UNITED STATES ENVIRONMENTAL PROTECTION AGENCY

Chapter 6 –Session 1 Overview: Model Development and Diagnostic Testing

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Session 1 Overview: Model Development and Diagnostic Testing

6.0 Introduction

As mandated by the Clean Air Act, the EPA develops National Ambient Air Quality Standards (NAAQS), to protect human health and the environment. The development of optimal emission control strategies that are both environmentally protective and cost effective to comply with the NAAQS requires air quality simulation models that can reliably predict the impact of emission control strategies on ambient levels of criteria pollutants such as ozone and particulate matter. AMAD research presented in this session supports the implementation of these air regulations by developing and providing comprehensive atmospheric modeling systems to the client offices. Through detailed treatment of physical and chemical processes affecting the transport and fate of atmospheric pollutants, these modeling systems provide scientifically-sound tools to understand the relationships between sources of air pollution and ambient concentrations over spatial scales ranging from urban to continental and temporal scales ranging from hourly to decadal. This is accomplished through an integrated multi-disciplinary research approach involving physical, numerical, and computational modeling to develop a “numerical laboratory” wherein atmospheric physico-chemical interactions can be effectively simulated to guide the development of air pollution abatement strategies. Through diagnostic testing against measurements over a wide range of spatial and temporal scales, the models provide a framework to test and refine hypotheses and process formulations, thereby improving our understanding of key atmospheric processes influencing ambient levels of pollutants.

Program goals and strategic directions for research described in this session support Long Term Goal (LTG) 1 of the EPA Clean Air Research Multi-Year Plan (MYP), which aims to reduce uncertainty in standard setting and air quality management decisions based on advances in air pollution science. The primary focus of this LTG is the development and implementation of NAAQS for particulate matter and ozone, and air quality regulations for hazardous air pollutants.

6.1 Research Summary

Through detailed simulation of various physical and chemical processes such as horizontal and vertical transport, diffusion, emissions, deposition, atmospheric chemistry, and cloud processes, comprehensive Eulerian air quality models attempt to describe most major processes in the atmosphere that are considered key in determining the distributions and levels of the atmospheric chemical species. These models must address the increasing complexity arising from new applications that treat multi-pollutant interactions, with the need to facilitate the design of effective air quality management strategies that focus on simultaneously controlling emissions of multiple criteria pollutants and their precursors. Research in this session can be broadly categorized into four major areas of emphasis:

1. Modeling of atmospheric chemistry and transport processes to characterize the fate of air pollutants
2. Characterization and modeling of emissions
3. Improvements in meteorological models for air quality modeling applications
4. Diagnostic Analysis and Testing

6.1.1 Modeling of atmospheric chemistry and transport processes to characterize the fate of air pollutants

AMAD’s atmospheric chemistry and transport research program develops scientifically-credible and computationally-efficient comprehensive modeling systems that provide a fundamental understanding of atmospheric processes that dictate the levels of atmospheric concentrations and deposition of air pollutants. The Community Multiscale Air Quality (CMAQ) modeling system is an urban to continental scale multi-pollutant air quality modeling system. Over the past 5 years, the primary focus of AMAD’s research in this area has focused on developing and improving the representation of interactions between gas-, aqueous-, and particle-phase air pollutants.

To accurately represent the seasonal and spatial variations in airborne particulate matter composition, numerous scientific improvements were made in CMAQ model processes. New pathways for secondary organic aerosol (SOA) formation from precursors including isoprene, sesquiterpenes, benzene, glyoxal, and methylglyoxal were incorporated to improve the model’s ability to represent the contribution of organic carbon to airborne fine PM. Improvements in aerosol process representation in CMAQ now allow semi-volatile aerosol components to condense and evaporate from the coarse mode and nonvolatile sulfate to condense on the coarse mode; thus, dynamic interactions between inorganic gases and coarse-mode PM are represented. In addition, the model has been updated to represent sea-salt emissions from wave-breaking in the coastal surf zone. A new parameterization to represent the heterogeneous N_2O_5 hydrolysis on particle surfaces, which includes dependence on temperature, relative humidity, inorganic PM composition, and phase state was developed and incorporated into CMAQ. These enhancements result in an improved representation of partitioning of airborne nitrogen between the gas-phase and the fine and coarse PM modes.

During the past five years, numerous improvements have been made to the representation of gas-phase chemistry in the CMAQ modeling system. An updated and expanded version of the Carbon Bond mechanism (namely, CB05) was incorporated in CMAQ to more accurately simulate wintertime, pristine, and high-altitude conditions. Compared to its widely-used predecessor CBM-IV mechanism, CB05 includes nearly twice the number of reactions and includes numerous updates to the kinetic data. AMAD has also actively collaborated with external chemical mechanism developers to ensure that evolving mechanisms are consistent with recent measurements and theory. While the SAPRC99 mechanism is currently available as an option in CMAQ, work is underway to test and incorporate its successor, SAPRC07, in the model. Efforts are also underway to develop and test updates to the RACM2 mechanism (successor to the RADM2 mechanism previously available in CMAQ) and will be included for further testing in CMAQ. Enhancements to the treatment of gas-phase chemistry in CMAQ were also directed at improving the partitioning of airborne oxidized nitrogen as well as at providing a consistent treatment for multi-pollutant applications (O_3, PM, air toxics, Hg). Earlier versions of CMAQ significantly underestimated ambient HONO mixing ratios which also exhibited a diurnal profile opposite to that seen in limited observations. HONO is the largest source of OH radicals (which controls oxidation) during the morning hours. Recent studies suggest that HONO emissions and heterogeneous reactions (involving NO_2 and H_2O) on aerosol and ground surfaces can produce HONO in the atmosphere. The incorporation of these production pathways results in improved representation of both the diurnal variability and the magnitude of predicted ambient HONO mixing ratios and its impact on atmospheric photochemistry. To provide a consistent treatment of gas-phase atmospheric kinetics for multi-pollutant applications and assessment studies, Cl_2 chemistry was incorporated in the CB05 chemical mechanism in

