Appendix C: Chemical Transport Model Results for Sulfate Source Attribution Studies in the Northeast U.S.

Model description and performance evaluation

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ABSTRACT

Regional Scale Air Quality Modeling is being conducted by NESCAUM with two primary modeling systems based on the CMAQ and REMSAD models. Modeling efforts are being conducted on a 12-km Eastern U.S. domain consistent with the RPO projection national domain. Modeling for the 2002 base year and two future years (2009, 2018) has been conducted. CMAQ provides one-atmosphere results for multiple pollutants while the REMSAD model is used primarily for attribution of sulfate species in the Eastern US via the species-tagging scheme included in Version 7.10 and newer versions of the model. Performance evaluations for both models are provided and will be refined for CMAQ once final data inputs have been developed. Current model output suggests reasonable performance for the species of interest for each platform. Only minor changes are anticipated for scenarios conducted with the final inventory version and revised model code.

Appendix C: Chemical Transport Model Results for Sulfate Source Attribution Studies in the Northeast U.S.

C.1. Introduction

Air quality models have been extensively used as important tools within academia, federal and state governments, and other regulatory, policy, and environmental research communities to understand the underlying causes and formation mechanisms of air pollution on the local, regional and global scale. To assist states in developing effective solutions to air pollution issues, NESCAUM has established in-house air quality modeling capabilities that include emission processing, meteorological input analysis, and chemical transport modeling. Preliminary work has been conducted to develop a number of modeling platforms for a variety of applications (e.g. annual runs, episodic simulations, source tagging, etc.) and to evaluate model performance. These efforts form the foundation upon which future modeling studies will be built ranging from SIP modeling for Regional Haze, Ozone, and fine particles to studies of mercury deposition and potential air quality impacts of climate change.

This appendix introduces air quality modeling platforms established by NESCAUM for Regional Haze and fine particle pollution, evaluates model performance with respect to spatial and temporal variations of PM species and their precursors, and summarizes the current status and future improvements of each platform's development.

C.2. Platforms

Currently two regional-scale air quality models have been evaluated and used by NESCAUM to perform air quality simulations. These are the Community Multi-scale Air Quality modeling system (CMAQ; Byun and Ching, 1999) and the Regional Modeling System for Aerosols and Deposition (REMSAD; SAI, 2002). CMAQ was developed by USEPA, while REMSAD was developed by ICF Consulting/Systems Applications International (ICF/SAI) with USEPA support. CMAQ has undergone extensive community development and peer review (Amar et al. 2005) and has been successfully used in a number of regional air quality studies (Bell and Ellis, 2003;Hogrefe et al., 2004; Jimenez and Baldasano, 2004; Mao and Talbot, 2003; Mebust et al. 2003). REMSAD has also has been peer reviewed (Seigneur et al., 1999) and used by EPA for regulatory applications (http://www.epa.gov/otaq/regs/hd2007/frm/r00028.pdf and http://www.epa.gov/clearskies/air_quality_tech.html) to study ambient concentrations and deposition of sulfate and other PM species.

C.2.1. CMAQ

The CMAQ modeling system is a three-dimensional Eulerian model that incorporates output fields from emissions and meteorological modeling systems and several other data sources through special interface processors into the CMAQ Chemical Transport Model (CCTM). The CCTM then performs chemical transport modeling for multiple pollutants on multiple scales. With this structure, CMAQ retains the flexibility to substitute other emissions processing systems and meteorological models. CMAQ is

designed to provide an air quality modeling system with a "one atmosphere" capability containing state-of-science parameterizations of atmospheric processes affecting transport, transformation, and deposition of such pollutants as ozone, particulate matter, airborne toxics, and acidic and nutrient pollutant species (Byun and Ching, 1999).

MANE-VU has adopted the Inter-RPO domain description for its modeling runs.[1] This 36-km domain covers the continental United States, southern Canada and northern Mexico. The dimensions of this domain are 145 and 102 cells in the east-west and north-south directions, respectively. A 12-km inner domain was selected to better characterize air quality in MANE-VU and surrounding RPO regions. This domain covers the Northeast region including northeastern, central and southeastern US as well as Southeastern Canada. It extends from 66^{o}W~94^{o}W in longitude and 29^{o}N~50^{o}N in latitude with 172X172 grid cells (Figure C-1).

NY DEC has completed annual 2002 CMAQ modeling on the 36km domain to provide dynamic boundary conditions for all simulations performed on the 12km domain. Three-hourly boundary conditions for the outer domain were derived from an annual model run performed by researchers at Harvard University using the GEOS-CHEM global chemistry transport model (Park et al., 2004). Model resolution was species dependent at either 4° latitude by 5° longitude or 2° by 2.5°.

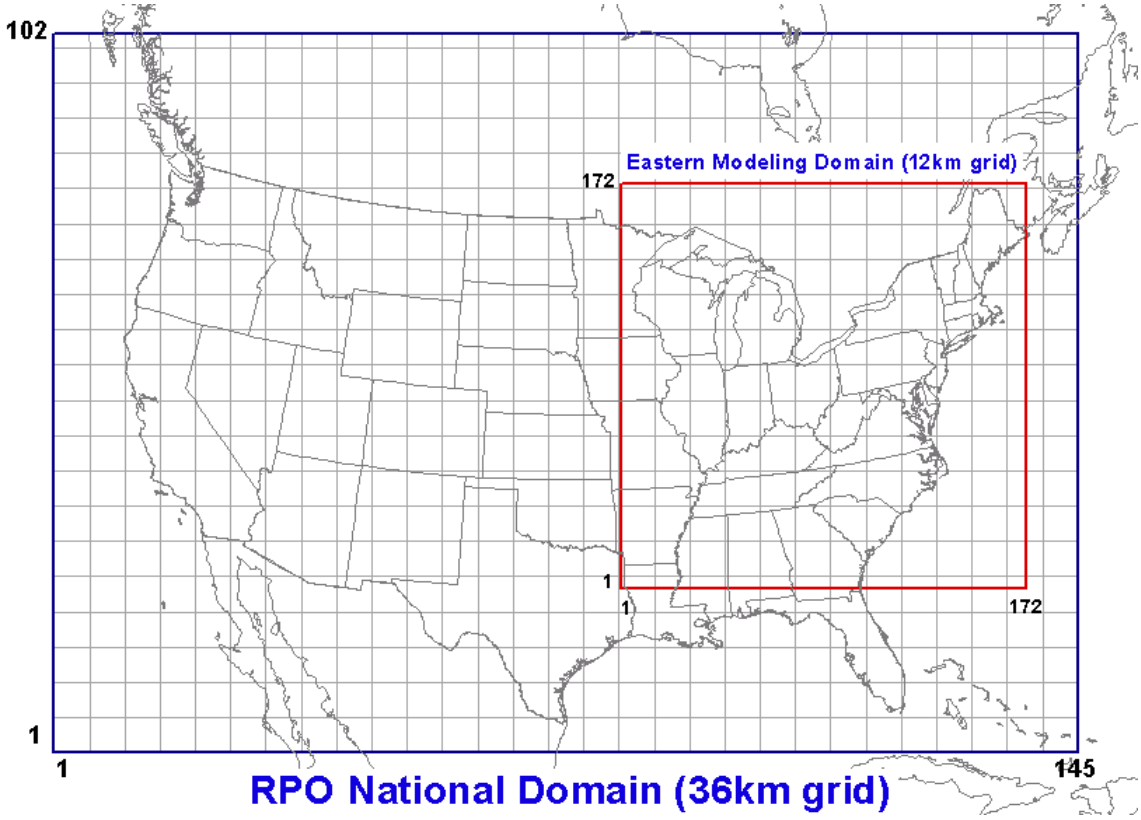
To date, MANE-VU SIP modeling on both 36km and 12km domains used CMAQ V4.4 with IOAPI V2.2 and NETCDF V3.5 libraries. The CMAQ model is configured with the Carbon Bond IV mechanism (Gery et al., 1989) using the EBI solver for gas phase chemistry rather than the SAPRC-99 mechanism due to better computing efficiency with no significant model performance differences for Ozone and PM as compared to observations.

