

The 2006 release of the Community Multiscale Air Quality (CMAQ) model (Version 4.6) includes upgrades to several model components as well as new modules for gas-phase chemistry and boundary layer mixing. Capabilities for simulation of hazardous air pollutants have been expanded to include more gas-phase toxics and several toxic metals. The mercury model has been updated and adapted to the new version of CMAQ. The CMAQ analysis tools, sulfur tracking and carbon source apportionment, have also been updated to operate with new physics, chemistry, and aerosol components.

An overview of the new upgrades for CMAQ version 4.6 is provided in Section 2. Then, in Section 3, we take a preliminary look at the model evaluation for the CMAQv4.6 release. Section 4 outlines plans for further model development over the next year and beyond. Finally, a summary is presented in Section 5.

2. UPGRADES FOR 2006

2.1 Gas Chemistry

An updated Carbon Bond mechanism (CB05) has been incorporated into the CMAQ modeling system. The CB05 mechanism contains 52 chemical species and nearly twice the number of reactions compared to the CB-IV mechanism. The major changes in the mechanism relative to the CB-IV fall into the following categories: kinetic updates, photolysis updates, extended inorganic reaction set, and better representation of the atmospheric chemistry of reactive species that are important in ozone and PM formation. Several

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additional species have been included, as described by Yarwood et. al. (2005), to more appropriately represent peroxy radical reactions, the production and decay of acetaldehyde and higher aldehydes, and the reactions of internally-bonded alkenes. Also, a more realistic description of terpene reactions and products has been included (Carter 2000).

In addition to the generalized gas chemistry solvers available in CMAQ (SMVGEAR and Rosenbrock) the CB05 may be run using a customized Euler Backward Iterative (EBI) solver for efficient execution. The details of the CB05 mechanism are contained in a report by Yarwood et al. (2005). Sarwar et al. (2006) describe the implementation and testing of the new mechanism in CMAQ. An analysis of the results with the CB05 mechanism versus the CB-IV mechanism will be shown in a presentation by Deborah Luecken. In general, summer hourly ozone (O₃) concentrations are about 9% higher compared to simulations using the CB-IV mechanism.

2.2 Heterogeneous Chemistry

Since 2003 (CMAQv4.3), the heterogeneous reaction probability for dinitrogen pentoxide (N₂O₅) hydrolysis was computed as a function of aerosol nitrate and sulfate concentrations. Recent laboratory evidence indicates that the probability is also influenced by temperature and relative humidity. Thus, the reaction probability has been changed so it is now a function of sulfate, nitrate, temperature, and relative humidity (Evans and Jacob 2005). To assist model users, the N₂O₅ heterogeneous reaction probability (i.e., GAMMA_N2O5) has been added as a new variable to the aerosol diagnostic output file.

The CB05 mechanism contains two gas-phase reactions involving N₂O₅ and water (H₂O) leading to nitric acid (HNO₃). In the beta version of the CB05 mechanism (released with CMAQ v4.5.1),

these reactions were turned off because there was some concern that they may be caused by wall effects in the smog chambers and modeled nitrate aerosol concentrations were persistently overpredicted. For CMAQv4.6 the gas-phase reactions involving N_2O_5 and H_2O leading to HNO_3 have been turned on. The SAPRC99 mechanism also contains a reaction involving N_2O_5 and H_2O leading to HNO_3 that was turned off since CMAQv4.3. This gas-phase reaction has also been turned on in the SAPRC99 mechanism for CMAQv4.6.

These changes in the heterogeneous and gas phase N_2O_5 hydrolysis reactions typically decrease net production of nitrate in the winter with a slight increase in the summer. During winter, the effects of turning on the gas-phase hydrolysis reactions are more than offset by the reduction in GAMMA_N2O5 values. In summer, the GAMMA_N2O5 values tend to increase, because of higher temperature and humidity, and the gas-phase reactions also lead to more nitrate production. A detailed description of the N_2O_5 hydrolysis treatment in CMAQ will be presented by Prakash Bhawe.

