WHITE PAPER:

IMPROVING AIR QUALITY-ECOSYSTEM MODELING CONNECTIONS TO EPA/ORD'S ECOSYSTEM RESEARCH PROGRAM

(*8 October 2010)*

Ecosystem & Hydrologic Linkage Team of the Modeling and Analysis Divison

Atmospheric Modeling and Analysis Division
National Exposure Research Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
Research Triangle Park, NC 27711

Air Quality – Ecosystem Modeling Connection

1. Introduction

As part of the EPA Office of Research and Development (ORD), the National Exposure Research Laboratory (NERL) is in the process of developing Integrated Transdisciplinary Research (ITR) programs to better address Problems of Broad, National Significance (PoBN). These high priority topics would benefit from more integrated collaborative research implementation across ORD's Laboratories and Centers. Energy and Climate, which includes environmental impacts on ecosystems, and Sustainable Water, which includes water quantity and quality impacts, have been identified as PoBNS's. NERL also considers Ecosystem Services research to be of broad national significance. Ecosystem responses are integral to all three problem areas. Just as there has been a movement away from assessing human exposure to air pollutants one chemical species at a time towards an integrated one-atmosphere approach, so too should there be an integrated one-atmosphere approach to assessing ecosystems exposure to air pollutants. With this in mind, we propose that now is the time to start advancing from simply a one-atmosphere to a one-biosphere approach that includes integration across multiple media and biogeochemical processes to more effectively address ecological interactions with the atmosphere as well as human systems. This overall vision will facilitate the air-surface intersection of atmospheric stressors-to-ecological receptor pathways. This overall vision is consistent with the EPA/NERL exposure science framework, as well as its emphasis on cross-disciplinary integration, and the vision is also consistent with the EPA/ORD research emphasis on Integrated Transdisciplinary Research.

This white paper addresses progress that can be made towards realizing this longer-term one-biosphere vision during the next 5 years by (1) describing critical issues, (2) describing the key air quality-exposure science questions relevant to the integrated assessment of air quality and ecosystem health, (3) discussing EPA/NERL's research program that will build on past linkages to improve exposure assessments through collaborative measurement campaigns, long-term monitoring and cross-disciplinary air-quality model refinement and decision tool development, and (4) outlining the research outcomes and products to be produced by this research program. The following sections of this white paper describe our early efforts to link air quality and ecosystem models and discusses key challenges and ways in which scientific and technical expertise within AMAD, across NERL and across the scientific community can be accessed to address these challenges.

Environmental Problem

A first step towards implementing a one biosphere paradigm is to better link air quality and ecosystem models. This is a critical initial research activity because atmospheric wet and dry deposition play an important role in terrestrial, freshwater aquatic and marine ecosystem functioning and degradation (Lovett and Tear, 2008; Driscoll et al., 2007; Vitousek, et al., 1997). For example, atmospheric deposition of sulfur and nitrogen is the primary source of acidifying chemicals that can impact aquatic and terrestrial ecosystems (Dennis et al., 2007). Acidification causes a cascade of effects that alter both terrestrial (DeHayes et al., 1999) and aquatic ecosystems (Driscoll et al., 2001). These effects include slower growth, the loss of soil fertility through acidification, the injury or death of forest vegetation and localized extinction of fish and other aquatic species. Atmospheric deposition is also an important source of excess nitrogen as a nutrient that can affect aquatic and terrestrial systems (Galloway et al., 2003). Excess nitrogen nutrient alters freshwater and terrestrial biodiversity, increases susceptibility of vegetation to insects and diseases, alters surface water quality and contaminates drinking water supplies (Driscoll et al., 2003). In the western U.S. microbial communities, such as lichen, are altered and diminished with increased nitrogen deposition (Fenn et al., 1998 and 2003). In the Rocky Mountains excess nitrogen deposition causes shifts in biodiversity and replacement of native plants (Baron, et al., 2000; Bowman et al., 2006). Excess nutrients alter estuarine systems through increased phytoplankton and algal productivity leading to eutrophication, loss of habitat, loss of dissolved oxygen, fish kills and decreased productivity (Valigura et al, 2001; Paerl et al., 2002). Nitrogen stressors from the atmosphere have been increasing, posing an increasingly serious problem (Galloway and Cowling, 2002). Atmospheric deposition is the primary source of mercury that is bio-accumulated in aquatic systems, affecting insects, birds and humans (Driscoll et al., 2007; Rimmer et al., 2005). Mercury methylation requires sulfur reducing bacteria, thus deposition of both sulfur and mercury is involved in mercury bio-accumulation (Jeremiason et al., 2006). Additionally, atmospheric re-emission and advection can be a key transport pathway for some pesticides (Bossi *et al.*, 2008).

An important aspect of the exposures discussed above is the exploration of changing exposure risk in light of future climate projections facilitated by the connection between air quality and ecosystem function. Atmospheric processes such as precipitation, solar radiation and temperature that drive pollutant deposition are also central drivers of the biogeochemical processes to which an ecosystem responds. The change in these driving variables in response to changing levels of greenhouse gases in the atmosphere will impact ecosystem functions and provision of services in conjunction with atmospheric deposition (C. Driscoll and K. Stolte, personal communication).

Past interactions and research with the scientists in the ecological community have produced ground breaking work linking air and ecosystem models. Atmospheric nitrogen deposition linkage of the Community Multiscale Air Quality (CMAQ) model with the Chesapeake Bay Watershed model (Linker et al., 2000) has gone through several advancements and improvements over the past two decades, revealing temporal and spatial scale issues that arise when trying to communicate between models. For example, airsheds were developed as part of this work showing that sources of air deposition extended well beyond the boundaries of the recipient watershed, illustrating the large difference in spatial scales of analysis (Dennis, 1997; Paerl et al., 2002). Nitrogen deposition simulations were also performed for Tampa Bay in support of the TMDL (Total Maximum Daily Load) analyses, to advance an understanding of how to address management of air quality to best reduce deposition to the watershed and total loadings to Tampa Bay. Air deposition for modeled base case and futures scenarios were linked with aquatic models for a critical loads analysis for Shenandoah National Park to examine the extent of atmospheric deposition reductions needed to support recovery (Sullivan et al., 2008). General analyses using acidic deposition futures were carried out, including the Shenandoah study, to assess the progress of acid rain controls, the progress of ecosystem indicator recovery as a result of controls that have been implemented and estimation of the amount of additional deposition reductions required to attain a prescribed degree of recovery (Driscoll, et al., 2001 and

2003). These analyses uncovered inconsistencies between current deposition estimates using data and future deposition estimates using air quality model projections and the need for multi-year atmospheric deposition model predictions. Ongoing collaborative work between AMAD and the Ecosystem Research Division (ERD) is addressing the linkage of meteorological model predictions of precipitation with watershed models in headwater watersheds to uncover and assess linkage issues at the level of hydrology (Golden, et al., accepted). Discussions are continuing to develop and refine recommendations for priority indicators or measures for climate change studies (C. Driscoll and K. Stolte, personal communication).

From these collaborations, valuable experience on connecting atmospheric model outputs of deposition with ecosystem models has been gained. From our experience, we can characterize the linkage of air and ecosystem models in the following manner: Linkage of atmospheric models to aquatic/watershed and terrestrial models is necessary, but this connection has been more or less ad hoc, often uses incomplete deposition budgets, and suffers some major problems and gaps. However, our experience has been sufficiently rich to lead to the identification of advances that will significantly address linkage issues and critical gaps that, if overcome, will improve cross-communication of air and ecosystem models.

Overall Research Goals

Over the next 5-years, AMAD's goal for air-ecosystem linkage is to:

- Produce exposure estimates of deposition at the spatial and temporal scales responsible for the receptor response

This will involve the marriage of the local ecosystem scale (tens of km^2) with the regional airshed scale (thousands to millions of km^2). The aim is (1) to provide support for SO_X-NO_X welfare standard and critical load assessments and determinations, (2) to support ecosystem and ecosystem services assessments coupled with air management drivers, and (3) to support the assessment of the sustainability of ecosystem health and services under future climate.

Beyond the 5-year time frame, AMAD's goal for air-ecosystem research is to:

- Link deposition exposure models to receptor response models in an integrated, transdisciplinary, one-biosphere manner for current and future climate conditions

This longer term goal supports the vision of advancing towards a one-biosphere perspective.

