Evaluation of CMAQ v4.7 Sulfate Predictions for 2002 - 2006

K. Wyat Appel and Shawn J. Roselle

Atmospheric Modeling and Analysis Division, National Exposure Research Laboratory, Office of Research and Development, U.S. EPA, RTP, NC 27711

Motivation and Background

- Particulate sulfate (SO_4^{2-}) constitutes a large portion of the total fine particulate mass observed in the eastern United States, especially in the summer when emissions of sulfur dioxide (SO_2) are large.
- Accurate predictions of atmospheric SO_4^{2-} concentrations are necessary in order to capture a large portion of the total fine particulate mass in the atmosphere and correctly predict the chemistry for other atmospheric pollutants that is highly dependent on the SO_4^{2-} predictions.
- The Community Multiscale Air Quality (CMAQ) model is a state-of-the-science air quality model that predicts the formation, transport and fate of a large number of gaseous and particulate species, including SO_4^{2-}.
- The performance of CMAQ model SO_4^{2-} predictions has generally been regarded as good, and consistently performs better than most other predicted particulate species.
- A set of CMAQ model simulations were performed for the 2002-2006 period. An operational performance evaluation of the full time period showed that the CMAQ model predictions for SO_4^{2-} were generally poor in the summer. In 2002 and 2005, the CMAQ model significantly underpredicted fine particulate SO_4^{2-} concentrations, while in 2003, 2004 and 2006 the fine particulate SO_4^{2-} concentrations were also underpredicted, although to a smaller degree.
- This work investigates some of the causes of poor SO_4^{2-} predictions in the summer periods. The main focus will be on the gas- and aqueous-phase pathways for SO_4^{2-} production and how errors in meteorology may affect these mechanisms for SO_4^{2-} production in the atmosphere. Additionally, the impact on SO_4^{2-} predictions from integrating observed satellite cloud information into the CMAQ model is shown.

MM5 Simulation Characteristics

Options	MM5
Cloud Microphysics	Reisner 2
Radiation Scheme	RRTM longwave / Dudhia Shortwave
Cumulus Parameterization	Kain-Fritsch2
Land Surface Model	PX
Cloud Scheme	ACM-2
Surface-Layer Scheme	Pleim
Soil Moisture Nudging	NAM-218
Initial / Boundary Conditions	NAM-218

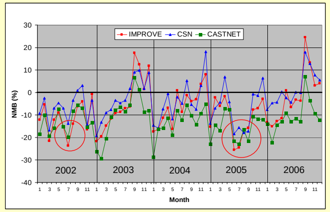
- The MM5 simulations include grid nudging (FDDA) and were run for the five year period of 2002 – 2006 on a 12km x 12km horizontal grid domain over the eastern United States with 24 vertical layers.

CMAQ Simulation Characteristics

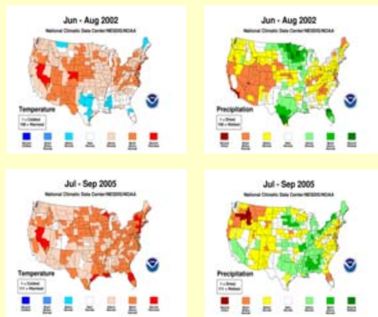
- CMAQ version 4.7 which includes
  - Updated Heterogeneous N_2O_5 Gamma Calculation
  - AERO5 (includes Secondary Organic Aerosols (SOA) and dynamic course mode)
  - Updated cloud integration time-step and sub-grid scale clouds
  - Updated HONO chemistry
  - Other various code fixes and updates
- EPA 2002 National Emissions Inventory (NEI) with year specific updates including
  - Continuous Emissions Monitoring (CEMs) data
  - Updates to mobile and biogenic emissions

CMAQ SO_4^{2-} Model Performance

Fine Particulate SO_4^{2-} Normalized Mean Bias

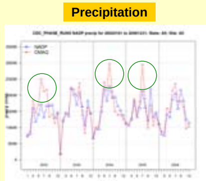


While SO_4^{2-} is often underpredicted slightly (other than in the winters of 2003, 2004 and 2006), there are large underpredictions in the summers of 2002 (June – August) and 2005 (June – September)

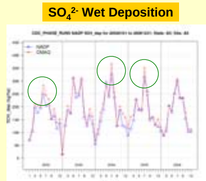


The summers of 2002 and 2005 were dry in the eastern United States. The summer of 2002 was extremely dry as well, while in 2005 several tropical systems impacted the southeast U.S.. Extremes of these type can be challenging for meteorological models such as MM5 to predict accurately. The plot below on the left shows that MM5 significantly overpredicts precipitation in the summer of 2002 and July of 2005.

Precipitation



SO_4^{2-} Wet Deposition



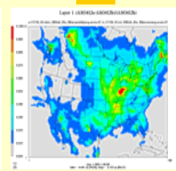
Large positive biases in precipitation correlate to the large positive biases in SO_4^{2-} wet deposition in July of 2002, 2004 and 2005. However, biases in SO_4^{2-} wet deposition do not account for all the underprediction in fine particulate aerosol SO_4^{2-} during the summers of 2002 through 2006.

Possible Reasons for Poor Model Performance

1) Too little gas phase SO_4^{2-} production. To investigate this issue, CMAQ was run using clear sky photolysis values to estimate the maximum increase in SO_4^{2-} through the gas-phase production pathway. The plots below show the absolute and ratio difference in SO_4^{2-} between the base CMAQ simulation and the CMAQ simulation using clear sky photolysis.