2.3 Aerosols

The CMAQ model uses a thermodynamic aerosol equilibrium model known as ISORROPIA that has been updated for Version 4.6. The developer of ISORROPIA, Dr. Athanasios Nenes, released the latest version in March 2006. The most significant change from the version used in CMAQv4.5.1 is correction of an error in how activity coefficients were adjusted for temperature. Tabulated activity coefficients for temperatures in the range 260.5 - 285.5 K have been corrected, resulting in increased ammonium nitrate aerosol concentrations. In a test simulation of 9 days in January 2001, the maximum increases were $1.8 \mu\text{g}/\text{m}^3$ and $0.5 \mu\text{g}/\text{m}^3$ for aerosol nitrate and ammonium, respectively.

Another revision to the aerosol model was made to ameliorate the very high concentrations (e.g., $1000 \mu\text{g}/\text{m}^3$) of aerosol water output from ISORROPIA in previous versions of CMAQ. We decided to limit RH to 95% during the ISORROPIA calculations so that concentrations of aerosol H_2O would not resemble cloud or fog-like conditions.

The effects of the RH limitation are most pronounced in polluted areas with high relative humidity, where aerosol water decreases result in similar decreases in ammonium and nitrate aerosols. A second order effect is that sulfate and all other aerosol species increase slightly due to

the smaller particle sizes, and hence, lower deposition velocities. Visual range should increase in the areas where aerosol water decreases. Also, the sum of the Aitken and accumulation modes should be a better approximation for $PM_{2.5}$ than in previous releases of the code.

2.4 Toxics and Mercury

Version 4.6 has two new mechanisms for Hazardous Air Pollutants (HAPs). One expands the SAPRC99 mechanism for HAPs used in version 4.5. The other mechanism adapts to the new CB05 mechanism. Each new mechanism simulates a larger number of gas phase HAPs than version 4.5 and also simulates several toxic metals and diesel contributions to particulate matter. Details of the toxic capabilities of CMAQv4.6 will be presented by Bill Hutzell.

The gaseous chemistry, aqueous chemistry and wet and dry deposition processes in the standard version of CMAQ were modified to simulate atmospheric mercury. For CMAQv4.6, the mercury related modifications will also be linked to the CB05 gas phase mechanism and the AE4 aerosol module. Dry deposition of elemental mercury (Hg^0) gas has been added to the model with deposition velocity (V_d) values provided from the Meteorological-Chemical Interface Processor version 3.1 (MCIP v3.1). The functionality of the mercury codes will also be coordinated with the special codes of the air toxics version of CMAQ so that a comprehensive one-atmosphere multipollutant model might be developed. Russ Bullock will present a detailed description of the mercury modeling capability of CMAQv4.6.

2.5 Other Upgrades

A new planetary boundary layer (PBL) model has been implemented in CMAQv4.6. The PBL model is a new version of the Asymmetric Convective Model (ACM) that has been an option in CMAQ for many years. The new version (ACM2) combines the non-local convective mixing of the original ACM with local eddy diffusion to better represent the full range of turbulent transport within the convective boundary layer (CBL). The ACM2 generally produces smoother profiles in the lowest model layers than the original ACM and more well-mixed profiles than the EDDY option. The ACM2 also simulates shallower effective mixed layers than the EDDY option. Compared to EDDY, the ACM2 produces slightly greater ground-level O_3 concentrations, especially

in the late afternoon. A presentation by Jonathan Pleim will provide further details of the ACM2 formulation, testing, and evaluation.

The Plume-in-Grid (PinG) capability and the tracking models for Carbon Apportionment and Sulfate have been updated to be compatible with the AE4 aerosol mechanism and the CB05 gas-phase chemistry mechanism. A restart file that contains the last time step of the entire 4-D concentration array has been added for the purpose of providing initial concentrations to start the next simulation period. The traditional concentration output file, normally used for this purpose, can consume a lot of disk space. Applications can optionally reduce the number of species and layers saved to the concentration file, thereby freeing up a large amount of disk space for multi-day scenarios. There have also been various updates to the Parallel I/O code library, thus requiring the new version for CMAQv4.6 compilation.