2. Science and Policy Drivers

The National Research Council (NRC) and the Clean Air Act Advisory Committee (CAAAC) have urged EPA to pay increased attention to ecosystem protection and to develop its capacity in this direction (NRC, 2004; CAAAC, 2005). In addition, the NRC and CAAAC recommended that EPA explore the use of critical loads in the development of secondary National Ambient Air Quality Standards. The NRC further noted that "concentration-based standards are inappropriate for some resources at risk from air pollutants, including soils, ground waters, surface waters and coastal ecosystems. For such resources, a deposition-based standard would be more appropriate." In response to the 2004 NRC report, EPA is considering secondary standards to ensure ecosystem protection and is emphasizing their development. Additionally, the most recent 2005 National Acid Precipitation and Assessment Program (NAPAP) report states "… scientific studies indicate that the emission reductions achieved by Title IV of the Clean Air Act are not sufficient to allow recovery of acid-sensitive ecosystems. Estimates from the literature of the scope of additional emission reductions that are necessary in order to protect acid-sensitive ecosystems range from approximately 40-80% beyond full implementation of Title IV."

EPA is now addressing the secondary standards to protect ecosystems separate from the primary national ambient air quality standards, the first time they have been separated. In the near-term the development and revision of secondary standards is focused on ozone damage from exposure to air concentrations, and adverse effects of acidification, from sulfur (S) and nitrogen (N) deposition, and nutrient enrichment from atmospheric deposition of N. The near-term priority for addressing adverse effects from deposition is development of a combined secondary standard for SO_X and NO_X air concentrations to address acidification since the total deposition of S plus N is acidifying. After acidification, the next priority is to develop secondary standards addressing nitrogen nutrient enrichment of aquatic, estuarine and terrestrial ecosystems. Because the CAA stipulated the National Ambient Air Quality Standards (NAAQS) were for ambient air concentrations, not deposition, the secondary standards must be set in terms of air concentrations and the air quality model serves a critical function to connect air quality and deposition. CMAQ is used to develop an Atmospheric Deposition Transformation Function for the NO_X-SO_X secondary standard development. The role of ammonia in acidic deposition and nutrient enrichment is expected to significantly increase as NO_X and SO_X controls related to O_3 and $PM_{2.5}$ are implemented under the current Clean Air Act and under potentially new welfare standards for NO_X and SO_X, requiring increased attention in the future. Following the setting of new secondary standards, program assessments will be expected to document whether emission control policies are working as intended to achieve ecosystem protection, necessitating reliable estimates of atmospheric deposition and creating a further need to connect air and ecosystem models in a more integrated manner for these assessments.

The management concept of critical loads supports the development of secondary standards. Following the NRC suggestion of exploring the use of critical loads, a multi-agency group working on critical loads (associated with atmospheric deposition) as an approach to ecosystem management for land management agencies has been established. The agencies are the NPS, USFS, USGS and EPA. Additional work on critical loads is also being pursued as part of the U.S.-Canada Air Quality Agreement. As with the NO_X and SO_X welfare standard development, the critical load studies are concentrating on acidification of fresh water and terrestrial systems and excess nutrients in fresh water and terrestrial systems (the latter mostly in the western US, e.g., Geiser et al., 2010). Atmospheric deposition is the source of the critical load being considered at this time. Thus, atmospheric deposition is central to the development of critical loads and estimation of critical load exceedances as a basis for ecosystem management and protection. These deposition estimates also need to be state-of-the-science.

Part of NERL’s mission is to conduct ecological exposure research for the EPA, to carry out its mission to protect human health and the environment (NERL, 2009). Within this mission, NERL has a goal of fostering Integrated Multi-Disciplinary Research (IMDR) which is consistent with the recently articulated ORD goal of developing Integrated Transdisciplinary Research (ITR). These research approaches call for ORD researchers to develop sustainable solutions to environmental problems by engaging partners from multiple disciplines who transcend traditional scientific disciplines throughout the research process. The air-ecosystem linkage research will bring together researchers from at least three NERL Divisions as well as other agencies (as noted in the collaboration section below) forming an important contribution to the NERL goal of creating an integrated, multidisciplinary research program across its Divisions. The research can be a central core, from the atmospheric side, for the development of ITR capabilities for ecosystem research to develop sustainable solutions and for being a multidisciplinary learning laboratory for EPA ORD.

The climate change research program at EPA is equally concerned about ecosystem response as well as air quality and human health. This requires that changes in climate be expressed in ways that can be translated to changes in ecosystem functioning and provision of services. It is critical for climate studies that model-generated meteorology, air deposition and hydrology be internally consistent. Otherwise, the climate signal from meteorological models provided to ecosystem researchers will have an unknown degree of contamination, leading to biased assessments. This calls for air-ecosystem research to advance research efforts to create that consistency.

Air-ecosystem linkages are important to the EPA/ORD’s Ecosystem Services Research Program (ESRP). ESRP has selected nitrogen as a priority pollutant for its research regarding ecosystem exposure, sustainability and the enhancement or degradation in quantity and quality of ecosystem services. The atmosphere is an important source of nitrogen. The linkage of air and watershed models can be an important contribution to the place-based (geographically focused) studies within the ESRP, particularly the Future Midwest Landscape and the Albemarle-Pamlico Watershed studies. State-of-the-science estimates of continental atmospheric deposition are required for the national mapping of nitrogen and ecosystem services research project. The ESRP has as a major long-term research objective answering questions regarding ecosystem sustainability under climate and land use change (change in stress).

Bioaccumulation of deposited Hg and pesticide exposure are important issues associated with toxic pollutants. The proposed mercury regulations, intended to reduce mercury emissions from coal fired power plants, are being revised and there is continued interest in the effects of mercury on humans and on ecosystems. The adequate representation of mercury bi-directional exchange has important implications for assessments of source responsibility, providing important answers to current management questions.

Other agencies are also interested in improved modeling of surface exchange. For example, NH_3 and N_2O surface exchange are important to USDA. Attention is being paid to regulating NH_3 emissions at both USDA and EPA because of the fine particle effects and deposition of nitrogen effects. N_2O emissions are of concern because N_2O is a greenhouse gas affecting climate and it is the single most important ozone depleting substance in the stratosphere.

3. Key Linkage Issues

To gain insight on priority research directions, we interviewed several ecosystem modelers with experience in using atmospheric deposition data in their modeling and critical load analyses. These modelers were asked to identify what they believed were the critical needs to improve the linkage between air and ecosystem models and to overcome some of the critical gaps. Combining their input with our experience and with perspectives from other modeling communities has led us to identify the following list of research issues:

- The paradigm gap
- Spatial and temporal scale issues
- Improved and more complete total deposition estimates
- Precipitation and hydrology issues

Many of the research needs associated with these issues are discussed in Seigneur and Dennis (in press) as part of the NARSTO assessment of multi-pollutant air quality management. Regional air quality models would appear to provide the best, universal approach and spatial coverage for linkage with ecosystem models. They will be more acceptable to the ecosystem modeling scientists if many of these issues can be addressed or reduced; however, they most likely cannot be eliminated. The paradigm gap is fundamental and provides an overarching context for setting the stage to define and address the other three specific research issues.

The Paradigm Gap

There is a major paradigm gap that thwarts easy linkage of atmospheric deposition predicted by regional atmospheric models to ecosystem models. This paradigm gap affects ecosystem studies and especially affects climate change studies. This gap stems from a fundamental lack of sufficient universally applicable ecosystem process information and too many degrees of freedom regarding the behavior of the system to perform the modeling from a universal applicability perspective. This limitation leads to a heavy reliance on calibrated ecosystem models. While calibrated models perform well for the particular system and historical input data (scenarios) for which they are calibrated, the models are not easily generalizable to other ecosystem locations and other inputs and errors in input data may invalidate the model. Atmospheric models, on the other hand, work from a perspective of universal applicability (first principles) so that the same model parameterizations can apply everywhere and at anytime and the models can be applied for multiple input data sets and they do not require calibration or long spin-up times. However, the downside of this approach is that outputs of the atmospheric models have error, at times significant error, relative to observations. Of particular concern are errors in the location, amount and timing of precipitation and in the associated water balance and surface water hydrology. These errors typically present challenges in linking with the ecosystem models because the atmospheric modeling system predictions do not match the conditions used to calibrate the ecosystem model. The paradigm gap is expected to introduce potentially serious errors in climate change analyses. In these analyses, an inconsistency is created by mixing the calibration base case with the futures prediction from the atmospheric model; the base case used by the ecosystem models is not consistent with the hydrologic balance of the base or the futures case coming from the atmospheric model. As a consequence, an artificial degree of change, a bias, can be introduced into the analysis of change which is particularly problematic for the assessment of climate change impacts. The following three issues should be viewed in the context of the paradigm gap because it structures our research response to them. The paradigm gap particularly underlies the precipitation and hydrology issue. Although many aspects of the following issues will be addressed over time, the paradigm gap will not soon disappear.