Meteorological inputs for CMAQ are derived from the Fifth-Generation Pennsylvania State University/National Center for Atmospheric Research (NCAR) Mesoscale Model (MM5)[2] system meteorological fields. A modified Blackadar boundary layer scheme is used as well as physics options including explicit representations of cloud physics with simple ice microphysics (no mixed-phase processes) and the Kain-Fritsch cumulus parameterization. As shown in Figure C-2, out of the 29 MM5 vertical layers, CMAQ has 22 vertical layers from the ground surface up to ~200hPa, with 10 layers below 850hPa (including 1 layer below 10m) to resolve boundary layer processes, 6 layers in between 850hPa and 500hPa, and 3 layers in between 500hPa and 300hPa. The domain has a finer vertical resolution within the troposphere so that it can capture complex atmospheric circulations between the east coast of the US and the northern Atlantic Ocean in the boundary layer, the free and upper troposphere, and potentially cross-tropopause transport.

[1] The modeling system for 2002 annual simulation is applied with a Lambert Conformal Conic projection with parallels at 33N and 45N. A spherical earth radius of 6370km is used for all elements of the system (MM5/SMOKE/CMAQ).

[2] http://www.mmm.ucar.edu/mm5/

Figure C-1. Modeling domains used in MANE-VU air quality modeling studies with CMAQ. Outer (blue) domain grid is 36 km and inner (red) domain is 12 km grid. The gridlines are shown at 180 km intervals (5 x 5 36km cells or 15 x 15 12km cells



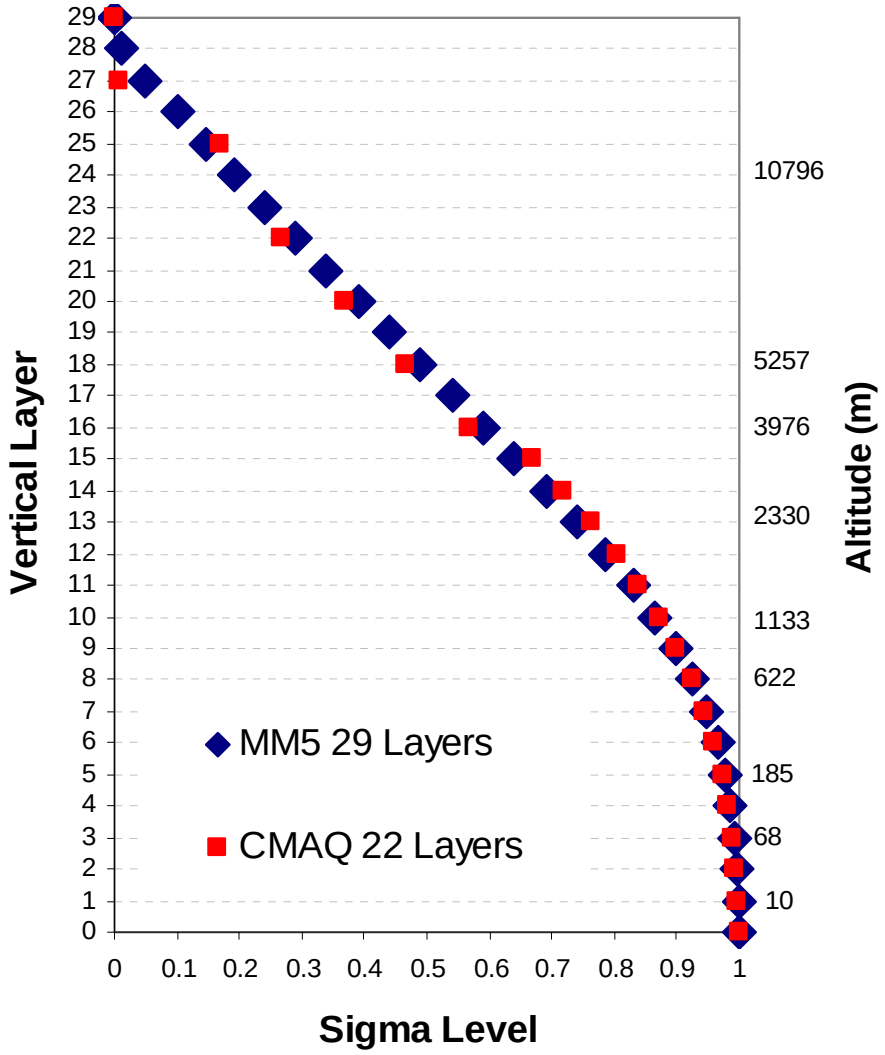
Five modeling centers are working collectively to maximize efficiency of computing resources in MANE-VU for SIP modeling. These centers include NY DEC, NJ DEP/Rutgers, VA DEQ, UMD, and NESCAUM. Annual CMAQ modeling on 12km domain is divided into 5 periods. UMD is responsible for the period from January 1 to February 28; NJ DEP and Rutgers are responsible for the period from March 1 to May 14; NY DEC is responsible for the period from May 15 to September 30; VA DEQ is responsible for the period from October 1 to October 31; and NESCAUM is responsible for the period from November 1 to December 31. Each period uses a 15 day spin up run to minimize/eliminate the impact of the default initial concentration fields. Each group performs CMAQ simulation on its period for a series of scenarios including 2002 Base Case, 2009 Base Case, 2018 Base Case, 2009 Control Case, and 2018 Control Case. All scenarios adopt the same meteorological field (2002) and boundary conditions, varying only emission inputs. To ensure consistency, a benchmark test was conducted by each modeling group.