CMAQ. Research has also focused on improving the estimation of photolysis rates used in chemistry calculations. A developmental on-line photolysis rate module that incorporates the radiative impacts of aerosol loading simulated by the model has been included in CMAQ. This enhancement allows the investigation of potentially important impacts of scattering and absorbing aerosols in modulating photolysis rates and atmospheric photochemistry regulating the formation of secondary air pollutants. An additional module that assimilates satellite-derived information on the location and opacity of cloud fields has also been incorporated to more accurately represent the spatial variability in the photolysis rate attenuation from clouds and its subsequent impacts on atmospheric chemistry.

CMAQ also contains the option to simulate the atmospheric fate of mercury compounds and 40 other Hazardous Air Pollutants (HAPs). The selection of HAPs included in CMAQ was based on consultation with the EPA Office of Air Quality Planning and Standards and includes the 33 HAPs identified under the Integrated Urban Air Toxics Strategy as posing the greatest potential public health concern in the largest number of urban areas, as well as several additional HAPs which are significant contributors to ozone and secondary particulate matter formation. This extended capability allows air quality managers to transition from the traditional pollutant-by-pollutant control approach to multi-pollutant control, and enables a robust examination of the benefits/dis-benefits of various control strategies.

Over the past five years, research efforts have also resulted in improvements in the representation of atmospheric advective, turbulent, and cloud transport processes in CMAQ. The three-dimensional advective transport representation in CMAQ was modified to improve mass conservation associated with mass-inconsistent wind and density fields generated by the driving meteorological models. The convective cloud scheme was also modified to improve the representation of cloud mixing on the vertical distribution of air pollutants in the atmosphere. Research efforts have also focused on improvements to the representation of dry deposition, a persistent sink for many airborne pollutants. To improve consistency between the dynamical and chemical calculations, the estimation of dry deposition velocities has been closely linked with surface-flux calculations in the meteorological model. Additionally, treatment of bi-directional surface exchange of species such as NH_3 and mercury has also been recently incorporated, thereby improving the representation of the atmospheric budgets of these compounds.

Another area of emphasis in the development of the CMAQ modeling system has been in improving its overall computational efficiency. Research and development in this area have focused on refining the numerical and structural aspects of the modeling system to enable its practical use while accommodating the increasing science complexity and increasing computational demands associated with expanding geographic domains and finer grid cell structures. AMAD's research has focused on developing numerically-accurate and computationally-efficient algorithms for the various physical and chemical modules included in CMAQ. Research was performed to investigate the trade-offs between accuracy and computational efficiency of the integration scheme employed to solve the ordinary differential representing the gas-phase kinetics. Based on extensive testing options to use either an Euler Backward Iterative (EBI) or a Rosenbrock solver were included in CMAQ as these provided the optimal balance between numerical accuracy and computational efficiency. Similarly, detailed performance profiling of the CMAQ code indicated that ~47% of the computational effort in the aerosol module was related to the calculation of coagulation coefficients; revisions to this aspect of the aerosol module resulted in increasing the computational efficiency of aerosol calculations by about a factor of 2. Recognizing that the time-steps for calculation of advective transport could be limited aloft due to faster winds, a layer-dependent time-step calculation was implemented to improve the overall computational efficiency of CMAQ. In addition, through collaborations with Sandia National Laboratory, AMAD developed several optimizations in the CMAQ code to improve its computational performance and scalability on “commodity” computational architectures, thereby promoting its operational use by the external user community.

6.1.2 Characterization and modeling of emissions

Emissions of air pollutants constitute a key input to air quality models and uncertainties inherent in their estimation can greatly influence the predictive accuracy of air quality modeling systems. The accurate characterization of anthropogenic and natural emissions and their relative contribution to air quality degradation is important, given the large costs associated with environmental decisions aimed at regulating emissions. AMAD’s research in this area focuses on improving the representation of the spatial and temporal variability of source emissions that are affected by meteorological variability, and characterizing the impact of uncertainties in emission estimation on air quality simulation model results. Quantitative anthropogenic emission inputs to the CMAQ model rely on the U.S. EPA National Emission Inventories, which are regularly updated by the EPA Office of Air Quality Planning and Standards. Over the past five years, AMAD’s research has led to the development of techniques to quantify emissions from non-traditional sources. These sources represent natural (e.g., biogenic VOC and NO_x, wildfires, sea-salt emissions) and/or non-regulated sectors (e.g., ammonia emissions from agricultural and animal husbandry), which are currently poorly characterized, but have a significant impact on ambient levels of criteria pollutants such as O_3 and PM.