Spatial and temporal scale issues

Since the ecosystem responses to stressors are controlled by local conditions and local exposures, ecosystem models require very fine spatial resolution to characterize the hydrology and distinguish deposition to small catchments and areas with varying topography and geology. However, the atmospheric models that are needed to provide a complete characterization of the atmospheric emissions and chemistry operate at a coarser level. A challenge of air-ecosystem linkage is the marriage of these two very disparate scales. Management strategies used in ecological studies need to recognize that airsheds for the local deposition impacting ecosystems are large and regional in extent (see Paerl et al., 2002). It is known that differences in dry deposition arise as a function of different types of land cover. Deposition to specific vegetation species or land cover types is needed for input to the ecosystem models while typical regional air quality models provide one dry deposition value per grid cell. Differences in deposition to different vegetation types can be addressed at the sub-grid level, however, the land cover specifications need to be consistent. Land use specification is important to both ecological and atmospheric models, but they have not historically been consistent. Therefore, it will be important to update and harmonize the land use data in CMAQ with those routinely used by the ecological community. Using this same finer scale base data and aggregating to regional data will provide an important consistency between the models. The capability to vary land use to study the effects of land use change is an investigatory area of great interest to the EPA Ecosystem Services Research Program and the greater ecological modeling community. In areas of complex terrain dry deposition can vary significantly due to differences in terrain influences on meteorology and air concentrations. Empirical studies have examined the range of deposition values in complex terrain that might be expected within a grid and underline the need for local, spatial detail in deposition estimates for critical load studies (Weathers et al., 1995 and 2006). Developing these data from regional model output presents a challenge. Terrain influences may be able to be partially addressed by development of empirical surrogate relationships of dry deposition variation with terrain features that are available from GIS information and maps. Such semi-empirical sub-grid models of dry deposition variability could further reduce the dry deposition uncertainty in complex terrain and would be of great interest to the ecosystem modelers.

The temporal scales of interest in ecosystem models can vary from daily to annual time scales and may be sensitive to short-lived episodic events (e.g., flooding events, intermittent toxic releases, and pesticide applications), while air models operate hourly and are often aggregated up to longer averaging times. Also of issue is the need for multiple years of atmospheric deposition for establishment of the base case because the ecosystem models are typically calibrated using several years of data. In addition, several ecosystem models require 150 years for a spin up period, starting with pre-industrial conditions and some climatic events of interest such as droughts occur over prolonged time periods, e.g., months to decades.

Improved and more complete total deposition

Ecosystem analyses need to include total deposition (both wet and dry) for a more accurate representation of the deposition budget. For greater credibility, wet deposition estimates should be improved to address biases and known missing processes. A summertime under-prediction bias is hypothesized to be associated with the omission in CMAQ of a parameterization for the production of NO_X aloft due to lightning. Two missing processes of concern to the ecological community are cloud impaction and dissolved organic nitrogen (DON). Cloud-water impaction at high elevation is an important source of deposition in complex terrain affecting critical load calculations. DON is estimated to constitute approximately 25% of the total wet nitrogen deposition. DON from the atmosphere is now thought to be an important source of nitrogen for ecosystem exposure and is considered to be a missing process in air deposition models that requires attention. Other chemical budgets important to ecosystem biogeochemical processes, in particular base cations, are not represented in the atmospheric models for either wet or dry deposition but should be addressed in future model development. Dry deposition is an important part of total deposition yet this component is typically missing from many ecosystem analyses because of the sparseness and incompleteness of measured dry deposition data. Dry deposition's magnitude is often equal to or greater than that of wet deposition. Since atmospheric models include a more complete characterization of the chemical species than most monitoring networks, using the dry deposition from atmospheric models provides ecosystem models with a more complete accounting of the pollutant budget. The importance of an air quality model for dry deposition estimates creates a responsibility and need to reduce the uncertainty in the model estimates.

Reducing uncertainty in the dry deposition algorithms especially involves keeping them state of the science. This includes incorporation of bi-directional air-surface exchange where it is critical and evaluating dry deposition algorithms with new measurements. It is well established that air-surface exchange is bi-directional for NH_3 (Walker et al., 2006) and Hg, two very important species involved in ecosystem effects. Other species, for example semi-volatile pesticides, may also need to be considered in the future. Formulations for bi-directional air-surface exchange with terrestrial and aquatic surfaces are required in CMAQ for scientific credibility (keeping it state of the science). Incorporation of bi-directional exchange is expected to significantly influence the estimated range of influence of a source and the estimation of source attribution as well as the wet and dry partitioning of total NH_3 deposition (Dennis et al., in press). Second, the evaluation of dry deposition algorithms has received limited attention due, in part, to lack a of measurement methods capable of supporting field study investigations of dry deposition. The importance of the sulfur, nitrogen and mercury species to deposition and ecosystem effects is now well accepted. Advances in instrument sensitivity and sampling frequency for key species have occurred that should create new opportunities for evaluation of deposition algorithms. The implementation of these advances in collaborative field studies should be encouraged to develop further credibility in CMAQ's dry deposition estimates.

Precipitation and hydrology issues

Errors in the meteorological model predictions of precipitation influence estimates of wet deposition, introducing errors in them. It is a real concern that the meteorological model predictions of precipitation location, amount, and duration have levels of error such that the predicted precipitation is inconsistent with the hydrologic response in the real world. Ecosystem models are calibrated on observed precipitation and hydrology to develop a water balance. Given the calibrated nature of the ecosystem models, observed precipitation and wet deposition are the preferred inputs for the ecosystem models because of the large errors in the prognostic model precipitation estimates. Unfortunately, this produces inconsistent total chemical budgets across models, due to the paradigm gap, when an air model is used to provide deposition estimates. It is important to remove/address this inconsistency (bridge the gap) to establish successful ecological and air quality model linkage. This suggests a necessity to link hydrology to the meteorological model's precipitation predictions to create an internally consistent data set of precipitation and hydrology as one of the research directions. It is also critical to improve our ability to simulate precipitation at the fine scale to improve the linkage with the hydrologic models and to improve the accuracy of the wet deposition estimates. Additionally, simulated precipitation and temperature improvements to account for orographic effects are needed in complex terrain and mountainous settings which are home to many sensitive ecosystems in the U.S. This is a challenging issue and the level of improvement needed may need to be established by a risk assessment of the sensitivity of the receptor of interest to the variety of precipitation-driven stressors (e.g., exposure risk to precipitation inputs), rather than metrics associated with comparisons against observed precipitation data.

4. Key Science Questions

Science Questions

The issues in the previous section need to be addressed to achieve the research goals for air-ecosystem linkage articulated above. Science questions associated with the issues are posed to help formulate components of the research. The science questions are:

- What are the sources of deposition bias and how can they be resolved?
- How can air-ecosystem spatial and temporal scale mismatches be addressed or resolved?
- What atmospheric processes are missing or not sufficiently state-of-the-science and what is needed to incorporate or improve them to be state-of-the-science?
- What are the sources of the paradigm gap between CMAQ, a universal, generalized parameter model, and calibrated ecosystem/watershed models? What approaches will bridge the paradigm gap?
- How will climate and land use change impact atmospheric composition, biogeochemical cycling and ecosystem health?

Related Management Questions that Create Science Questions

- Which sources of atmospheric emissions contribute to terrestrial and aquatic ecosystem degradation?
    - Where is the deposition coming from; where is it going to?
        - How large and where are the airsheds? What is the range of influence?
    - Who is the deposition coming from?
        - Economic sectors
        - Geographic regions
- What is the accuracy of the deposition estimates?
- What is the "policy-relevant" background for nitrogen deposition?

