Instrumented Models for Diagnostic Model Evaluation: Decoupled Direct Method in 3-D, Carbon Apportionment, and Sulfur Tracking

Sergey Napelenok, Prakash Bhave, Shawn Roselle

2009 AMAD Peer Review: Session 2

Model Evaluation: Establishing Model's Credibility

Environmental Issue

Operational model evaluation provides an overall assessment of modeling results. In complement, diagnostic evaluation and the use of instrumented models help identify *model processes* that require further improvements.

From the perspective of both model evaluation and model development, it is often useful to determine not only the state of an environmental system, but also its response to perturbation in various parameters that define it. Instrumented models offer a unique interpretation of standard air quality model output and are useful to better understand the physical and chemical processes occurring in the atmosphere as re-created by the model.

Furthermore, instrumented models are useful in regulatory applications in the areas of source apportionment, estimation of uncertainty, selection and evaluation of control strategies, and future projections of air quality conditions.

Research Objectives

Three instrumented models are presented: Decoupled Direct Method in three dimensions (CMAQ-DDM-3D), Carbon Apportionment (CMAQ-CA), and Sulfur Tracking (CMAQ-ST). Each has been employed in a variety of applications, extracting additional utility model predictions.

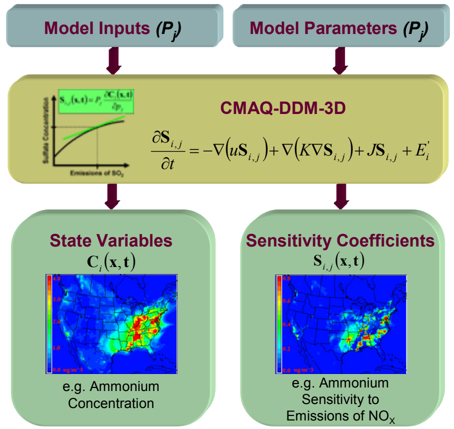
❖ CMAQ-DDM-3D provides an efficient and accurate approach for calculating sensitivity of atmospheric pollutant concentrations to changes in photochemical model parameters (emissions, chemical reaction rates, initial/boundary conditions, etc.)

❖ CMAQ-CA allows quantification of absolute contributions from different emission sources to primary organic carbon (OC) and elemental carbon (EC). Molecular tracer and radiocarbon techniques have been developed to measure source specific contributions. The combination of these measurements and carbon apportionment allows evaluation of both the air quality model and the emissions inventory.

❖ CMAQ-ST allows for analysis of the sulfate productions pathways. It tracks sulfate production from gas phase and aqueous phase chemical reactions, as well as contributions from emissions and initial/boundary conditions.

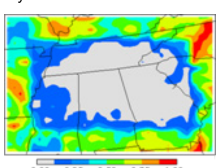
Decoupled Direct Method in 3D

CMAQ-DDM-3D operates within the underlying atmospheric model to compute local, semi-normalized sensitivities to perturbations in input and model parameters.



CMAQ-DDM-3D has been used in a variety of applications. In one application, direct sensitivities were calculated to estimate a reduced form model of ozone concentrations as a function of emissions of NO_x, VOC, and initial conditions. This information was then used to quantify uncertainty in model outputs through a Monte Carlo analysis (Pinder et al., 2008b; Poster 2.6).

In another application, CMAQ-DDM-3D sensitivities were used to quantify the relationship between source region emissions and receptor concentration in an inverse modeling study of NO_x emissions in the southeastern United States (Napelenok et al., 2008; Poster 2.4). Sensitivity coefficients were calculated for emissions from predefined geographical regions in the modeling domain in order to establish an emissions scenario that would minimize the differences between model predictions of column NO_2 values and those observed by the SCIAMACHY satellite.