AMAD pioneered the development of the Biogenic Emissions Inventory System (BEIS) for estimating natural volatile organic compound emissions from vegetation and NO from soil. The system was further enhanced to provide estimates of vegetation emissions of sesquiterprenes, a major precursor for secondary organic aerosol formation. AMAD’s emission characterization research has also focused on exploiting emerging and non-traditional data sources to improve the quantification of atmospheric emissions. Current research is attempting to integrate the National Land Cover Database to better characterize the spatial distribution of vegetation and associated atmospheric emissions of VOCs. AMAD exploited the availability of emerging remotely sensed information on location of wildland and prescribed fires to develop a combined satellite-ground based fire emission estimation methodology that has significantly reduced the uncertainty in daily emission estimates from this source category.

6.1.3 Improvements in meteorological models for air quality modeling applications

An accurate description of the dynamical state of the atmosphere is critical for describing the fate and transport of airborne pollutants. Meteorological models are key components of an air quality modeling system in that they provide detailed three-dimensional information on air flow, turbulence, and clouds which are critical for accurately simulating atmospheric pollutant transport and dispersion, temperature and humidity which regulate atmospheric kinetics leading to secondary pollutant formation, and radiation which initiates photo-chemical reactions in the atmosphere. Atmospheric conditions such as light winds, clear sky, and subsidence inversion, which lead to accumulation of air pollutants, are not the primary foci of numerical weather prediction research where the emphasis is on extreme weather conditions which may not always be conducive for severe air quality episodes. AMAD’s meteorological modeling research focuses on (1) improving the linkage between meteorological and air quality models to maintain consistency in data and process treatment common to both modeling systems (e.g., mass consistency during transport calculations, cloud mixing), (2) developing and refining physical process components in the meteorological model that are critical for air quality applications (e.g., boundary-layer mixing, surface exchange and deposition), and (3) reducing uncertainty in key meteorological variables through development and application of data assimilation techniques.

Since the majority of the pollutants are emitted into the lowest atmospheric layers, planetary boundary layer (PBL) processes dictate, to a large extent, the dilution of emissions, chemical concentrations and reactions influencing surface level air quality. AMAD has conducted research to develop a PBL model that produces realistic fluxes and profiles of both meteorological variables as well as chemical species, thereby providing consistent and accurate representation of PBL processes and their impacts on air quality. AMAD's research on data assimilation techniques demonstrated the impacts of reduced uncertainty in resulting meteorological fields on improving the accuracy of air quality predictions. In addition, AMAD's research in meteorological model development has led to the successful transition from the Mesoscale Meteorological Model version 5 (MM5) to the Weather Research and Forecasting (WRF) model for air quality applications.

6.1.4 Diagnostic Analysis and Testing

Diagnostic analysis and testing of process modules, numerical algorithms, and overall model predictions, is an integral part of AMAD's model development activities. The CMAQ model includes several options to output additional diagnostic information. Process analysis has been implemented to output the local tendencies of each modeled process and provides valuable information for model budget analysis. Diagnostic evaluations using an instrumented version of CMAQ indicated that biogenic secondary organic aerosol was largely responsible for underpredictions of summertime organic carbon when compared to field measurements in North Carolina (Poster 1.3). Dynamic interactions between inorganic gases and coarse mode particulate matter (PM) in CMAQ were tested using field experimental data from the Tampa Bay BRACE study, including size-resolved compositional data of the PM. The International Consortium for Atmospheric Transport and Transformation (ICARTT) field study provided a rich set of field data from 2004 in the northeast U.S. coastal areas, which was used to evaluate several different CMAQ model applications. Vertical profiles of sulfate and organic carbon particulate, and NO_y were compared with CMAQ-predicted profiles (Poster 1.2). Radar wind profiler data have been processed to estimate the depth of the mixed layer; these data were used to evaluate the modeled estimates of mixed layer depth from the WRF model (Poster 1.6). In addition, several of the posters in Session 2 on Model Evaluation discuss applications of model diagnostic evaluation using the CMAQ model.

Over the past five years, the deployment of CMAQ as part of the NOAA-EPA National air quality forecasting system has also assisted in the diagnostic analysis of model upgrades. Daily developmental forecast simulations were used to continuously examine the performance of the model under varying dynamical and chemical conditions, which helped in identifying systematic biases and guiding further development of specific process modules. In addition, detailed analysis of model predictions and measurements from the 2004 ICARTT and the 2006 Texas Air Quality field studies was performed to develop diagnostic process-based insights and identify adequacies/inadequacies in the modeling system.