5. Research Directions and Approach

To answer these science questions requires a coordinated research strategy covering a variety of activities and tasks to best use AMAD's limited resources. The strategy involves: (1) Improvements in CMAQ's bi-directional and uni-directional air-surface exchange characterizations, evaluation of these improvements, and quantification of the uncertainty of these estimates; (2) Harmonization of land cover with ecosystem models and improvements in the capability and flexibility of the WRF/CMAQ land surface characterizations and air-surface exchange in complex terrain; (3) Incorporation of missing deposition pathways into CMAQ; (4) Linkage of hydrologic models with WRF/CMAQ to provide a consistent connection between precipitation and hydrology for a system-consistent linkage to ecosystem models and development of improved WRF precipitation fields; (5) Research on close coupling of the WRF/CMAQ/hydrology system with watershed models to test fidelity regarding biogeochemical cycling; (6) Application of expanded model capability to simulate ambient air-quality and ecosystem exposure to support assessments addressing sensitive ecosystems and ecosystem services and assessments of scenarios depicting climatic and land use change, particularly climate change. The applications may involve post-processing CMAQ deposition data for critical loads assessments and error analysis.

(1) Further Develop CMAQ Air-Surface Exchange

Directions: Improve the parameterizations of the air-surface exchange of atmospheric pollutants, both bi-directional and uni-directional for aquatic and terrestrial landscapes. Improvements will target vegetated canopy atmospheric resistances and the relative partitioning between deposition to physiologically mediated leaf tissue and vegetation and soil surfaces. Develop techniques to account for deposition of pollutants at high elevations *via* impaction of cloud water.

Approach: CMAQ currently estimates bi-directional exchanges of NH_3 over terrestrial surfaces and of Hg over terrestrial and aquatic land surfaces. This is a first for a 3-dimensional regional air quality model. Deposition and emission fluxes are simultaneously estimated for NH_3 based on the two layer canopy compensation point resistance model of Nemitz *et al.* (2001) and Hg following the two layer canopy compensation point resistance-capacitance model of Sutton *et al.* (1998). The ambient concentration at which the net air surface exchange is zero, i.e., the compensation point, in both the NH_3 and Hg surface exchange models is dynamic and solved from bi-directional exchange estimates from air-soil, -vegetation surface, and -stomatal interfaces. Quantification of the deposition to the canopy and soil components with different physical and chemical resistances provides insights that can improve the net uni-directional deposition estimate and the net bi-directional exchange flux. The prototype NH_3 bi-directional exchange model uses *a priori* specified emission potentials for soil, vegetation surfaces, and stomatal exchange taken from the literature, while the Hg bi-directional exchange model parameterizes dynamic compensation points using partitioning coefficients and first order reduction of previously deposited divalent mercury in the surface media published in the literature. The NH_3 bidirectional exchange model will be refined to use dynamic fertilizer application rate inputs to be able to derive *in situ* emission potentials from fertilized agricultural soils. This will be accomplished by development of a Fertilizer Emissions Scenario Tool for CMAQ (FEST-C) based on the USDA Environmental Policy Integrated Climate (EPIC) model in collaboration with the USDA. This tool will become part of the CMAQ input processing system. FEST-C will first treat chemical fertilizer application, but will then be extended to deal with manure application, dependent on having the relevant driving information. The NH_3 bidirectional exchange model will also be expanded to include bi-directional exchange over aquatic ecosystems. The Hg bidirectional model currently assumes that vegetation is a sink for divalent mercury species due to the lack of a published reduction mechanism for divalent mercury on vegetation surfaces and uses a simple single layer water surface model to estimate air-surface water exchange. Bi-directional Hg exchange model algorithms will be refined to include the reduction of divalent mercury species on vegetation surfaces and a multiple compartment parameterization of surface waters. Furthermore, other pollutants, most notably persistent organic pollutants (POPs), exhibit bi-directional exchange that can significantly enhance the transportation and ecosystem exposure. Future research and collaborations will expand the bi-directional model to include POPs and other semi-volatile pollutants that exhibit long range transport and are responsible for deleterious effects on ecosystem health.

The parameterization of bi-directional surface exchange requires more detailed vegetation and soil parameters and field data to guide the parameterizations. AMAD model development and evaluation teams will engage in collaborative development work with other EPA laboratories, such as NRMRL, and federal and academic research institutions to advance surface exchange parameterizations. AMAD will partner with process-oriented scientists, at EPA and elsewhere, engaged in laboratory and *in situ* field measurements, *via* collaborations to gather data needed to advance in-house air-surface exchange, bi- and uni-directional, model development and evaluation. Top down and bottom up inverse modeling techniques using satellite and special field observations will be used to assess the sensitivities of the newly developed model air-surface exchange parameterizations and to identify sensitive model variables and variables that are not routinely measured.

(2) Harmonize Land Surface Characterizations and Address Subgrid Variability of Air-Surface Exchange

Directions: Advance and modernize the WRF/CMAQ land surface parameterizations and leaf area index (LAI) estimates to improve connections or linkage to ecosystem models, to address paradigm mismatches and develop community confidence in the system. Develop sub grid scale land use specific deposition estimates, and improve meteorological and dry deposition simulations in complex terrain at higher spatial resolutions. Develop parameterizations of sub grid variation in dry deposition due to complex terrain.

Approach: CMAQ, MM5, and WRF have historically used 1 km land use data from the 1992 National Land Cover Database (NLCD) with categories defined using the USGS 24 category system. An updated version of the NLCD is now available which represents land cover from 2001 with a resolution of 30 m. This new data set is being incorporated into WRF and CMAQ along with MODIS data for Mexico and Canada. The native NLCD and MODIS land use categories will be retained as this is consistent with the categories used by ecosystem models and will facilitate linkages between the air and ecosystem models. Additional capabilities will be developed to allow modification of the land use designations to explore future scenarios of land use change and their impact on air quality and air-surface exchange.

Currently, CMAQ output consists of grid-averaged deposition velocities and fluxes. Deposition depends greatly on the characteristics of the underlying surface and therefore will vary with land cover type. Ecological applications often require knowledge of the deposition to individual land cover types within a grid rather than the grid-averaged value. CMAQ is being modified to calculate and output sub-grid scale land-use specific deposition estimates (Mosaic approach) which will provide the information needed for ecosystem assessments. Making these changes within CMAQ rather than the meteorological model allows the use of previously generated meteorological files rather than requiring the meteorological model to be rerun.

Leaf area index (LAI) is an important input to the land surface model in WRF and to the deposition algorithms in CMAQ. The current method for obtaining LAI for WRF and CMAQ uses the deep soil temperature for predicting leafout and crop growth. Fixed land use category maximum and minimum values for LAI are provided in the models. Alternative sources such as the EPIC model may provide similar input for alternative land use and climate scenarios. Advances in remote sensing technology provide an opportunity for developing techniques for using satellite data to obtain spatially and temporally explicit estimates of LAI for input to the models. Collaboration with ESD’s Landscape Characterization Branch will be pursued to further this approach.

Deposition in complex terrain is poorly understood and not often measured. Consequently, little air quality model development has occurred in this area. However, numerous sensitive ecosystems occur in complex terrain and current deposition estimates to these areas are likely inadequate to provide the information needed for use in, for example, critical loads deposition assessments. While WRF and CMAQ model long term development will work to improve meteorological and surface exchange capabilities in complex terrain at higher spatial resolutions, there is a need to find alternative means to improve these estimates for current deposition assessments. AMAD will need to partner with ecosystem scientists to collect sub grid-scale deposition or air concentration data in complex terrain for sulfur, nitrogen and mercury species to empirically tie within-grid dry deposition variability with GIS-available metrics.

(3) Address Missing Deposition Processes

Directions: Incorporate processes or pathways that are currently known to be missing in CMAQ that have an important effect on deposition or are highly desired by the ecosystem modeling community. Interpret model evaluation results and error analysis in terms of missing processes to further identify candidates related to missing processes, pathways or emissions, as distinct from spatial issues of emissions accuracy.

Approach: Recent model evaluation analyses have identified the lack of a lightning NO_X generation processes in CMAQ as a source of a major summer under-prediction bias in wet nitrate deposition and NO_X concentrations aloft in the free troposphere. Collaborative research with NASA is culminating in a parameterization that can be driven by WRF to include lightning-generated NO_X in CMAQ.

Cloud impaction is an important source of wet deposition in mountainous, complex terrain that is important to terrestrial critical load modelers, but it is not computed by CMAQ. CMAQ can be modified to include a model of cloud impaction. The initial approach will be to incorporate a simple model, such as that described in *Katata, et al.* (2008), to meet the needs of the critical load modelers without creating a large computational burden in CMAQ.