C.2.1.1. Meteorology

All meteorological fields have been simulated using the (MM5) system. Originally developed by Penn State and NCAR, MM5 is a model with limited-area primitive equations of momentum, thermodynamics, and moisture with the option of hydrostatic and non-hydrostatic physics. It is designed to simulate mesoscale

atmospheric circulation. Domains are uniform rectangular grids representing three-dimensional regions of the atmosphere. The horizontal coordinated system is equally spaced geographically and uses the Arakawa-B gridding scheme. The resolution can be as high as 1km. Sigma (σ) is a terrain-following vertical coordinate that is a function of pressure at the point (for hydrostatic) or reference state pressure (non-hydrostatic) (P), the surface pressure (P_{s0}), and the pressure at the top (P_{top}) of the model. $\sigma = (p-p_{top}) / (p_{s0}-p_{top})$

Figure C-2. Vertical Structure of Air Quality Modeling Domains



Professor Dalin Zhang's group from University of Maryland (UMD) was tasked to provide the 2002 annual meteorological field for air quality modeling. The UMD MM5 model runs are made on two nested domains: the 36km resolution National US Continental domain (Domain 1) and the 12km resolution Eastern US domain (Domain 2). The inner (12km) domain uses a finer resolution terrain data. Initially, a set of test runs for the period of August 6 to 16, 2002 was conducted. UMD ran the non-hydrostatic MM5 v3.5.3 with 3 PBL schemes (a) modified Blackadar [BL], (b) the Pleim-Xiu scheme with the soil module [P-X], and (c) modified Blackadar with soil module [SSIB]. The simulated meteorological fields were compared to the measurements from

Techniques Development Laboratory of National Weather Service (TDL NWS) and Clean Air Status and Trends Network (CASTNET). The TDL data are reflective of urban/suburban settings, while the CASTNET sites are more representative of rural areas. There are 48 CASTNET sites and about 800 TDL sites within Domain 2 (as shown in Figure C-3). Overall, the BL scheme shows a better correspondence to the measured data than the other two schemes, although it poorly captures the diurnal pattern of humidity. While the P-X scheme shows a better correspondence with the observed diurnal pattern for humidity, it fails to perform well for wind speed and temperature (Hao et al., 2004). The model utilizes a terrain-following sigma coordinate with 29 layers with the first level at 10 m and a radiative upper-boundary condition at 50hPa. Based on test run results, the boundary layer processes are determined using the Blackadar high-resolution planetary boundary layer parameterization. Physics options also included explicit representations of cloud physics with simple ice microphysics (no mixed-phase processes) and the Kain-Fritsch cumulus parameterization. The model was initialized with the analyses of the National Center for Environmental Prediction (Eta Model). TDL data are used for MM5 nudging. A modeled wind field map (Figure C-4) shows typical prevailing mesoscale flows from the midwest US to the east coast.

Figure C-3. Observation Network sites within 12km resolution domain

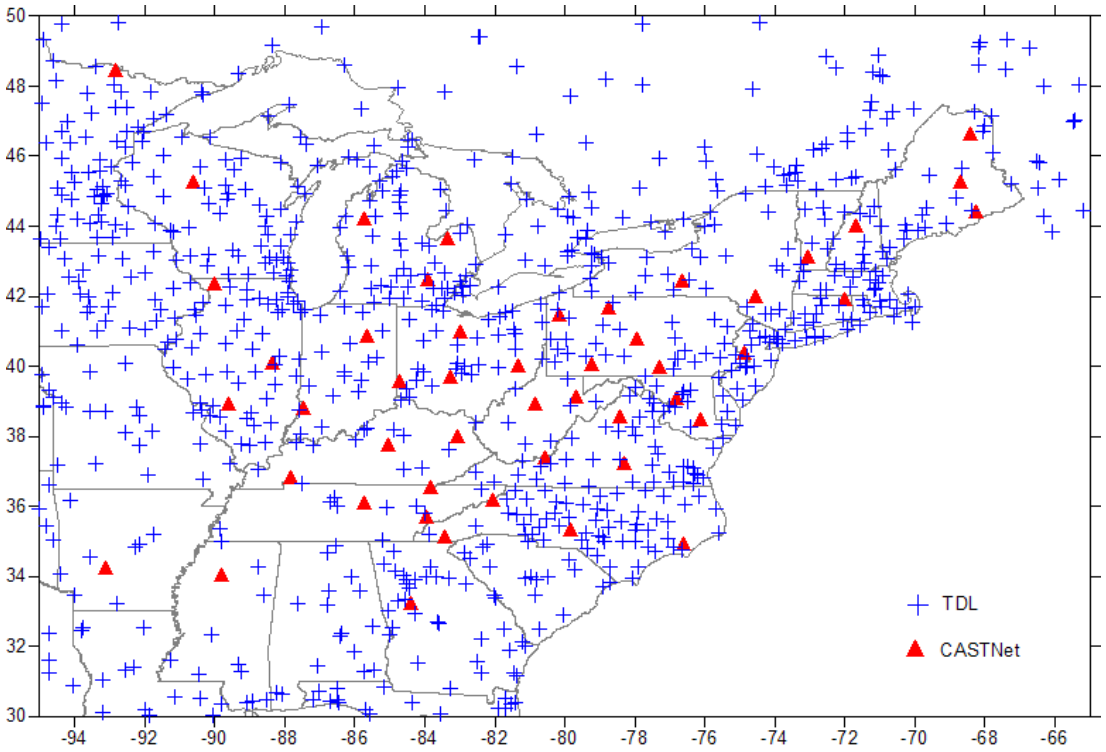
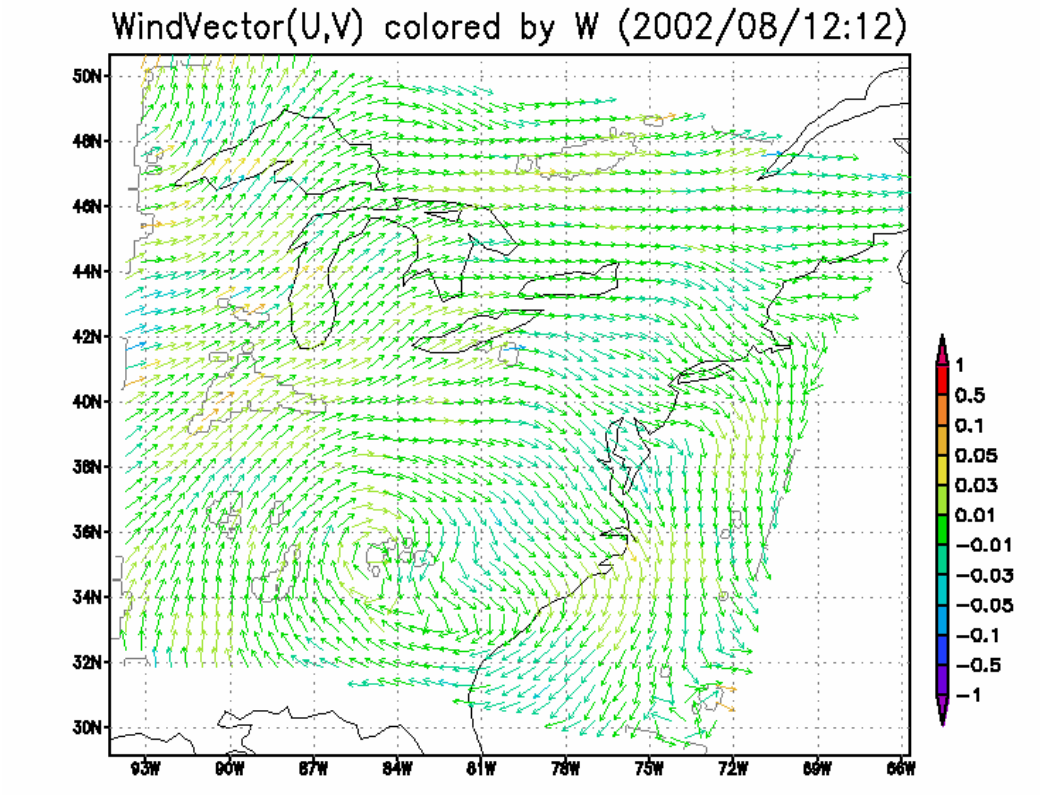