6.2 Future Research

AMAD's integrated multi-disciplinary modeling research will continue to develop and enhance comprehensive atmospheric modeling systems through exploration of the development of novel new modeling methodologies to extend the CMAQ modeling system to address emerging environmental problems. Model applications to date have clearly demonstrated the continued need to account for and to improve the representation of interactions of atmospheric processes occurring at the various spatial and temporal scales. Model simulations over annual cycles have pointed to the need for more robust methods for specification of lateral boundary conditions. While linking with global scale atmospheric chemistry models has been pursued and will continue to be investigated, it is also recognized that biases in the global model can propagate and influence regional CMAQ calculations, confounding the interpretation of model results. The specification of the lateral boundary conditions, to a large extent, dictates the simulated variability in the free troposphere, which, in turn, can impact the simulated surface-level background values for a variety of trace species. The tightening of the NAAQS to lower threshold values (e.g., the recent revisions to the O_3 NAAQS) places additional requirements on the ability of atmospheric chemistry transport models to accurately represent the entire spectrum of ambient concentrations including the background values. Expansion of the modeling system to hemispheric scales provides opportunities to consistently represent processes at all scales and will improve the characterization of long-lived pollutants (e.g., mercury). The extension also supports future applications to study the linkage and interactions between global climate and air quality. On the other end of the spectrum, emerging agency problems focusing on air quality-human exposure linkage will require application of the model at significantly finer resolutions to capture variability in ambient concentrations of a number of pollutants (O_3, PM, air toxics) and resultant human exposure. These extensions will require further development and enhancement of various physical and chemical process modules/algorithms included in the modeling system. Research will focus on improving the chemical interactions between gas-aerosol-aqueous phases. We will also work to modify chemical mechanisms used by the model to be consistent with the latest science to include new information (e.g., reactive nitrogen reactions, improvements in aromatic chemistry), and new compounds to address emerging issues such as biofuels and chemicals that contribute to global warming.

Research efforts will also focus on reducing the uncertainty in PM predictions by the model. To better understand the discrepancies between model predictions and measurements especially during the cool seasons, efforts will focus on characterizing unspeciated $PM_{2.5}$ (trace element $PM_{2.5}$ composition, non-carbon organic matter, sampling artifacts associated with gravimetric $PM_{2.5}$ measurements). Anticipating the promulgation of coarse PM standards, efforts to simulate emissions and transport of natural windblown dust and interactions between crustal ions and gases will continue. To support model applications dealing with air quality-climate interactions, algorithmic development of methods to estimate and represent the optical and radiative properties of airborne PM will continue.

AMAD's research will also continue to develop and refine meteorological models for air quality applications. Investigation of the use of non-traditional upper air data in data assimilation to improve the characterization of atmospheric dynamical conditions will be conducted. In addition, higher resolution and more up-to-date land information from the National Land Cover Dataset (NLCD) will be incorporated to improve land-surface fluxes in the meteorological model as well as the estimation of dry deposition velocities for use in the chemistry-transport model.

Research on further development of boundary layer schemes to better represent stable atmospheric conditions (especially winter-time) and their impact on simulated pollutant levels and distributions will be conducted.

To address the needs of emerging assessments for both air quality-climate interactions and for finer scale air quality applications, AMAD recently embarked on the development of a coupled atmospheric dynamics-chemistry model. A prototype coupled atmospheric modeling system based on the WRF meteorological model and the CMAQ chemistry-transport model has been developed, wherein direct radiative effects of absorbing and scattering aerosols in the troposphere estimated from the spatially and temporally varying simulated aerosol distribution can be fed-back to the WRF radiation calculations, resulting in a “2-way” coupling between the atmospheric dynamical and chemical modeling components. Additionally, coupling the modeling systems provides means for finer scale applications, wherein higher frequency of data exchange between meteorological and chemical calculations is necessary to capture the effects of meteorological variability on modeled concentrations. Research efforts will continue to further develop and refine various components of this coupled modeling system to facilitate its use in addressing emerging Agency problems dealing with both air quality-climate interactions as well as fine-scale modeling to understand the relationships between air quality and human health.

6.3 Impact

The CMAQ modeling system, the primary product from AMAD’s model development research, has been and continues to be extensively used by EPA and the states for air quality management (State Implementation Plans and rulemaking: Clean Air Interstate Rule, Clean Air Mercury Rule, Renewable Fuels Standard Act-2), thereby providing critical regulatory support to assure protection of public health and the environment. AMAD’s research on developing techniques for estimating emissions of biogenic VOCs and NO, and emissions from wildfires and sea-salt have helped better quantify the impact of these “non-controllable” sectors in air quality management and assessment studies. The multi-pollutant capability in CMAQ is now allowing model-based air quality assessment studies to transition from the traditional pollutant-by-pollutant approach to an integrated multi-pollutant air quality management approach, wherein benefits/dis-benefits of various control strategies can be more robustly examined. Frequent updates of the CMAQ modeling system are released to the user community through the Community Modeling and Analysis System (CMAS) center which also provides user support for the system and has helped in creation of a dynamic and diverse CMAQ community with over 1000 users worldwide. The evaluation of research and regulatory application of the model by this community over geographic regions outside the contiguous U.S. has helped improve the robustness of the modeling system and helped advance our understanding of a variety of air pollution-related problems.

In partnership with the NOAA National Weather Service, through a series of phased-development activities starting in 2004, CMAQ was deployed as part of a National Air Quality Forecasting system to provide reliable daily air quality forecast guidance across the continental United States; this product now serves as an operational tool that local and state air quality forecasters can use when creating daily Air Quality Index (AQI) outlooks and issuing health advisory warnings for more than 290 million people across the country. CMAQ forecasting applications have also served to provide guidance for planning aircraft measurement missions during recent field studies such as the 2004 International Consortium for Atmospheric Transport and Transformation (ICARTT) and the 2006 Texas Air Quality study, with detailed post-mission analysis providing in-depth process based analysis of field data and also providing guidance for future model development research.