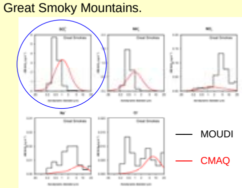
Absolute Difference

Ratio



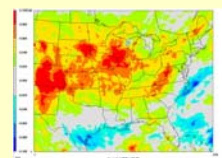
Average increase in SO_4^{2-} at observation sites is ~3.5 - 4.5%, which is relatively small when compared to the total underprediction in SO_4^{2-}. Therefore, too little gas-phase production is not the primary cause for the SO_4^{2-} underpredictions.

2) Particle size distribution. Errors in the size distribution of SO_4^{2-} particles could result in too many particles in the course particle size and not enough in the fine particle size range. To investigate this, CMAQ predicted SO_4^{2-} particle sizes were compared to size resolved observations (MOUDI) of SO_4^{2-} from several cities across the United States for several time periods. Below is an example from a site in the Great Smoky Mountains.



Sulfate particle size distribution (circled in blue) is too broad and shifted toward larger particle sizes. This results in a too much dry deposition of SO_4^{2-} and hence lower concentrations of ambient fine particulate SO_4^{2-}. Similar results were found for the other cites and time periods examined. Until the issue is resolved users should continue to use the sum of the fine (i) and accumulation (j) modes to represent fine particle mass.

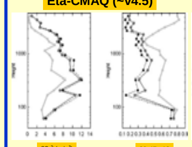
3) CMAQ vertical resolution. CMAQ model simulations show lower concentrations of SO_4^{2-} when fewer vertical layers are used. Some of the underprediction could be mitigated by using a greater number of vertical layers (e.g. 34 instead of 24).



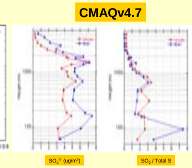
Shown to the left is the ratio difference in SO_4^{2-} between a 14 layer CMAQ simulation and a 34 layer CMAQ simulation (34/14). In general SO_4^{2-} concentrations are higher in the 34 layer simulation, with some increases exceeding 10%. On average, the increase in SO_4^{2-} is approximately 5-6%.

4) Vertical distribution of SO_4^{2-}. Analysis of CMAQ v4.5 simulations showed that the model predicted too much SO_4^{2-} aloft. Improvements in CMAQ v4.7, including a new convective mixing scheme, were expected to improve this overprediction aloft.

Eta-CMAQ (~v4.5)



CMAQv4.7



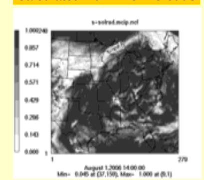
The plots to the left show that while the overprediction of SO_4^{2-} aloft was corrected somewhat in the new model, but is still largely overpredicted.

Integrating Satellite Cloud Observations into CMAQ

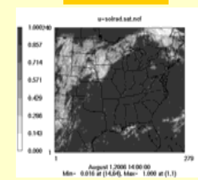
The analyses presented in the previous section suggest only relatively minor improvements in SO_4^{2-} underpredictions. It is likely that errors in the meteorological predictions, specifically the cloud predictions, may be responsible for a larger portion of the errors in SO_4^{2-} predictions.

One solution to improve the cloud predictions in the CMAQ model is to incorporate the observed cloud information into the modeling system. Researchers at the University of Alabama at Huntsville and the Atmospheric Modeling and Analysis Division have developed a technique for incorporating the observed cloud information from satellites into the CMAQ modeling system. However, the observed cloud information can currently only be used to adjust the radiation values used in the photolysis calculations in the CMAQ model. Below is an example of the effect that the integrated cloud product has on both ozone (O_3) and SO_4^{2-} predictions in CMAQ.

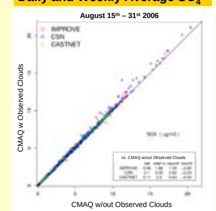
Calculated from MCIP Clouds



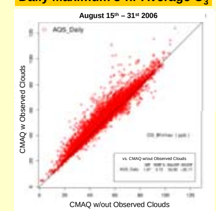
Satellite Derived



Daily and Weekly Average SO_4^{2-}



Daily Maximum 8-hr Average O_3



Using the observed clouds to adjust photolysis in the CMAQ model results in greater photolysis (suggesting the CMAQ model predicts too much cloud cover) and hence greater concentrations of SO_4^{2-} (through gas-phase production) and much higher ozone mixing ratios. The increase in SO_4^{2-} is relatively small, which is consistent with the sensitivity run in which clear sky photolysis rates were used.

Summary

Results from a multi-year operational evaluation of the CMAQ model v4.7 show SO_4^{2-} is underpredicted, particularly in the summer periods.

Several factors may be contributing to the underprediction, including too little photolysis, particles sizes that are too large, vertical layer structure and not enough vertical mixing of SO_4^{2-} from aloft. However, these factors may contribute only a small portion to the SO_4^{2-} underprediction.

Future work will focus on improving the cloud predictions in the both the meteorological model (through incorporating satellite cloud observations) and the CMAQ model (by way of a new cloud scheme).

Acknowledgements: Rohit Mathur and Prakash Bhave from the U.S. EPA and Lara Reynolds, Nancy Hwang and Lucille Bender from CSC.