Base cation deposition, mainly from soil emissions, is highly desirable for acidification calculations for aquatic systems to help complete the chemical balance driving these systems, but CMAQ does not track these emissions at this time. New work to help PM2.5 non-attainment modeling is characterizing and identifying the species involved in soil emissions. When it is completed under the EPA Air Program, this work will allow CMAQ to provide ecosystem modelers with base cation deposition estimates.

Dissolved organic nitrogen (DON) deposition is an important source of nitrogen through wet deposition that is currently not incorporated in current air deposition models. The difficulty is that the chemical constituents are many and not well known, coupled with a lack of understanding of where they are coming from. This means that we do not currently know how to characterize emissions of DON species. Collaborations with EPA/NRMRL and with the National Atmospheric Deposition Program will provide empirical data on the amounts of DON depositing across the country under different mixes of land use. These data will be assessed to provide more information to guide the next steps for developing interim estimates of DON deposition and incorporating DON into CMAQ.

(4) Link Hydrological Processes and Models to WRF/CMAQ and Improve Precipitation Fields

Directions: Extend the WRF meteorological model to provide an internally consistent representation of the hydrologic cycle, including precipitation, soil moisture, evapotranspiration and overland flow. Focus on the elements of the hydrological cycle that impact ecosystem loading and exposure of atmospheric pollutants. Provide an internally consistent representation of precipitation and overland flow. Also, focus on the elements of the hydrological cycle that affect the response of ecosystems to climate change. Develop a linkage between climate-model-produced-precipitation and associated modeled hydrological processes to address paradigm mismatches and support ecosystem exposure scenario estimates under climate change.

Improve the modeling of amount, location, and duration of precipitation by WRF to reduce the error in wet deposition estimates from CMAQ. Extend the WRF precipitation predictions to finer grid scales, such as 4 km, with a particular emphasis on regions with complex terrain. Further develop techniques for accounting for occult deposition, particularly cloud-water deposition in mountainous terrain.

Approach: Develop extensive collaboration with other research groups within EPA, other federal agencies and academia to extend WRF/CMAQ capabilities to include hydrology consistently linked to WRF. AMAD (ecosystem and climate teams) would develop the linkage for retrospective WRF and down-scaled WRF climate simulations with the intent of providing national coverage. Primary groups for collaboration are the NOAA Hydrology Laboratory of the National Weather Service (NWS), the US Geological Survey (USGS) and Pacific Northwest National Laboratory (PNNL). AMAD/EPA will sponsor a workshop co-sponsored with USGS and NOAA to develop a sound research direction for this work and develop a conceptual plan to guide the research. A major consideration is how tightly the hydrology should be coupled with the meteorological model's internal water budget in order to provide some degree of feedback or whether the hydrology should be purely calculated off-line as a post-processing step. Attention will also have to be paid to within-grid heterogeneity of the hydrologic processes affecting evapotranspiration. Land surface models for WRF to be considered include the NOAH Land Surface model, the Community Land Cover Model (CLM4) model and the Variable Infiltration Capacity Model (VICM). Hydrology models to consider for adaptation in this effort could be the NOAA/NWS Hydrology Laboratory Research Distributed Hydrologic Model (HLRDHM), which is operational at 1 km grid size and the USGS Precipitation Runoff Modeling System (PRMS). The connection would be targeted for WRF with a 12 km continental domain and 4 km regional test domains. In conjunction with recommendations from the planning workshop, AMAD would consult with NOAA and USGS on approaches to establish the degree of error introduced by the use of larger WRF grid sizes. The NOAA Earth System Research Laboratory Hydrometeorological Testbed (HMT) that is planned to start in 2010 in the Neuse Basin in North Carolina will collect high temporal and spatial precipitation and hydrological data that will be a potential source of test data for the WRF-hydrology linkage. The ecosystem and climate teams within AMAD will coordinate closely on working with the down-scaled meteorology to produce consistent hydrology for climate change studies. AMAD and ERD will coordinate closely on evaluating the influence of the internally consistent hydrology predictions on watershed model water balance and chemical biogeochemical response predictions. This research is a major undertaking that will coordinate with the larger meteorological and hydrology community.

Improvement in WRF precipitation simulation for linkage of air quality to ecosystem and watershed models will arise from improved/expanded data assimilation, improved boundary condition definition and improved model physics, in addition to advances in land surface modeling stemming from the above work to link hydrology with WRF. Advances in these areas will come from a combination of in-house AMAD research and collaborative research with the WRF community. In-house scientists have incorporated an "obs-grid" data assimilation approach, to good effect, and are expanding assimilation to include soil moisture (e.g., Pleim and Gilliam, 2009) which will improve surface heat and moisture flux simulation from soil and vegetation. Long-term research by UNC scientists is yielding promising results in expanding the application of 3-D variational techniques to include, for example, radar data assimilation and scientists at the University of Alabama are exploring the assimilation of satellite data to improve cloud characterization.

Recent WRF simulations make use of advanced data products such as the 12-km North American Model (NAM) analysis to provide model boundary conditions (Appel, et al, 2009), but these data are not available prior to 2005. In-house research is exploring the use of alternative, higher resolution reanalysis data such as the North American Regional Reanalysis (NARR). The precipitation errors in complex terrain at different grid resolutions will be quantified using the Parameter-elevation Regressions on Independent Slopes Model (PRISM) data set. AMAD will continue to explore the value of the radar-based precipitation estimates in complex terrain for establishing ground truth. Tests will compare performance at 12 km and 4 km grid sizes. Collaboration will be developed with researchers interested in improving WRF predictions in complex terrain, including precipitation predictions, such as researchers at the University of Washington. The University of Washington research is also examining WRF simulations at 4 km grid sizes. Improvements derived from these enhancements will be evaluated with scientists at ERD and Syracuse University in light of improved hydrologic model performance.

Research regarding the incorporation of improved model physics will follow the same successful in-house/external collaboration design. For example, in-house scientists have successfully implemented the Asymmetric Convective Model 2 (ACM2; Pleim, 2007a, b), but further refinements are being explored through collaboration with scientists at Texas A & M regarding the use of Kalman filtering techniques to refine ACM2 parameter values.

(5) Explore Close Coupling of WRF/CMAQ/Hydrology and Watershed Models and Identify One-Biosphere Linkage Issues

Directions: Assess the performance of watershed models using internally consistent precipitation and hydrology input from the WRF/CMAQ system as compared to using observed precipitation and hydrology. Assess the fidelity or robustness of ecosystem biogeochemical response when internally consistent precipitation and hydrology is provided to bridge the paradigm gap between air and watershed models. Do these data bridge the gap or do they introduce a lack of robustness of response to stressor change? Address the fidelity and robustness of assessing ecosystem response to climate change with linked air-watershed biogeochemical models with exposure inputs based on a down-scaled WRF/CMAQ/Hydrology system.

Approach: Conduct collaborative, multi-model assessments of (1) the impact of precipitation errors on watershed water balance estimates, (2) the impact of the WRF vs observed precipitation differences on predicted biogeochemical cycling of nitrogen, (3) the impact of the internally consistent WRF/Hydrology data on watershed water balance modeling, and (4) the impact of the internally consistent WRF/CMAQ/Hydrology data on biogeochemical cycling of nitrogen, particularly with respect to assessing the effects of climate change. AMAD will evaluate the internally consistent meteorology/hydrology/chemistry from the standpoint of the robustness of assessments of ecosystem exposure scenarios using calibrated biogeochemical models.

AMAD will collaborate with NERL/ERD and academia to compare hydrologic routing and depiction of the watershed water balance based on input of WRF precipitation with the routing predicted by calibrated watershed and ecosystem models, such as the Grid Based Mercury Model (GBMM) of ERD and the PnET-BGC model of Syracuse University, an integrated biogeochemical model developed to simulate forest and aquatic ecosystems, calibrated on observed precipitation and hydrology. This will eventually be compared to the hydrologic routing produced by the WRF/Hydrology linked model set when those data are available. AMAD will also collaborate with NERL/ESD to test and compare the hydrology predictions from the WRF/Hydrology system and GIS-based models for small catchment regions and examine the ability to represent regional hydrology with available GIS data layers. AMAD will evaluate biases between observed and modeled hydrological processes and their impact on the ecosystem model predictions. We would be looking for internal consistency and ability of the WRF/Hydrology model set to support calibrated ecosystem models and test whether a "calibration" based on the WRF/Hydrology linked model set is distorted or is basically equivalent relative to a calibration based on observed data.