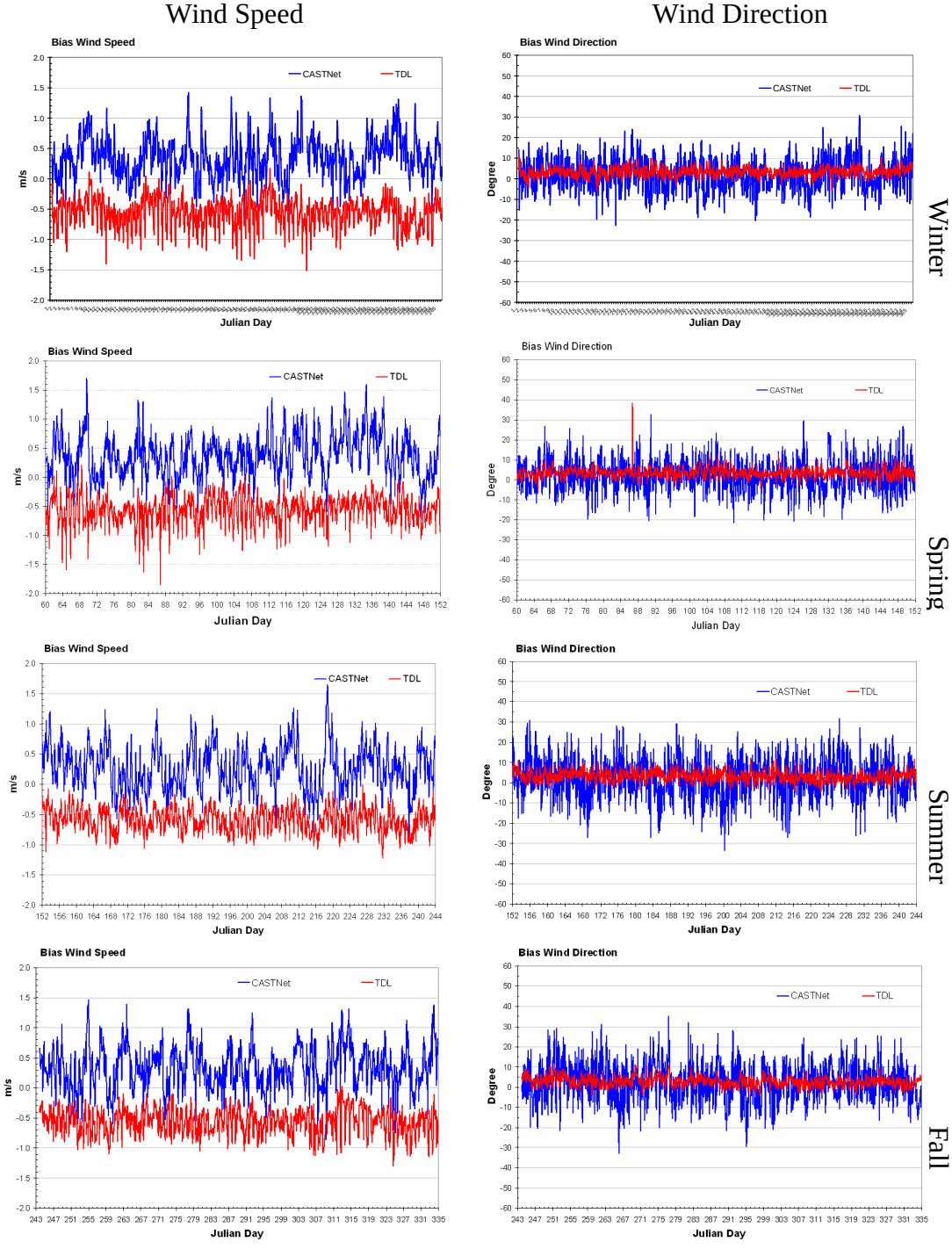
Figure C-4. MM5 modeled wind field map at 12:00 UTC on August 8, 2002



The MM5 generated 2002 annual 12km resolution meteorological field has been evaluated by NESCAUM using ENVIRON's METSTAT program. Model results of surface wind speed, wind direction, temperature, and humidity are paired with measurements from CASTNET and TDL networks by hour and by location and then statistically compared.

Figure C-5 presents domain-wide average hourly bias of wind speed (left panel) and wind direction (right panel) between MM5 results and two sets of measurement for every season in 2002 (a-Winter including Jan. Feb. and Dec.; b-Spring including Mar. Apr. and May; c-Summer including Jun. Jul. and Aug; d-Fall including Sep. Oct. and Nov.). It shows that MM5 capably predicts wind speed with reasonably small bias and equal consistency. Within the domain, MM5 tends to overestimate wind speed (hourly bias up to 1.7m/s) at CASTNET sites, and underestimate wind speed (hourly bias up to -1.85m/s) at TDL sites. Seasonal mean bias of MM5 wind speed to CASTNET wind speed is ~0.3 to 0.4m/s, while seasonal mean bias of MM5 wind speed to TDL wind speed is about ~-0.5 to -0.6m/s. No significant seasonal variation on this wind speed bias is observed. MM5 prediction of wind direction shows a larger variation with CASTNET wind direction (hourly bias from ~-30 degree to ~30 degree) than with TDL wind direction (hourly bias from ~-5 degree to ~10 degree). However, seasonal mean bias of MM5 wind direction to CASTNET wind direction (~2 degree) is smaller than seasonal mean bias of MM5 wind direction to TDL wind direction (~3 degree) because the large variation of positive and negative bias offset each other.