Recognizing that meteorological requirements for air quality modeling studies are different from the needs for weather prediction modeling, AMAD's research on development and improvement of specific process modules and data assimilation techniques within the meteorological models has promoted and advanced the use of prognostic meteorology models for driving air quality simulations; these improvements have been made available to the wider user community through incorporation in publicly released versions of the WRF (and predecessor MM5) modeling systems.

6.4 Session Posters

The following six (6) posters will be presented in this session:

(1) The Community Multiscale Air Quality (CMAQ) Model: An Interdisciplinary Approach for Multipollutant Modeling Analysis (1.1)
(2) Atmospheric Chemistry Mechanisms: Current State and Future Needs (1.2)
(3) Modeling Atmospheric Particulate Matter: Description and Evaluation of the CMAQ Aerosol Module (1.3)
(4) Emissions Modeling: Fires, Biogenics, and Sea-Salt (1.4)
(5) Multiscale Meteorological Modeling for Air Quality Modeling Applications (1.5)
(6) Planetary Boundary Layer Modeling for Meteorology and Air Quality (1.6)

Abstracts for each of these posters follow.

The Community Multiscale Air Quality (CMAQ) Model: An Interdisciplinary Approach for Multipollutant Modeling Analysis (1.1)

Rohit Mathur, Shawn Roselle, Jonathan Pleim, Kenneth Schere, Jeffrey Young,
Prakash Bhave, William Hutzell, Tanya Otte, David Wong, Deborah Luecken,
Russell Bullock, Golam Sarwar, Christopher Nolte, Annmarie Carlton,
Sergey Napelenok, George Pouliot, Jesse Bash, Donna Schwede, and Robin Dennis

Collaborators: Tad Kleindienst, Ed Edney, John Offenberg (EPA/NERL/HEASD); Tyler Fox and staff (EPA/OAQPS); CMAS Center staff (UNC-Chapel Hill Institute for the Environment); Carlie Coats (Baron Advanced Meteorological Services); Ted Russell, Athanasios Nenes (Georgia Institute of Technology)

EPA and the states are responsible for implementing the National Ambient Air Quality Standards (NAAQS) for ozone and particulate matter (PM). New standards for 8-h average ozone and daily average $PM_{2.5}$ concentrations have recently been implemented. Air quality simulation models, such as the Community Multiscale Air Quality (CMAQ) model, are central components of the air quality management process at the national, state, and local levels. The CMAQ model, used for research and regulatory applications by the EPA, states, and others, must have up-to-date science in order to ensure the highest level of credibility for the regulatory decision-making process. The research goals under the CMAQ model development and evaluation program are (1) to develop, evaluate, and refine scientifically credible and computationally efficient process simulation and numerical methods for the CMAQ air quality modeling system; (2) to develop the CMAQ model for a variety of spatial (urban through continental) and temporal (days to years) scales and for a multipollutant regime (ozone, PM, air toxics, visibility, acid deposition); (3) to adapt and apply the CMAQ modeling system to particular air quality/deposition/climate-related problems of interest to EPA, and use the modeling system as a numerical laboratory to study the major science process or data sensitivities and uncertainties related to the problem; (4) to evaluate the CMAQ model using operational and diagnostic methods and to identify needed model improvements; (5) to use CMAQ to study the interrelationships between different chemical species as well as the influence of uncertainties in meteorological predictions and emission estimates on air quality predictions; (6) to collaborate with research partners to include up-to-date science process modules within the CMAQ model system; and (7) to pursue computational science advancements (e.g., parallel processing techniques) to maintain the efficiency of the CMAQ model.

The CMAQ model was initially released to the public by EPA in 1998. Annual updated releases to the user community and the creation of a Community Modeling and Analysis System (CMAS) center that provides user support for the CMAQ system and holds an annual CMAQ users conference have helped to create a dynamic and diverse CMAQ user community of over 1000 users throughout the world. CMAQ has been and continues to be extensively used by EPA and the states for air quality management analyses (SIPs; CAIR, CAMR, RFS-2 rulemakings), by the research community for studying relevant atmospheric processes, and by the international community in a diverse set of model applications. Future research directions include development of an integrated WRF-CMAQ model for two-way feedbacks between meteorological and chemical processes and models, and extension of the CMAQ system to hemispheric scales for global climate-air quality linkage applications and to the neighborhood scale for human exposure applications.

Atmospheric Chemistry Mechanisms: Current State and Future Needs (1.2)

Deborah Luecken, Annmarie Carlton, William Hutzell, Rohit Mathur,
Prakash Bhave, Golam Sarwar, Rob Pinder

Collaborators: Edward Edney, Tadeuz Kleindienst, John Offenberg (EPA/NERL); Barbara Turpin (Rutgers); Barbara Ervens (NOAA); William Carter (University of California, Riverside); William Stockwell (Howard University); Greg Yarwood (Environ Corporation)

An accurate characterization of atmospheric chemistry is essential for developing reliable predictions of the response of air pollutants to emissions changes, to predict spatial and temporal concentrations, and to quantify pollutant deposition. In the past, air quality modelers have largely focused on single-pollutant issues, but it has since become clear that it is more appropriate to treat chemistry in an integrated, multiphase, multipollutant manner (National Research Council, 2004). For example, both inorganic and organic aqueous-phase chemistry can influence formation of secondary organic aerosol (SOA) through cloud processing (Carlton et al., 2006; 2007). High-NO_x versus low-NO_x conditions influence both ozone and SOA formation (Ng et al., 2007). In the past five years, our requirements for air quality modeling have also changed: the new National Ambient Air Quality Standards (NAAQS) for ozone and fine particulate matter ($PM_{2.5}$) have shifted our focus from urban-scale ozone episodes (~7 days) to regional/continental-scale simulations over longer time periods (one month to one year). In addition, our chemical mechanisms must adapt quickly to address emerging issues of high importance, such as changing climatic conditions and the impacts of biofuels.