AMAD will collaborate with NERL/ERD and academia to evaluate the response surface of the processing/cycling of nitrogen predicted by biogeochemical models under changing environmental conditions. The response surface and its position in parameter space will be evaluated for a suite of inputs with the hydrology and chemistry based on several different input data sets: observation-based inputs, WRF-precipitation-based input, WRF/Hydrology system input, and WRF/CMAQ/Hydrology system input. The goal is to establish whether the nitrogen response surface is significantly moved in parameter space due to the use of the internally consistent WRF/CMAQ/Hydrology system set compared to a set based on observations that is consistent with the model calibrations. This research will provide guidance on how best to link the WRF-consistent hydrology with ecosystem models for management and climate change studies. AMAD will also develop guidance on how best to provide hydrology coupled to precipitation for critical load models such as the Model of Acidification of Groundwater in Catchments (MAGIC) that do not have internal routing capability. The ecosystem and climate teams within AMAD will coordinate closely on working with the down-scaled meteorology to produce consistent hydrology for climate change studies based on the guidance developed above. The hydrology will be characterized for a climate study period to establish a consistent baseline for climate change studies involving ecosystem response to changes in precipitation that avoid or at least mitigate the paradigm gap.

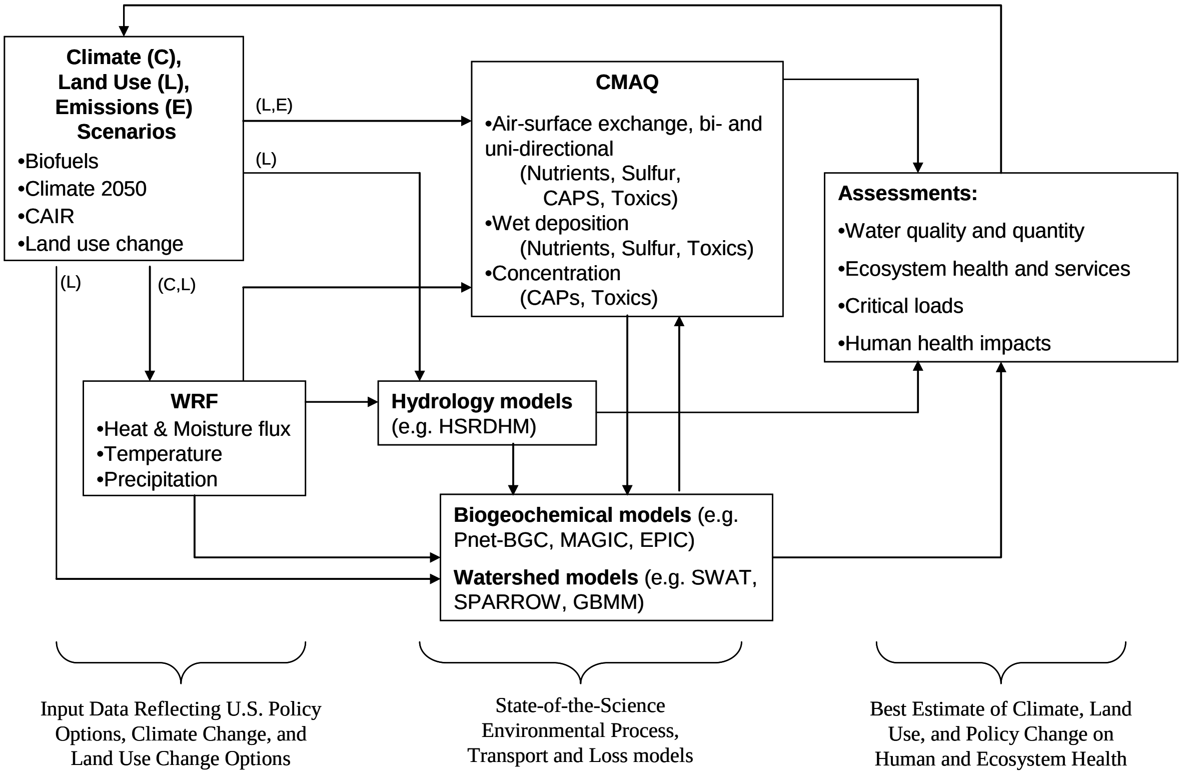
(6) Application of Expanded Model Capabilities

Directions: Assess the ability of CMAQ and WRF to address current and future ecosystem exposure, and the development of future policy through modeling scenarios with policy and climate change forcing. Target critical load studies and climate change assessments. Communicate information on uncertainties to client stake-holders.

Approach: Develop targeted model applications to support EPA, federal agency and academic partners. Potential application studies are expected to be EPA SO_X-NO_X welfare standards assessments, Chesapeake Bay TMDL studies, multi-agency (NPS, USFS and EPA) critical loads assessments (deposition) and Ecosystem Services Research Program (ESRP) scenario assessments involving land use change and climate change. AMAD will provide CMAQ deposition and air quality estimates for the ESRP research involving nitrogen at the national and regional level. AMAD will develop post-processing procedures to address precipitation error and potential emission errors, leading to estimates of uncertainty of relevance to the stake-holder communities. AMAD will engage in providing improved and post-processed deposition estimates for critical load studies conducted by EPA and the other federal agencies, learning to enhance the use of CMAQ deposition estimates for this purpose. AMAD will engage in critical load inter-comparisons with Canadian models under the US-Canada agreement between EPA and Environment Canada. Key variables driving ecosystem response to climate change will be identified with academic and federal partners, and guidance will be developed on metrics for assessing the impacts of climate change. The AMAD ecosystem team will develop studies to examine the potential impact of climate change on ecosystems and ecosystem services based on the AMAD down-scaled meteorology in coordination with the AMAD climate team. Results from these application studies will further guide in-house model development and application. AMAD will use CMAQ as a laboratory to study the interactions with deposition and the hydrological cycle to spur the development of methods and metrics for the application of regional air quality models to ecosystem exposure studies under climate change scenarios.

A conceptual diagram of how the component parts of the above research integrate into the overall modeling system to provide the capabilities needed to address air-ecosystem linkage and climate change research needs, and how to answer the science questions is shown below.

Linking Air Quality to Ecosystems and Watersheds to assess Land Use, Land Cover and Climate Change Effects



Implications for Model Development, Evaluation and Application Planning

Extend and refine bi-directional exchange algorithms in CMAQ

- Collaborative development of advanced bi-directional exchange (emission and deposition) algorithms for Hg, NH_3 and other bi-directionally mobile species for water and terrestrial (soil and vegetation) surfaces. Include improved modeling of NO and the addition of N_2O**.**
- Collaborative development and evaluation (with USDA) of the fertilizer tool for creating NH_3 emissions from soils due to fertilizer application to support NH_3 bi-directional exchange calculations in CMAQ**.**
- Collaborative development and evaluation (with USDA) of the bi-directional flux and regional transport of pesticides in the WRF/CMAQ system.
- Develop adjoint model for NH_3 bi-directional exchange version of CMAQ

Harmonize air-ecosystem and WRF/CMAQ land surface characterizations

- Convert WRF/CMAQ to use NLCD land use and the CMAQ Mosaic approach to estimate dry deposition output by land-use category within a grid.
- Establish consistent treatment of variables that are common, e.g., land cover, soil across CMAQ, WRF, BEIS/MEGAN, EPIC, etc.

- Enhance national-scale vegetation distribution information (e.g., LAI) and expand to include additional remotely sensed and ground-based survey (e.g., USDA Cropland Data Layer, USGS LANDFIRE) data sources (ESD).

Address missing or inaccurate process descriptions

- Enhance WRF and CMAQ capabilities to simulate complex terrain using a smaller grid size (e.g., 4 km) **.**
- Address missing process descriptions
  - Lightning NO_X
  - Cloud impaction in high elevation terrain
  - Base cation deposition
  - Approach to address dissolved organic nitrogen (DON) deposition

Extend tool capabilities related to agricultural and land use management

- Enhance WRF and CMAQ to respond to more specific land cover information e.g., corn, peanuts as opposed to "agricultural land", wetlands, irrigated lands, etc.
- Integrate the capability to model the effects of agricultural management and non-point source pollution releases beyond fertilizer application into CMAQ to improve current land surface parameterization.
- Upgrade land-surface model to account for water budgets and denitrification of water-column N and potential production of N_2O (greenhouse gas).
- Link CMAQ flux algorithms (e.g., early morning release of ammonia and agri-chemicals that have accumulated in the vegetation canopy overnight) to local-scale human exposure.