Figure C-5. 2002 Seasonal Average Hourly Bias of Wind Speed and Direction

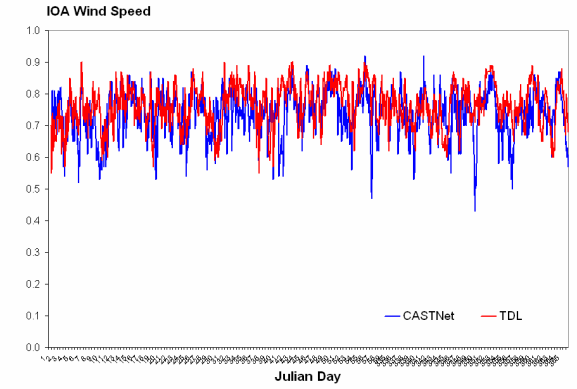


Index of Agreement (IOA) is a statistical measure of difference between prediction and measurement, calculated as a ratio of Root Mean Square Error to the sum of the difference between prediction and mean observation and difference between observation and mean observation. IOA varies from 0 to 1, with a value of 1 indicating the prefect agreement between model prediction and observation, and a value larger than 0.5 IOA indicating acceptable model performance. Domain wide average hourly IOA of

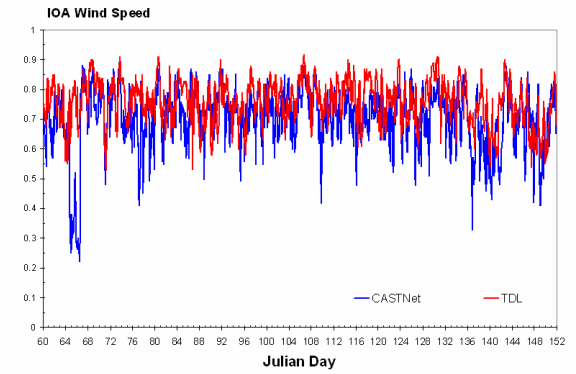
wind speed are presented in Figure C-6. MM5 predicted wind speed values are in good agreement (IOA from ~0.5 to ~0.9) to both CASTNET data and TDL data with similar IOA variation. Seasonal mean values of IOA are ~ 0.7. No particular season of the year stands out in terms of its agreement with measurement.

Figure C-6. 2002 Seasonal Hourly Average Index of Agreement for Wind Speed

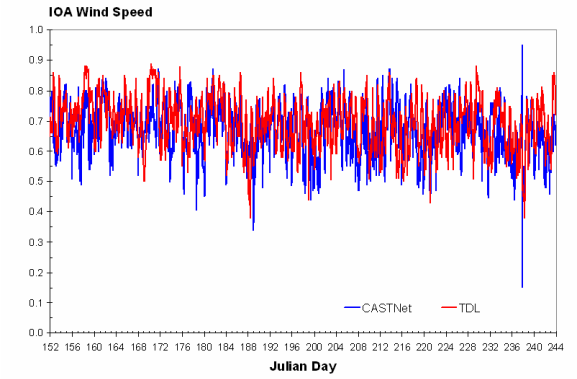
a) Winter



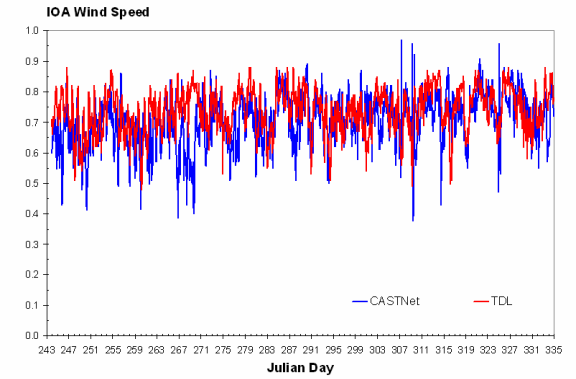
b) Spring



c) Summer



d) Fall



Quarterly correlation coefficients in Figure C-7 show good MM5 performance on hourly wind speed for each observation site. MM5 predictions exhibit similar spatial patterns of correlation with CASTNET (left panel) and TDL (right panel) measurements – stronger correlation in north than in south. Over the year, the model has stronger correlation in the 1st quarter (Jan. Feb. Mar., top 1st row), 2nd quarter (Apr. May Jun., 2nd row) and 4th quarter (Oct. Nov. and Dec., bottom row) than it does in the 3rd quarter (Jun. Jul. Aug., 3rd row), with an average of 0.1 correlation coefficient difference. Generally MM5 predictions and measurements have strongest correlation (0.8~0.9) within Midwest US, strong correlation (0.7~0.8) within Northeastern US and along coastline, and acceptable correlation (0.5~0.7) within Southern US and inland coast. MM5 predictions consistently show very similar spatial patterns and temporal variations for wind direction (as shown in Figure C-8) and wind speed. There is strong correlation (>0.7) between prediction and measurement for wind direction at most of sites.

Figure C-7. Quarterly correlation coefficient (r) of hourly wind speed between modeling and measurement for each observation site in 2002