The object of our research in this area is to develop, refine, and implement chemical mechanisms for use in the Community Multiscale Air Quality (CMAQ) model to:

- Ensure that CMAQ and other models that are used for regulatory and research purposes have scientifically justifiable chemical representations, are appropriate for the application being studied, and are consistent with our most up-to-date knowledge of atmospheric chemistry;
- Ensure that interactions between gas-, aqueous- and particle-phase chemistries are adequately accounted for, so that we can truly predict multimedia chemical effects of emissions changes; and
- Develop techniques, tools, and strategies so that we are able to efficiently expand current mechanisms to predict the chemistry of additional atmospheric pollutants that we anticipate will become important in the future.

Our efforts to improve the chemical mechanisms in CMAQ have resulted in more complete and up-to-date descriptions of the important chemical pathways that influence concentrations of the criteria pollutants ozone and particulate matter. The inclusion of chlorine reactions and the explicit chemistry for 43 Hazardous Air Pollutants (HAPs) has helped to expand the applications for which CMAQ can be used. The inclusion of additional chemical detail in the aqueous and aerosol modules is providing pathways for more complete descriptions of secondary organic aerosol formation and decay.

Modeling Atmospheric Particulate Matter: Description and Evaluation of the CMAQ Aerosol Module (1.3)

Prakash Bhave, Christopher Nolte, Kristen Foley, Wyat Appel,
Annmarie Carlton, Sergey Napelenok

Collaborators: Francis Binkowski* (UNC Chapel Hill); Jerry Davis* (NC State Univ.); Edward Edney (ORD); James Kelly* (CARB); Athanasios Nenes (Georgia Tech.); Adam Reff* (OAQPS); Uma Shankar (UNC Chapel Hill); Barbara Turpin (Rutgers Univ.); Anthony Wexler (UC Davis); K. Max Zhang (Cornell Univ.)
* Former members of the NERL Atmospheric Modeling and Analysis Division

Atmospheric particulate matter (PM) is linked with acute and chronic health effects, visibility degradation, acid and nutrient deposition, and climate change. Accurate predictions of the PM mass concentration, composition, and size distribution are necessary for assessing the potential impacts of future air quality regulations and future climate on these health and environmental outcomes. The objective of this research is to improve predictions of PM mass concentrations and chemical composition, by advancing the scientific algorithms, computational efficiency, and numerical stability of the CMAQ aerosol module.

To achieve this objective, we have focused efforts on five areas in which previous versions of the CMAQ aerosol module were deficient. First, we doubled the computational efficiency of the aerosol module by improving the computations of coagulation coefficients and secondary organic aerosol (SOA) partitioning. Second, we worked with the developer of ISORROPIA, CMAQ's thermodynamic partitioning module for inorganic species, to smooth out discontinuities. Third, we developed a new parameterization of the heterogeneous hydrolysis of N_2O_5 as part of a larger effort to mitigate model overpredictions of wintertime nitrate aerosol concentrations. Fourth, we vastly improved the treatment of SOA by incorporating several new SOA precursors and formation pathways. Fifth, we implemented an efficient scheme to treat the dynamic interactions between inorganic gases and the coarse PM mode.

As a result of this research, the CMAQ aerosol module has been enhanced greatly over the past five years. During that time period, the aerosol module has been used for regulatory and forecasting applications (e.g., EPA-CAIR, NOAA-NCEP) because it is scientifically credible, computationally efficient, and numerically stable. With the recent scientific enhancements, our clients have increased confidence in the utility of CMAQ predictions of PM for future regulatory applications (e.g., RFS2 rulemaking). Meanwhile, the community of CMAQ users outside EPA continues to grow rapidly.

Emission Modeling: Fires, Biogenics, and Sea Salt (1.4)

Thomas Pierce, George Pouliot, William Benjey, Prakash Bhave,
David Mobley, Robert Pinder, Alice Gilliland

Collaborators: Cornell University (K. Zhang); EPA/NRMRL (C. Geron); EPA/OAQPS (Emissions Inventory/Assessment Group); EPA/Region VI (D. Roy); EPRI (E. Knipping); NASA (A. Soja, K. Pickering); NCAR (A. Guenther, C. Widenmeyer); and, the USFS (Fire Sciences Group)

The NARSTO 2005 assessment report stressed that emissions are at the cornerstone of air quality management decision-making. While OAQPS bears the responsibility for maintaining the National Emissions Inventory (NEI) for traditional anthropogenic sources (e.g., electrical generating units and mobile sources), many nontraditional emission categories (e.g., fires and biogenics) remain poorly characterized. During the 1980s, AMAD began to identify these categories and started to work with outside groups (e.g., NCAR, NASA, and the U.S. Forest Service) to build new and improved emission estimation tools. This poster highlights AMAD's work on fire, biogenics, and sea salt. Not shown because of space limitations is research on speciation, fugitive dust, lightning NO_x, and ammonia emissions from natural landscapes and agricultural activities. The goal of this research is to improve the characterization of emissions that will reduce the uncertainty in air quality model simulations, such as with the CMAQ model.