3. PRELIMINARY EVALUATION

Each major new version of CMAQ is accompanied by an operational evaluation comparing model simulated concentrations to measurements from several air quality monitoring networks (e.g. Eder and Yu 2006). For the 2006 release, CMAQv4.6 is being run for four months representative of the four seasons (January, April, July, and October) in 2001. Two modeling domains are used: a coarse grid ($\Delta x = 36$ km) covering all of continental US, southern Canada, and northern Mexico and a nested fine grid ($\Delta x = 12$ km) covering the eastern US. Two vertical grid configurations are also being tested: 14 layers, as has been used for the past several years, and 34 layers. The 34 layer run uses a vertical grid that is identical to the MM5 meteorological model simulation. Comparison of the two vertical grid structures will be presented by Wyatt Appel. A few preliminary results of the 12 km 14 layer January and July simulations are presented here.

3.1 January 2001

Figure 1 shows concentrations of particulate matter smaller than $2.5 \mu\text{m}$ in diameter (PM_{2.5}), broken down by chemical composition averaged over all the Status and Trends Network (STN) sites within the 12 km modeling domain for January 2001. Overall, this year's model (CMAQv4.6) produced slightly more PM_{2.5} than last year's model (CMAQv4.5) with about 40%

overprediction compared to the observations. The great bulk of the overprediction is in the total carbon (TC) and Other categories. The predominately secondary inorganic aerosols (NO₃, NH₄, SO₄) compare very well between the two model versions and to the observations. The Other category contains mostly primary aerosol material whose emissions are not well defined. The TC is a combination of organic carbon (OC) that is generally underpredicted by the model, and elemental carbon (EC), that is generally overpredicted in the winter. Note that EC is also mostly primary aerosol and thus subject to uncertainties in emissions.

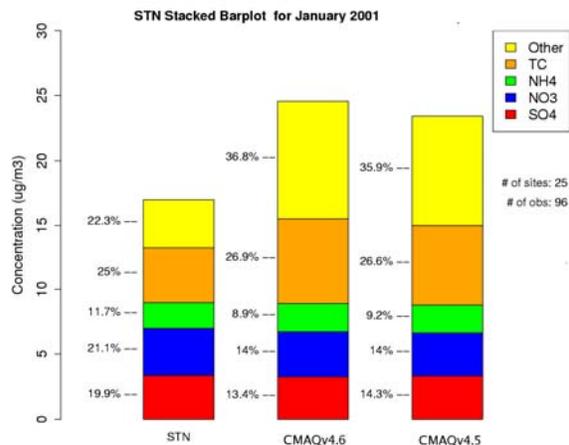


Figure 1. Comparison of observed and modeled aerosols for January 2001

3.2 July 2001

The most important air pollutant species in the summer are O₃, sulfate aerosols, and carbonaceous aerosols. Figure 2 shows a comparison between CMAQv4.5 and CMAQv4.6 modeled peak daily 8-h average O₃ concentrations for July 2001. Overall, CMAQv4.6 simulates higher O₃ concentrations than CMAQv4.5 by an average of about 10%. Figure 3 shows time series of 8-h peak daily O₃ concentrations from both model versions compared to measured values at AQS monitoring sites. The upper panel of Fig. 3 compares modeled and measured concentration averages for all sites in the SW region of the 12 km domain (LA, TX, MS, MO, OK) and the lower panel includes all sites in the Mid-Atlantic region (WV, KY, TN, VA, NC, SC, GA, AL). In all cases CMAQv4.6 predictions are higher than CMAQv4.5. During high ozone episodes, such as July 22-23 in the SW region and July 9-11 and July 15-16 in the mid Atlantic region,

CMAQv4.6 predictions were usually closer to the observations.

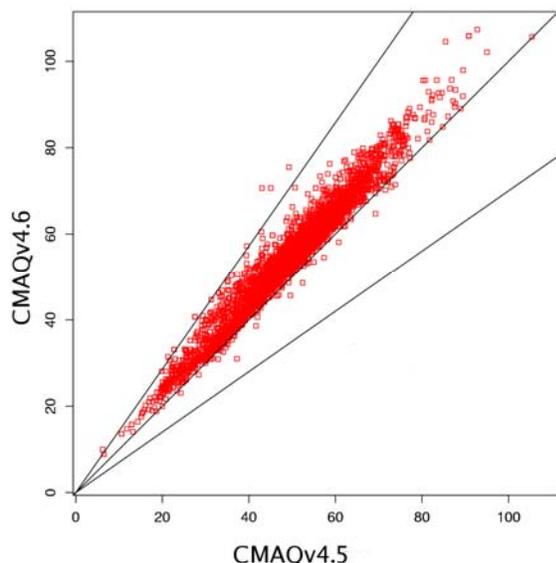


Figure 2. Comparison of observed and modeled peak 8 h average ozone concentrations at the AQS network sites for July 2001.