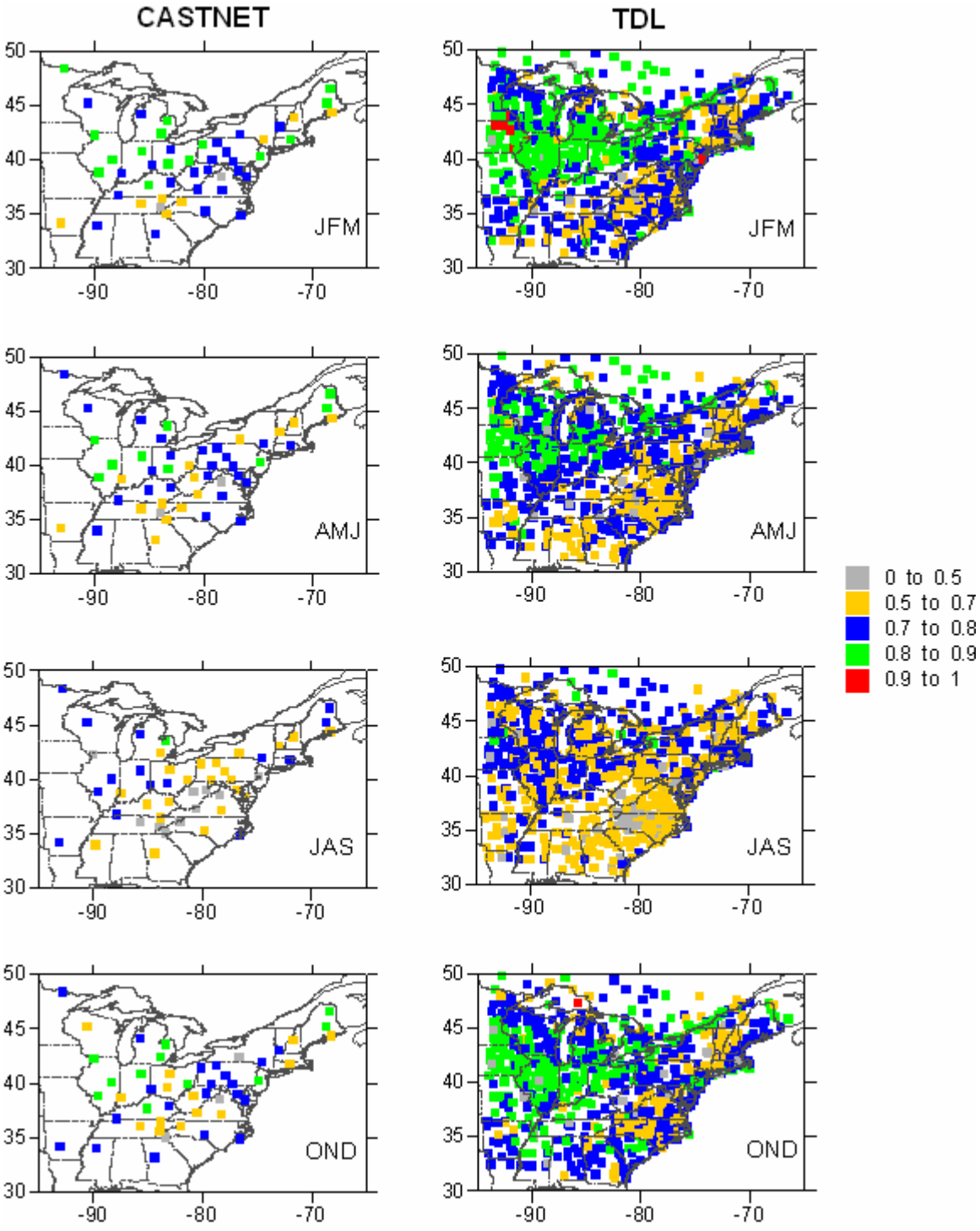


Figure C-8. Quarterly correlation coefficient (r) of hourly wind direction between modeling and measurement for each observation site in 2002

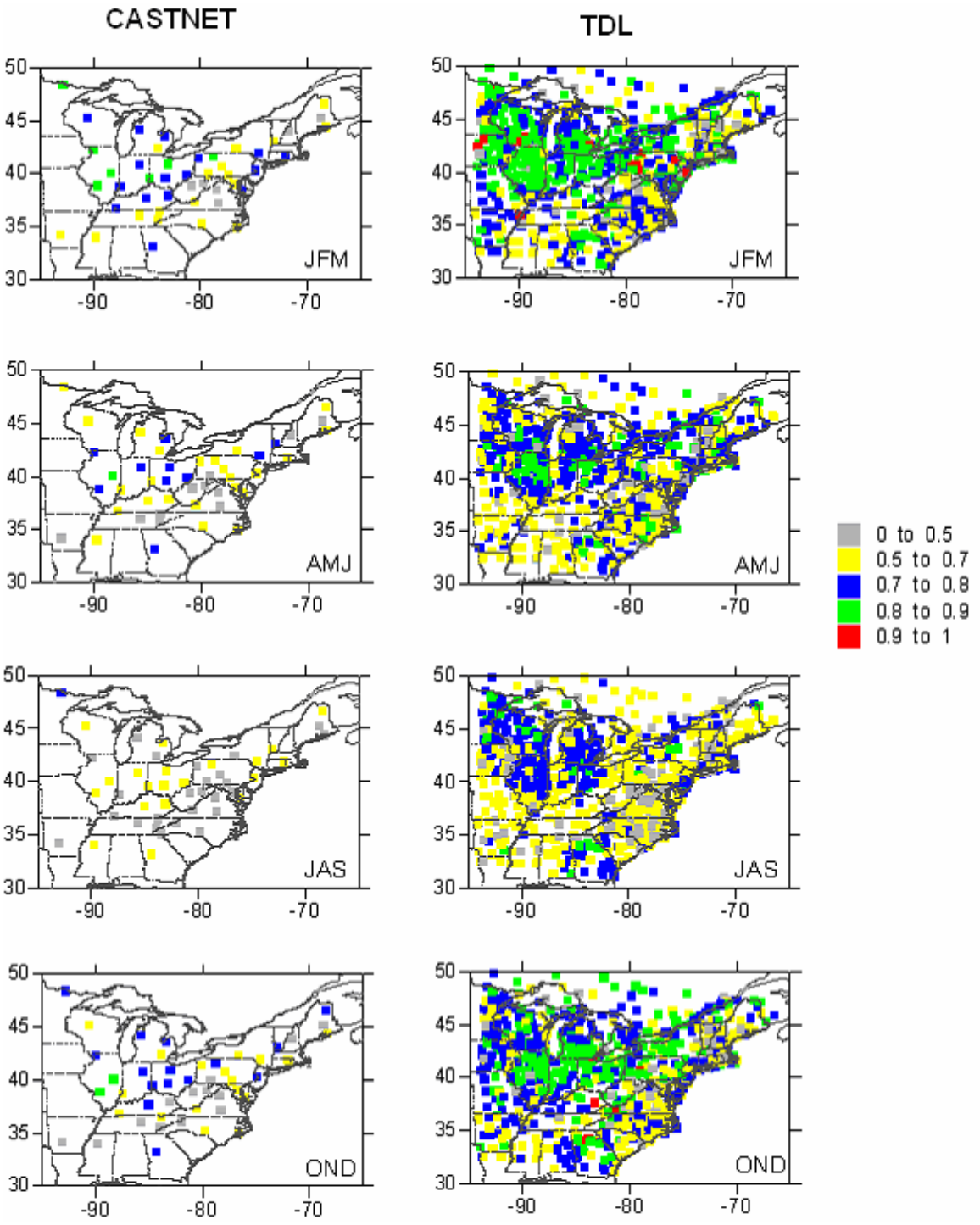


Figure C-9 presents domain wide average hourly bias of surface temperature between MM5 results and CASTNET and TDL for every season. MM5 tends to underestimate temperature at TDL sites throughout the year and at CASTNET sites for non-ozone season months. The seasonal mean temperature bias values are from ~-1K (Winter) to ~-0.3K (Summer) for TDL sites and ~-1K(Winter) to ~0.5K(Summer) for CASTNET sites. MM5 predictions show significantly larger variations of temperature bias at CASTNET sites (-4K~9K) than at TDL sites (-3K~1K). Domain wide average hourly IOA values of temperature are shown in Figure C-10. Model predicted temperatures have significantly better agreement with TDL data (average IOA as ~0.95) than with CASTNET data (average IOA as ~0.85), although both indicate accurate MM5 performance on temperature. Figure C-11 shows the spatial distribution of quarterly correlation coefficients between MM5 prediction and measurement of surface temperature. It reveals very strong correlation (>0.95) over most of the domain for TDL data, with strong correlation (>0.8) for the majority of CASTNET sites. No spatial patterns or quarterly variations are apparent. MM5 performs consistently well throughout the year and the domain.

The TDL network also provides humidity measurements. Comparison between MM5 prediction of hourly surface humidity and TDL measurement are presented in Figure C-12. MM5 captures the general trend of humidity change. It tends to underestimate humidity during the ozone season (seasonal mean bias as ~0.35g/kg), and overestimate it during the rest of year (seasonal mean bias range from ~0.17 to ~0.4), as shown in. Domain wide average hourly humidity bias shows a large diurnal variation, as much as 2g/kg. Domain wide average hourly IOA in Figure C-13 shows that MM5 predicted humidity values are in good agreement with TDL data (average IOA as ~0.9) throughout year. Spatial distribution of quarterly correlation coefficient in Figure C-14 shows a distinctive spatial pattern and temporal trend. MM5 results have stronger correlation to TDL data in the northern US than in the Southern US. Through the year, the strongest correlation between MM5 prediction and measurement occurs in the 4th Quarter (>0.95), followed by the 1st and 2nd Quarters, and finally, the 3rd Quarter, which shows the weakest correlation (0.5~0.9).