Enhance ability to address within-grid deposition variability

- Develop new CMAQ output options e.g., MOSAIC to estimate dry deposition output by land-use category and the Watershed Deposition Tool to better communicate CMAQ simulation outcomes to our hydrologic and ecosystem exposure clients.
- Enhance land-surface model to account for fine scale processes and complex terrain, particularly as it relates to sub-grid scale dry-deposition variability.
- Develop a GIS-based approach with CMAQ that uses sub grid information on terrain and other key variables to account for sub grid variation in dry deposition due to variation in terrain.

Develop link between WRF and hydrology models. Improve WRF precipitation.

- Develop and evaluate a post-WRF/CMAQ processing tool or system able to estimate hydrology metrics such as runoff that are consistent with modeled (e.g., WRF-generated) precipitation.
- Enhance CMAQ-hydrology linkage to subgrid hydrology and biogeochemical models such as the Pnet-BGC systems for aquatic systems response to assess critical loads and climate change.
- Improve WRF precipitation simulations, including extremes to support linkage to calibrated hydrological models, to support estimation of ecosystem services and ecosystem exposure.
- Improve and evaluate downscaled climate precipitation scenarios against historical climate means and variability to establish credibility in the ecological and hydrological

communities. Exploration of alternative methods of using existing precipitation scenarios (e.g., ensemble approaches).

Advance model evaluation and model intercomparison techniques

- Evaluate CMAQ atmospheric deposition predictions and define biases, input errors, and missing input components, such as base cations, and those relating to the nitrogen budget (e.g., lightning NO_X).
- Develop additional CMAQ tools in-house and through collaborations for addressing source apportionment management questions (e.g., DDM-3D).
- Conduct an inverse modeling study to examine differences introduced by the NH_3 bi-directional parameterization.
- Conduct model intercomparisons with Canada on critical load calculations.
- Design approaches to post process CMAQ deposition to reduce error (e.g., precipitation adjustment of wet deposition or data fusion) to obtain most accurate deposition estimate.
- Develop new diagnostic output options, e.g. flux components as opposed to net flux, additional chemical species to make better use of collaborative field study results for model development and evaluation.

Advance model application capability

- Assess barriers and potential approaches to development of long term multi-year CMAQ deposition values needed by biogeochemical/watershed models
- Conduct targeted research applications
  - Critical load/Secondary Standards/TMDL Studies
  - Ecosystem Services Research Studies for national atlas, and place-based research oriented towards nitrogen
  - Water quality/water quantity studies
  - Climate change studies with down-scaled meteorology

Anticipated Major End Points

1-2 Years

CMAQ with advanced bi-directional air-surface exchange algorithms incorporated for NH_3 and Hg and updates to uni-directional dry deposition algorithms.

Lightning NO_X included in CMAQ (addressing a missing pathway)

FEST-C available to the community for retrospective and near-term assessments.

CMAQ with the Mosaic option implemented to output subgrid dry deposition by land use type and fraction.

Land use fully harmonized between WRF, CMAQ and ecosystem models (NLCD land use).

Method developed to post-process WRF precipitation and CMAQ wet deposition predictions to reduce wet deposition error for use in, for example, critical load studies.

CMAQ sulfur and nitrogen deposition results with bi-directional NH_3 available for multi-agency studies of critical loads in collaboration with the Office of Air Programs/Clean Air Markets Division.

Atmospheric deposition scenarios for Ecosystem Services Research Program national and place-based study assessments.

2-4 Years

Bi-directional NH_3 adjoint version of CMAQ operational

Continued CMAQ air-surface exchange evaluations against new flux data available through collaborative research efforts in coordination with Clean Air Markets Division

Cloud water deposition and tracking of base cations included in CMAQ (addressing two missing pathways)

WRF improvements implemented based on 3-D variational assimilation techniques and WRF physics options and data assimilation approaches defined that produce the most accurate simulation at a 4 km grid size.

Harmonized soils between WRF, CMAQ, FEST-C or EPIC, ecosystem and hydrology models.

FEST-C available to the community for land use change and climate change assessments.

Development of linkage between WRF and a hydrology model accomplished.

NH_3 inversion modeling initiated with new NH_3 adjoint version of CMAQ to replace previous investigations

Model intercomparison with Canadian models of critical load calculations for US-Canada Accord (follow-on the AQMEII model intercomparison)

Application study support for the next round of NO_X-SO_X welfare standard setting process and key TMDL assessments in collaboration with NCEA and OAQPS

Atmospheric deposition scenarios for Ecosystem Services Research Program national and place-based study assessments and for critical load assessments for alternative scenarios.

Preliminary investigations of the effects of land use and climate change on ecosystem health and water quality using new WRF/CMAQ/Hydrology systems

3-6 Years

CMAQ with bi-directional air-suface exchange of select pesticides developed in collaboration with USDA

Preliminary approach developed to address dissolved organic nitrogen (DON) in CMAQ (address a missing pathway)

Continued CMAQ air-surface exchange evaluations against new flux data available through new mobile flux platform deployed at selected CASTNet sites in coordination with Clean Air Markets Division

WRF/CMAQ with advanced hydrology surface layer developed with an uncertainty characterization

Initial advanced subgrid variability estimates for dry deposition developed for application studies.

FEST-C advanced to consider nitrogen fate and N_2O production for climate change assessments.

Guidance developed for linking WRF/CMAQ with ecosystem models to address the paradigm gap. Guidance developed for linking WRF/CMAQ with coupled hydrology to ecosystem models. Guidance developed for creating climate baseline meteorology and coupled hydrology

Application study support for the next round of NO_X-SO_X welfare standard setting process and critical load and key TMDL assessments in collaboration with NCEA and OAQPS

Atmospheric deposition studies and scenarios for Ecosystem Services Research Program national nitrogen, wetlands and place-based assessments for future conditions that include land use and climate change.

Integrated studies with atmospheric and ecosystem models assessing the impact of climate change on critical loads and ecosystem health.

References

Appel, K.W., Roselle, S.J., Gilliam, R.C. and Pleim, J.E., 2009. Sensitivity of the Community Multiscale Air Quality (CMAQ) Model v4.7 results for the eastern United States to MM5 and WRF meteorological drivers. Geoscience Model Development Discussion, 2:1081-1114.

Baron, Jill S.; Rueth, Heather M.; Wolfe, Alezander M.; Nydick, Koren R.; Allstott, Eric J.; Minear, J. Toby, and Moraska, Brenda, 2000. Ecosystem Responses to Nitrogen Deposition in the Colorado Front Range, *Ecosystems*, 3, 352-368.

Bossi, R., Skov, H., Vorkamp, K., Christensen, J., Rastorgi, S.C., Egeløv, A., Petersen, D., 2008 Atmospheric concentrations of organochlorine pesticides, polybrominated diphenyl ethers and polychloronaphthalenes in Nuuk, South-West Greenland. *Atmospheric Environment* 42, 7293-7303.

Bowman, William D., Julia R. Gartner, Keri Holland, and Magdalena Wiedermann. 2006. Nitrogen Critical Loads for Alpine Vegetation and Terrestrial Ecosystem Response: Are We There Yet? *Ecological Applications*, 16, 1183-1192.

Clean Air Act Advisory Committee, 1005. Air Quality Management Group Report to the Clean Air Act Advisory Committee, 190 pp, http://www.epa.gov/air/caaac/aqm/report1-17-05.pdf.

DeHays, D.H., Schaberg, P.G., Hawley, G.J., Strimbeci, G.R., 1999. Acid rain impacts on calcium nutrition and forest health, *Bioscience*, 49, pp 789-800.

Dennis, R.L., Mathur, R., Pleim, J.E. Walker, J.T., in press. Fate of ammonia emissions at the local to regional scale as simulated by the Community Multiscale Air Quality Model , *Atmospheric Pollution Research*, in press.

Dennis, R., Haeuber, R., Blett, T., Cosby, J., Driscoll, C., Sickles, J. and Johnston, J., 2007. Sulfur and nitrogen deposition on ecosystems in the United States. *Environmental Manager*, December 2007: 12-17.

Dennis, R.L., 1997. Using the Regional Acid Deposition Model to Determine the Nitrogen Deposition Airshed of the Chesapeake Bay Watershed, in Baker, J.E. editor, **Atmospheric Deposition to the Great Lakes and Coastal Waters**, Society of Environmental Toxicology and Chemistry, Pennsacola, FL, pp 393-413.