Until OAQPS and several Regional Planning Organizations spent nearly a million dollars building a 2002 biomass burning inventory for event-specific emissions, fire emissions were resolved only by month and by county. We used the 2002 inventory as a baseline for developing a combined satellite/ground-based inventory that can be constructed at a fraction of the cost. Our methodology has been incorporated into the NEI and has been used to estimate biomass burning emissions (on a daily basis) for the years 2003-2006.

AMAD was a pioneer in developing the Biogenic Emissions Inventory System (BEIS) for estimating biogenic volatile organic compounds from vegetation and NO from soil. We are now collaborating with NCAR to develop and evaluate the Model of Emissions of Gases and Aerosols from Nature (MEGAN), which represents an evolution of the BEIS system, but was only recently converted into Fortran computer code that is compatible with CMAQ. While BEISv3.14 is the latest operational biogenic emissions processor in the CMAQ system, the Division is performing rigorous tests with MEGAN and is seeking to integrate the National Land Cover Database (NLCD) into its structure.

Until recently, the characterization of sea-salt emissions did not exist in regional air quality simulation models. To meet this need, we have constructed a saltwater and shoreline geographical coverage file to drive emissions from open water and breaking waves using available flux equations that vary as a function of wind speed.

Simulations with the CMAQ modeling system suggest that model performance has improved with the addition of these more scientifically defensible emission algorithms. In addition, the underlying data and modeling systems have been distributed to the scientific community and are widely used by other modeling groups around the world. Over the next few years, we plan to continue work to integrate the MEGAN system, to incorporate NO production from lightning, and to model windblown dust.

Multiscale Meteorological Modeling for Air Quality Modeling Applications (1.5)

Robert Gilliam, Jonathan Pleim, and Tanya Otte

Collaborators: Aijun Xiu and Craig Mattocks, Institute for the Environment, UNC; Jimy Dudhia, NCAR; Lara Reynolds, CSC; David Stauffer and Nelson Seaman, Pennsylvania State University

Air quality models require accurate representations of atmospheric flow patterns and dispersion, cloud properties, radiative fluxes, temperature and humidity fields, boundary layer evolution and mixing, and surface fluxes of both meteorological quantities (heat, moisture, and momentum) and chemical species (dry deposition and evasion). Thus, meteorological models are critical components of the air quality modeling systems that evolve with the state of science. Because of this evolution, there is a need to frequently challenge our established models and configurations; this includes examining not only new physics schemes but also data assimilation strategies, which serve to reduce uncertainty in model output. It is also necessary to develop and refine physical process components in the models to address new and emerging research issues. Our meteorology modeling research program involves several key projects that have led to improved meteorological fields. First is the transition from the MM5 mesoscale model system to the Weather Research and Forecasting (WRF) model that represents the current state of science. Part of this effort was to implement in WRF the land-surface (Pleim-Xiu; PX), surface-layer (Pleim), and planetary boundary layer (Asymmetric Convective Model version 2; ACM2) schemes that had been used in MM5 and are designed for retrospective air quality simulations. Part of this effort included improving the PX land-surface physics that included a deep soil nudging algorithm and snow cover physics that dramatically improved temperature predictions in the winter simulations and in areas with less vegetation coverage. An additional effort was to work toward implementing in WRF the nudging-based four-dimensional data assimilation (FDDA) capability that had been available in MM5. Another effort has been a re-examination of FDDA techniques, including the use of a developmental analysis package for WRF ("OBSGRID") to lower the error of analyses that are used to nudge the model toward the observed state.

Current results of the implementation of new physics in WRF show that our configuration is comparable to or exceeds the level of MM5 in terms of the error in near-surface variables like 2-m temperature, 2-m moisture, and 10-m wind. This is true only when the new analysis package is used to improve analyses used for FDDA and soil moisture and temperature nudging in WRF. A new evaluation method that utilizes both wind profiler and aircraft profile measurements provides a routine method to examine not only the uncertainty of simulated wind in the planetary boundary layer, but also the less examined temperature structure. The WRF model has low error in temperature (median absolute error of 1.0 to 1.5 K or less) in the planetary boundary layer, which is generally less than the error near the surface. The model also simulates the evolution of the wind structure, including features like nocturnal jets and the convective mixed layer, with low error (<2.0 m s^{-1}). Our current configuration of WRF has met the requirements for the transition from MM5.

Acronyms

Sensitivity simulations are identified on the poster by mesoscale model, land-surface and planetary boundary layer options, and in one case, the tool used to generate analyses for FDDA. The run identifications include: WRF PXACM, WRF PXACM OG, MM5 PXACM and WRF NOAH YSU. **PXACM** indicates the Pleim-Xiu LSM (PX) and Asymmetric Convective Model Version 2 (ACM2). **OG** indicates OBSGRID was used. **NOAH** – National Centers for Environmental Prediction, Oregon State University, Air Force and Hydrologic Research Lab land surface model. **YSU** – Yonsei University PBL scheme implemented in WRF.