Figure 4 shows comparisons of $PM_{2.5}$ and its constituent chemical compositions for July 2001. For total $PM_{2.5}$ both versions compare well to the observations at the STN sites. Sulfate, the largest chemical component in summertime aerosols, is slightly overpredicted by CMAQv4.6 but to a slightly lesser degree than CMAQv4.5. Total Carbon, however, is significantly underpredicted by both model versions, which represents mostly underprediction of OC which dominates TC in the summer. The poor performance of CMAQ for organic carbon largely reflects incomplete science in secondary organic aerosol chemistry. Thus, development of new secondary organic aerosol (SOA) chemistry and thermodynamic modules based on the latest field and laboratory experiments is at the top of our list for next year. In addition, direct OC emissions from poorly characterized sources (e.g., wildfires and open-burning) may also play a role in the OC model underprediction.

4. FUTURE WORK

Major new developments in air quality modeling as well as meteorology modeling and the coupling between them are being worked on for release in 2007 and 2008.

4.1 WRF-CMAQ coupling

A new project to develop 2-way coupling between CMAQ and the Weather Research and Forecast (WRF) model is underway. The idea is to create a coupled model system with synchronous meteorology and chemistry calculations. This system will allow for data flow in both directions at each model time step thus facilitating chemical feedback to the meteorology. For example, the coupled system will be able to include aerosol feedback to the radiation model. With the coupled model we can develop integrated resolved-scale microphysics and aqueous chemistry; such that aqueous chemical species could be advected through the grid along with the microphysical components (i.e. cloud water, cloud ice, rain, snow, graupel). Indirect effects of aerosol on microphysics could also be incorporated. Furthermore, closer temporal coupling between meteorology and chemistry would result in more consistency between the chemical and meteorological simulations.

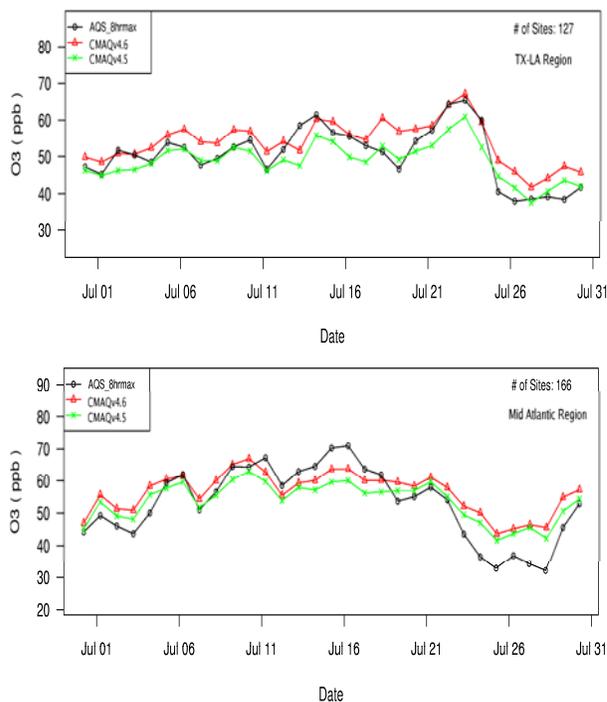


Figure 3. Time series of observed and modeled 8-hr daily maximum ozone concentrations for the TX-LA region (top) and the mid-Atlantic region (bottom) for July 2001

The coupled WRF-CMAQ would have similar capabilities as the WRF/CHEM that is currently

available as an extension of the WRF system. The WRF-CMAQ, however, takes a different approach where the meteorology and chemistry components are compiled together but are still structurally separate codes. This design will allow consistent air quality modeling in either 2-way coupled or 1-way sequential execution. A prototype working model is anticipated in 2008.