Based on this statistical comparison between model prediction and data from two networks for wind speed, wind direction, temperature, and humidity, MM5 performs well. An acceptable small bias, high index of agreement and strong correlation with CASTNET and TDL data are shown. Since MM5 uses TDL data for nudging, the model predictions are in better agreement with TDL data than with CASTNET data. MM5 performs better in Midwest and Northeast than Southeastern US.

Figure C-9. 2002 Seasonal Hourly Average Bias of Temperature

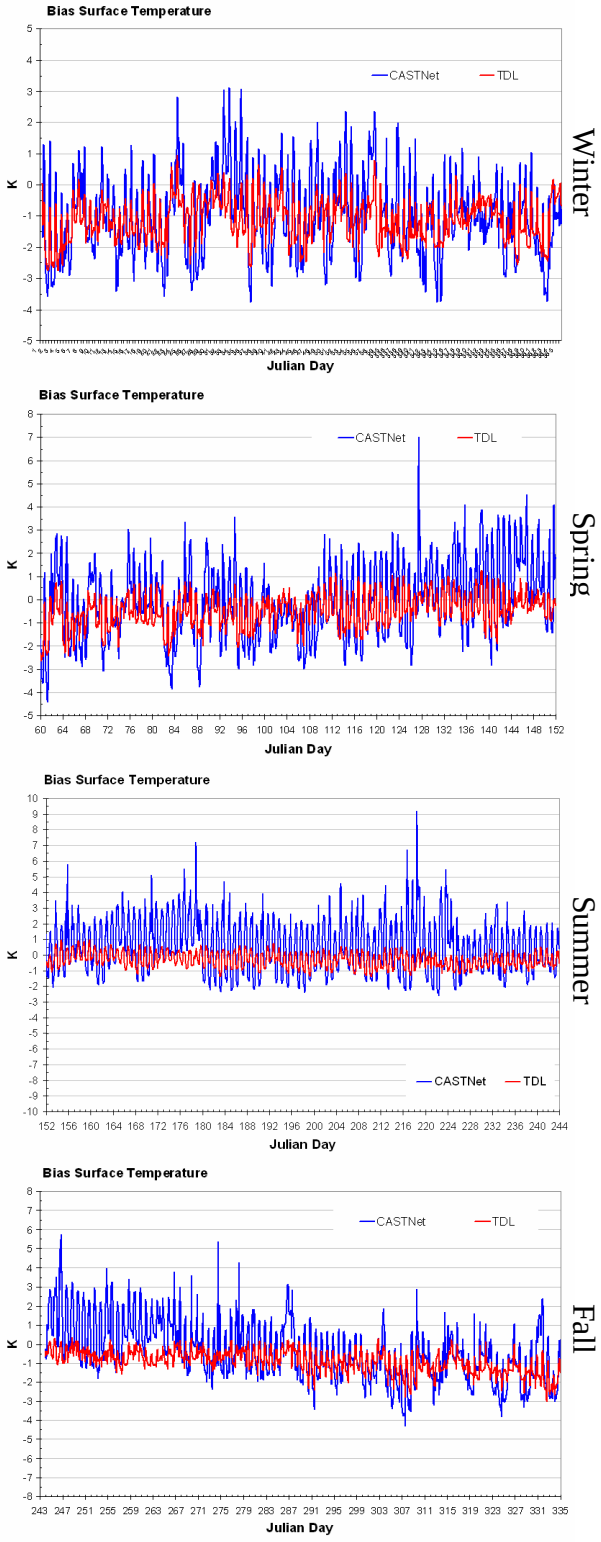


Figure C-10. 2002 Seasonal Hourly Average Index of Agreement for Temperature

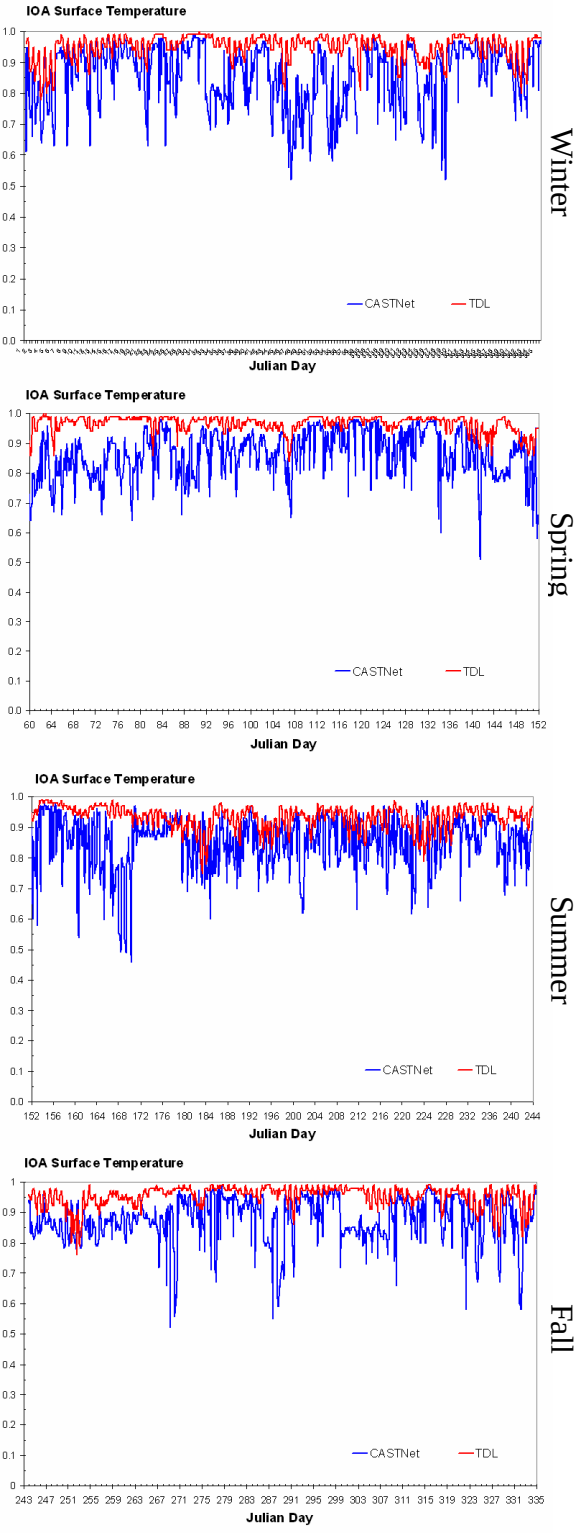


Figure C-11. Quarterly correlation coefficient (r) of hourly temperature between modeling and measurement for each observation site in 2002