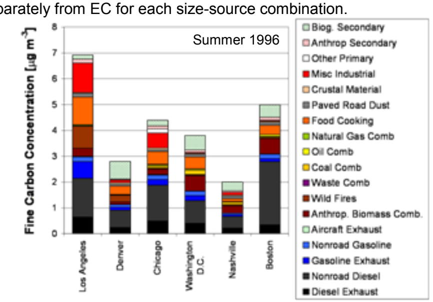


In the inverse modeling study, DDM-3D was also used in the domain selection process to find a balance between impacts from outside the source regions and domain size for computational efficiency. It was important in the inverse calculation to minimize the influence from non-contributing areas in the domain.

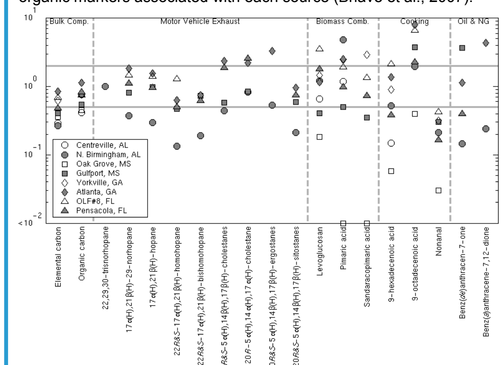
CMAQ-DDM-3D has been updated to include second and higher order sensitivity coefficients (Hakami et al., 2003), and been applied to calculate model responses to larger perturbations such as those typically analyzed in SIP modeling.

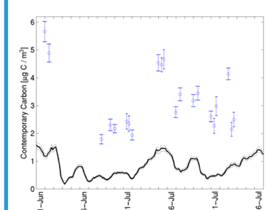
Carbon Apportionment

CMAQ-CA employs 64 additional species in order to track size, composition, and source of primary carbonaceous aerosol. Sixteen emissions categories are represented (e.g. diesel exhaust, wild fires, food cooking). Size distribution are tracked by two modes (Aitken and accumulation), and primary organic aerosol (POA) is tracked separately from EC for each size-source combination.



CMAQ-CA can be used to quantify sector emissions contributions on receptors in the domain as was done in a diagnostic evaluation of carbonaceous $PM_{2.5}$ through a comparison to measurements of organic markers associated with each source (Bhave et al., 2007).



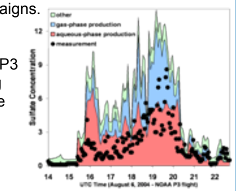


Carbon Apportionment also allows for direct evaluation of model results against radiocarbon measurements (Lewis et al., 2004), because the defined sources can be readily associated with either fossil fuel or contemporary carbon as shown in the figure on the left.

Sulfur Tracking

CMAQ-ST uses additional species to calculate sulfur contributions from all formation pathways. Currently tracked sulfur pathways include: primary emissions; initial and boundary conditions; gas phase oxidation of SO_2; and five separate aqueous oxidation pathways via hydrogen peroxide (H_2O_2), ozone (O_3), methyl hydrogen peroxide (MHP), peroxy acetic acid (PAA), and aqueous catalysis by iron (Fe^{++}) and manganese (Mn^{+}). Each species is advected, diffused, processed through clouds, and deposited through wet and dry pathways.

CMAQ-ST has been used to track sulfate formation pathways as predicted by the model during extensive measurement field campaigns. For example, it was possible to track model predictions along NOAA P3 aircraft flight paths taking measurements during the 2004 INTEX campaign (Singh et al., 2006).



Future Directions

The instrumented models are constantly updated with the latest science additions to the base model and will be distributed through the CMAS center. Additional in-house capabilities are also being explored including an effort to develop in-house capabilities for adjoint modeling (Hakami et al., 2007).

Impact

❖ Distributed to regulatory and academic communities and used in a wide range of applications.

❖ Used in comparisons of model results with non-traditional measurements (radiocarbon, organic tracers).

❖ Applied in modeling support for OAQPS/NCEA $PM_{2.5}$ assessment.

❖ Identified model processes warranting future model development.

Contributors/Collaborators

Dan Cohan, Rice University
Yongtao Hu, Georgia Institute of Technology
Talat Odman, Georgia Institute of Technology
Armistead Russell, Georgia Institute of Technology