Planetary Boundary Layer Modeling for Meteorology and Air Quality (1.6)

Jonathan Pleim, Robert Gilliam and Tanya Otte

Collaborators: Aijun Xiu, Institute for the Environment, UNC; Jimy Dudhia, NCAR; Jim Wilczak and Laura Bianco, ESRL, NOAA; Shaocai Yu, Science and Technology Corporation; Lara Reynolds, CSC

Air quality modeling systems are essential tools for air quality regulation and research. These systems are based on Eulerian grid models for both meteorology and atmospheric chemistry and transport. They are used for a range of scales from continental to urban. A key process in both meteorology and air quality models is the treatment of sub-grid-scale turbulent vertical transport and mixing of meteorological and chemical species. The most turbulent part of the atmosphere is the planetary boundary layer (PBL), which extends from the ground up to ~1-3 km during the daytime but is only tens or hundreds of meters deep at night.

The modeling of the atmospheric boundary layer, particularly during convective conditions, has long been a major source of uncertainty in numerical modeling of meteorology and air quality. Much of the difficulty stems from the large range of turbulent scales that are effective in the convective boundary layer (CBL). Both small-scale turbulence that is sub-grid-scale in most mesoscale grid models and large-scale turbulence extending to the depth of the CBL are important for vertical transport of atmospheric properties and chemical species. Eddy diffusion schemes assume that all of the turbulence is sub-grid-scale and therefore cannot realistically simulate convective conditions. Simple nonlocal-closure PBL models, such as the Blackadar convective model that has been a mainstay PBL option in NCAR's mesoscale model (MM5) for many years, and the original Asymmetric Convective Model (ACM), also an option in MM5, represent large-scale transport driven by convective plumes but neglect small-scale, sub-grid-scale turbulent mixing. A new version of the ACM (ACM2) has been developed that includes the nonlocal scheme of the original ACM combined with an eddy diffusion scheme. Thus, the ACM2 can represent both the super-grid-scale and sub-grid-scale components of turbulent transport in the convective boundary layer. Testing the ACM2 in one-dimensional form and comparing to large-eddy simulations (LES) and field data from the second and third GEWEX Atmospheric Boundary Layer Study, known as the GABLS2 (CASES-99) and GABLS3 (Cabauw, NL) experiments demonstrates that the new scheme accurately simulates PBL heights, profiles of fluxes and mean quantities, and surface-level values. The ACM2 performs equally well for both meteorological parameters (e.g., potential temperature, moisture variables, and winds) and trace chemical concentrations, which is an advantage over eddy diffusion models that include a nonlocal term in the form of a gradient adjustment.

The ACM2 is in the latest releases of the Weather Research and Forecast (WRF) model and Community Multiscale Air Quality (CMAQ) model and is being extensively used by the air quality and research communities. Comparisons to data from the TexAQS II field experiment show good agreement with PBL heights derived from radar wind profilers and vertical profiles of both meteorological and chemical quantities measured by aircraft spirals.

6.5 Session Publications

This section presents the products (generally from 2004-2008) associated with each poster in this session. Some products are associated with multiple posters, so they are listed as products under more than one poster.

The Community Multiscale Air Quality (CMAQ) Model: An Interdisciplinary Approach for Multipollutant Modeling Analysis (1.1)

Appel, W., P. Bhave, A. Gilliland, G. Sarwar, and S.J. Roselle. Evaluation of The Community Multiscale Air Quality (CMAQ) Model Version 4.5: Uncertainties and Sensitivities Impacting Model Performance: Part II - Particulate Matter. Atmospheric Environment. Elsevier Science Ltd, New York, NY, 42(24):6057-6066, (2008).

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Atmospheric Chemistry Mechanisms: Current State and Future Needs (1.2)

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Modeling Atmospheric Particulate Matter: Description and Evaluation of the CMAQ Aerosol Module (1.3)

Appel, K.W.; Bhave, P.V.; Gilliland, A.B.; Sarwar, G.; Roselle, S.J. Evaluation of the Community Multiscale Air Quality (CMAQ) Model Version 4.5: Sensitivities Impacting Model Performance; Part II – Particulate Matter, *Atmospheric Environment*, 2008, 42:6057-6066.

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Emission Modeling: Fires, Biogenics, and Sea Salt (1.4)

Miller, A., G. Hidy, J. Hales, C. Colb, A. Werner, B. Haneke, D. Parrish, C. Frey, L. Rojas-Bracho, M. Deslaruiers, B. Pennel, and **D. Mobley** (2006) Air emission inventories in North America: A critical assessment, *Journal of Air and Waste Management* 56(8):1115-1129.

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Multiscale Meteorological Modeling for Air Quality Modeling Applications (1.5)

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Pleim, J. E., 2007: A combined local and non-local closure model for the atmospheric boundary layer. Part 2: Application and evaluation in a mesoscale model. *J. Appl. Meteor. Climatol*., **46**, 1396-1409.

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Planetary Boundary Layer Modeling for Meteorology and Air Quality (1.6)

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