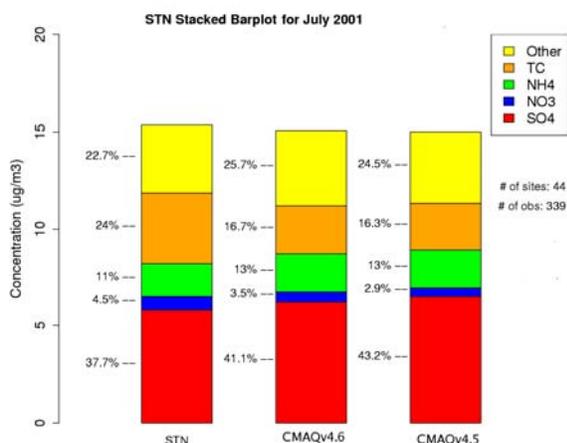


Figure 4. Comparison of observed and modeled aerosols for July 2001

4.2 Aerosols

The CMAQ aerosol module will undergo major upgrades for the 2007 release. The main improvements will be the incorporation of a new SOA module and a coarse particle chemistry module. New SOA precursors will include sesquiterpenes and isoprene. In the new aerosol module, the transfer of volatile inorganic material between the gas phase and the coarse-particle mode will be simulated. Also, a new algorithm to moderate biogenic emissions to account for in-canopy deposition will be applied, resulting in substantially lower emission fluxes at night when turbulent transport is limited.

4.3 Photolysis

Currently, photolysis rates are precomputed by an "off-line" model assuming clear-sky conditions and climatological estimates of temperature, pressure, and ozone vertical profiles. A single vertical profile of aerosol attenuation coefficients is used in the clear-sky photolysis rates (J-values) calculation. J-values are computed for various latitudes, altitudes, and hour angles. The resulting J-value look-up table is read

into CMAQ and interpolated to each specific grid cell for the local solar time. Subsequently, cloud effects are applied through parameterized estimates of cloud transmissivity.

A new computationally efficient "on-line" photolysis model has been developed for use in the 2007 release of CMAQ. The new model has seven wavelength bands in UV and visible and updated absorption cross-sections and quantum yields based on Fast-J (Wild et al., 2000). This model will compute J-values for each grid cell at each time step using CMAQ aerosols for extinction and scattering, grid specific surface albedo, and modeled meteorological and chemical profiles. Effects of clouds will also be added to the on-line calculations.

4.4 Cloud modeling

We have begun collaboration with Georg Grell of NOAA/ESRL to adapt the WRF/CHEM convective cloud model for CMAQ. The cloud mixing algorithm includes treatment of an updraft, downdraft, and compensating subsidence. We are also planning to update the aqueous chemistry module in CMAQ. We plan to test a more detailed aqueous mechanism, and may include it in the next model release. In addition, we will continue testing a generalized aqueous chemistry solver.

4.5 Satellite data assimilation

New techniques for satellite data assimilation in both the meteorology models (MM5 and WRF) and the chemical transport model (CMAQ) have been developed in collaboration with University of Alabama at Huntsville and NASA/MSFC. Satellite information, primarily from GOES, is used to derive several key components of the meteorology and atmospheric chemistry systems. Satellites give accurate information on cloud coverage, cloud height, and optical thickness that can be used to estimate more accurate surface solar insolation and photolysis rates. Also, satellite derived skin temperature can be used to nudge soil moisture. These techniques have been tested and evaluation and will be made operational for retrospective modeling at the USEPA over the next couple of years.

5. SUMMARY

A very active program of innovative development is continuing for the CMAQ modeling system. The 2006 release (CMAQv4.6) includes

major new components for gas phase chemistry and PBL mixing. CMAQ's air toxics and mercury capabilities continue to expand and improve. Next year will bring a new state-of-the-science secondary organic aerosol model to the CMAQ system along with an improved treatment for coarse particles.

The CMAQ system is now capable of using WRF modeled meteorology. This capability, however, is still evolving as the WRF model is being updated to include four dimensional data assimilation (FDDA) and the Pleim-Xiu land surface model (PX LSM). In addition to 1-way sequential execution, we are developing 2-way coupled WRF-CMAQ capability that will allow for chemistry feedbacks to the meteorological simulation.

for publication, it does not necessarily reflect their policies or views.

6. REFERENCES

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Disclaimer

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