Driscoll, Charles T., Gregory B. Lawrence, Arthur J. Bulger, Thomas J. Butler, Christopher S. Cronan, Christopher Eagar, Kathleen F. Lambert, Gene E. Likens, John L. Stoddard, and Kathleen C. Weathers. 2001. Acidic Deposition in the Northeastern United States: Sources and Inputs, Ecosystem Effects, and Management Strategies. *BioScience*, 51, 180-198.

Driscoll, Charles T., David Whitall, John Aber, Elizabeth Boyer, Mark Castro, Christopher Cronan, Christine L. Goodale, Peter Groffman, Charles Hopkinson, Kathleen Lambert, Gregory

Lawrence, and Scott Ollinger. 2003. Nitrogen Pollution in the Northeastern United States: Sources, Effects, and Management Options. *BioScience*, 53, 357-374.

Driscoll, CharlesT., Young-Ji Han, Celia Y. Chen, David C. Evers, Kathleen F. Lambert, Thomas M. Holsen, Neil C. Kamman and Ronald K. Munson. 2007. Mercury contamination in forest and freshwater ecosystems in the Northeastern United States. *BioScience,* 57, 17-28.

Fenn, Mark E., Mark A. Poth, John D. Aber, Jill S. Baron, Bernard T. Bormann, Dale W. Johnson, A. Dennis Lemly, Steven G. McNulty, Douglas F. Ryan, and Robert Stottlemyer. 1998. Nitrogen Excess in North American Ecosystems: Predisposing Factors, Ecosystem Responses, and Management Strategies. *Ecological Applications,* 8, 706-733.

Fenn, Mark E., Jill S. Baron, Edith B. Allen, Heather M. Rueth, Koren R. Nydick, Linda Geiser, William D. Bowman, James O. Sickman, Thomas Meixner, Dale W. Johnson, and Peter Neitlich. 2003. Ecological Effects of Nitrogen Deposition in the Western United States. *BioScience,* 53, 404-420.

Galloway, J.N., Cowling, E.B., 2002. Reactive Nitrogen and the World: 200 years of change, *Ambio*, vol. 31 no. 2, pp64-71.

Galloway, J.N., Aber, J.D., Erisman, J.W., Seitzinger, S.P., Howarth, R.W., Cowling, E.B., Cosby, B.J., 2003. The nitrogen cascade, *Bioscience*, 53, pp 341-356.

Geiser, Linda H., Jovan, Sarah E., Glavich, Doug A., Porter, Matthew K., 2010. Lichen-based critical loads for atmospheric nitrogen deposition in Western Oregon and Washington Forests, USA, *Environmental Pollution*, 158, 2412-2421.

Golden, H.E., Knightes, C.D., Cooter, E.J., Dennis, R.L., Gilliam, R.C., in press. Linking air quality and watershed models for environmental assessments: Analysis of the effects of model-specific precipitation estimates on calculated water flux, *Environmental Modeling & Software* (2010), doi:10.1016/j.envsoft.2010.04.015.

Jeremiason, J.D., Engstron, D.R., Swain, E.B., Nater, E.A., Johnson, B.M., Almendinger, J.E., Monson, B.A., Kolka, R.K., 2006. Sulfate addition increases methylmercury production in an experimental wetland, *Environmental Science and Technology*, 40, pp 3800-3806.

Katata, G., Nagai, H., Wrzesinsky, T., Klemm, O., Eugster, W., and Burkard, R, 2008. Development of a Land surface Modeling Including Cloud Water Deposition on Vegetation, *Journal of Applied Meteorology and Climatology*, 47, 2129-2146.

Linker, L.C, G.W. Shenk, R.L. Dennis, J.S. Sweeney, 2000. Cross-Media Models of the Chesapeake Bay Watershed and Airshed, *Water Quality and Ecosystem Modeling*, **1**, 91-122.

Lovett, G.M. and T.H. Tear, 2008. Threats from Above: Air Pollution Impacts on Ecosystems and Biological diversity in the Eastern United States, The Nature Conservancy and the Cary Institute of Ecosystem Studies, Bethesda, MD and Millbrook, NY. 28 pp.

(www.ecostudies.org/reprints/Threats_from_Above.pdf). (Available online at www.nature.org/wherewework/northamerica/states/maryland/)

National Exposure Research Laboratory, 2009. A conceptual Framework for U.S. EPA's National Exposure Research Laboratory, Report No. EPA/600/R-09/003, Office of Research and Development, Washington, DC 20460, 42 pp.

National Research Council (NRC), 2004. Air Quality Management in the United States. Report by the Committee on Air Quality Management in the United States, The National Academy Press, Washington, D.C.

Nemitz, E., Milford, C., Sutton, M.A., 2001. A two-layer canopy compensation point model for describing bi-directional biosphere-atmosphere exchange of ammonia. *Q. J. R. Meterol. Soc.*, *127*, 815-833

Paerl, H.W., Dennis, R.L., Whitall, D.R., 2002. Atmospheric Deposition of Nitrogen: Implications for Nutrient Over-enrichment of Coastal Waters, *Estuaries*, 25, No. 4B, 677-693.

Pleim, J.E. and R.C. Gilliam, 2009. An Indirect Data Assimilation Scheme for Deep Soil Temperature in the Pleim-Xiu Land Surface Model. *Journal of Applied Meteorology and Climatology*, 48(7):1362-1376.

Pleim, J.E., 2007a. A combined local and nonlocal closure model for the atmospheric boundary layer. Part I: model description and testing, *Journal of Applied Meteorology and Climatology*, 46, 1383-1395.

Pleim, J.E., 2007b. A combined local and nonlocal closure model for the atmospheric boundary layer, Part II: application and evaluation in a mesoscale meteorological model, *Journal of Applied Meteorology and Climatology*, 46: 1396-1409.

Rimmer, C.C., Mcfarland, K.P., Evers, D.C., Miller, E.K., Aubry, Y., Busby, D., Taylor, R.J., 2005. Mercury concentrations in Bicknell's thrush and other insectivorous passerines in Montane forsts of northeastern North America, *Ecotoxicology*, 14, pp 223-240.

Seigneur, C. and Dennis, R.L., in press. Chapter 8: Atmospheric Modeling, in *Technical Challenges of Risk- and Results-Based Multipollutant Air Quality management*, Hidy, G., J.R. Brook, K.L. Demerjian, L.T. Molina, W.T. Pennell, and R. Scheffe (editors), Springer, Dordrecht, 2010.

Sullivan, T.J., Cosby, B.J., Webb, J.R., Dennis, R.L., Bulger, A.J. and Deviney, F.A., 2008. Streamwater acid-base chemistry and critical loads of atmospheric sulfur deposition in Shenandoah National Park, Virginia. *Environmental Monitoring and Assessment*, 137(1-3): 85-99.

Sutton, M.A., J.K. Burkhardt, D. Guerin, E. Nemitz, D. Fowler, 1998. Development of resistance models to describe measurements of bi-directional ammonia surface-atmosphere exchange. *Atmos. Environ.* 32, 473-480

Valigura, R.A., Alexander, R.B., Castro, M.S., Meyers, T.P., Paerl, H.W., Stacey, P.E., Turner, R.E. (editors), 2001. **Nitrogen Loading in Coastal Water Bodies: An Atmospheric Perspective**, Coastal and Estuarine Studies No. 57, American Geophysical Union, Washington DC, 254 pp.

Vitousek, M., J.D. Aber, R.W. Howarth, G.E. Likens, P.A. Matson, D.W. Schindler, W.H. Schlesinger, D.G. Tilman, 1997. Technical Report: Human Alteration of the Global Nitrogen Cycle: Sources and consequences, *Ecological Applications*, vol. 7, no. 3, pp 737-750.

Walker, J.T., Robarge, W.P., Wu, Y., Meyers, T., 2006. Measurement of bi-directional ammonia fluxes over soybean using the modified Bowen-ratio technique, *Agricultural and Forest Meteorology*, 138, 54-68.

Weathers, K.C., Lovett, G.M., Likens, G.E., 1995. Cloud deposition to a spruce forest edge, Atmospheric Environment, 29, 665-672.

Weathers, K.C., Simkin, S.M., Lovett, G.M., Lindberg, S.E., 2006. Empirical modeling of atmospheric deposition in mountainous landscapes, Ecological Applications, 16, 1590-1607.