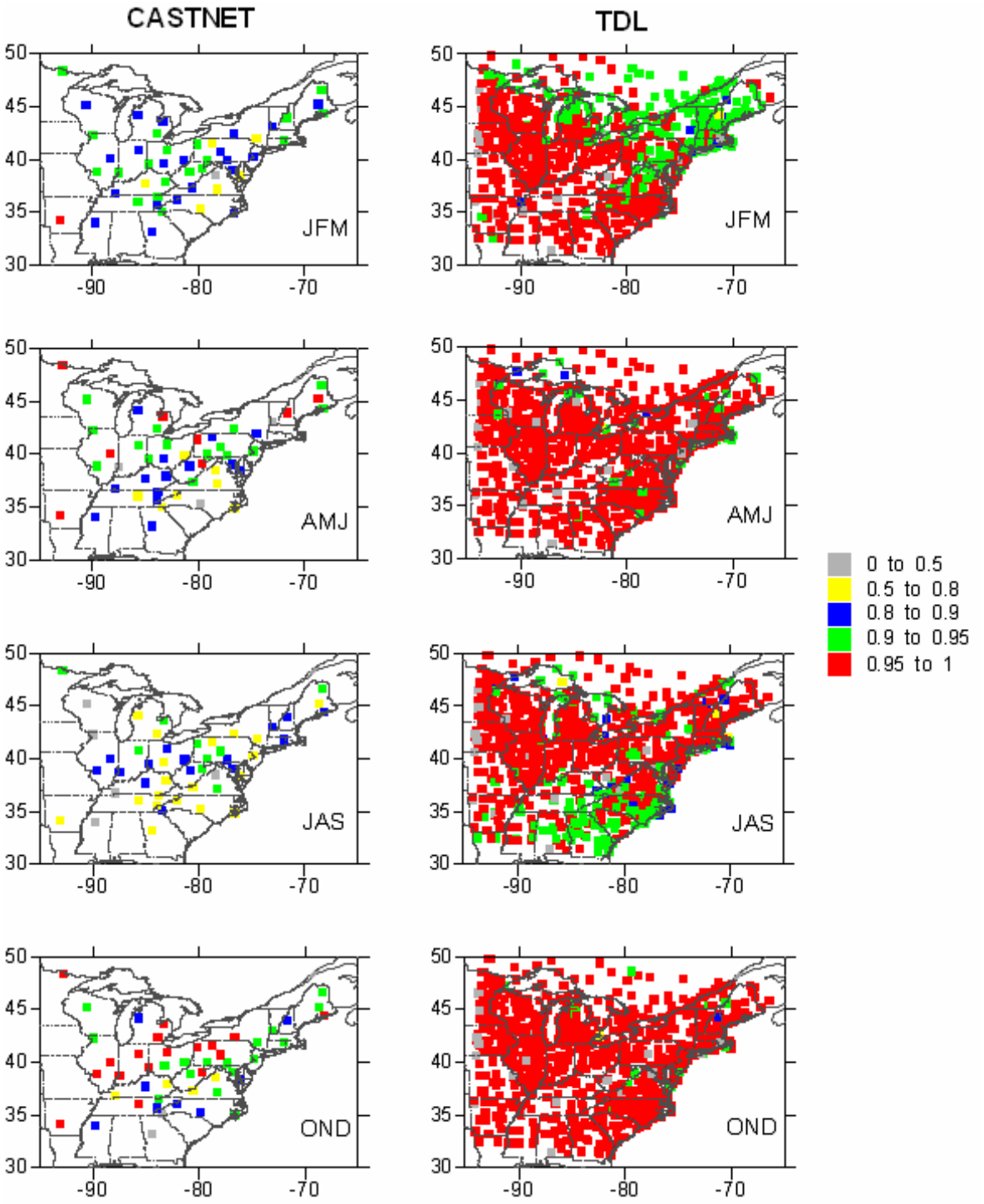


Figure C-12. 2002 Seasonal Average Hourly Bias of Humidity

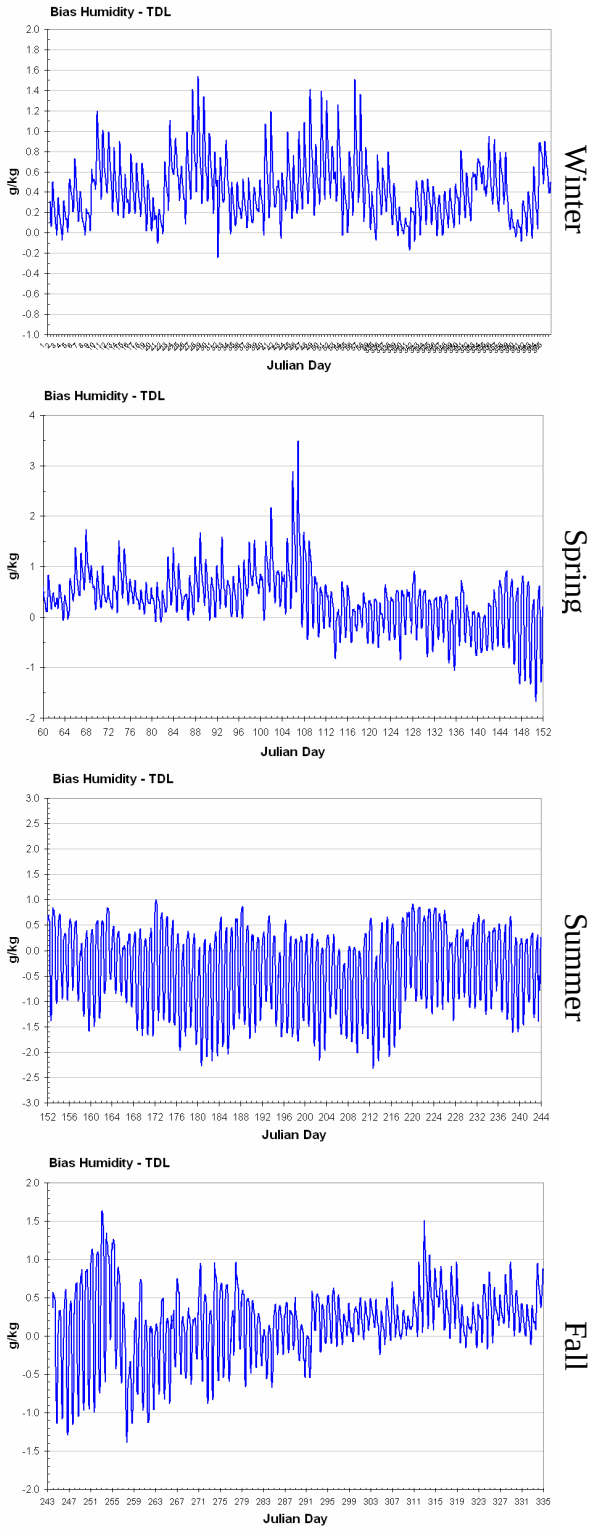


Figure C-13. 2002 Seasonal Hourly Average Index of Agreement for Humidity

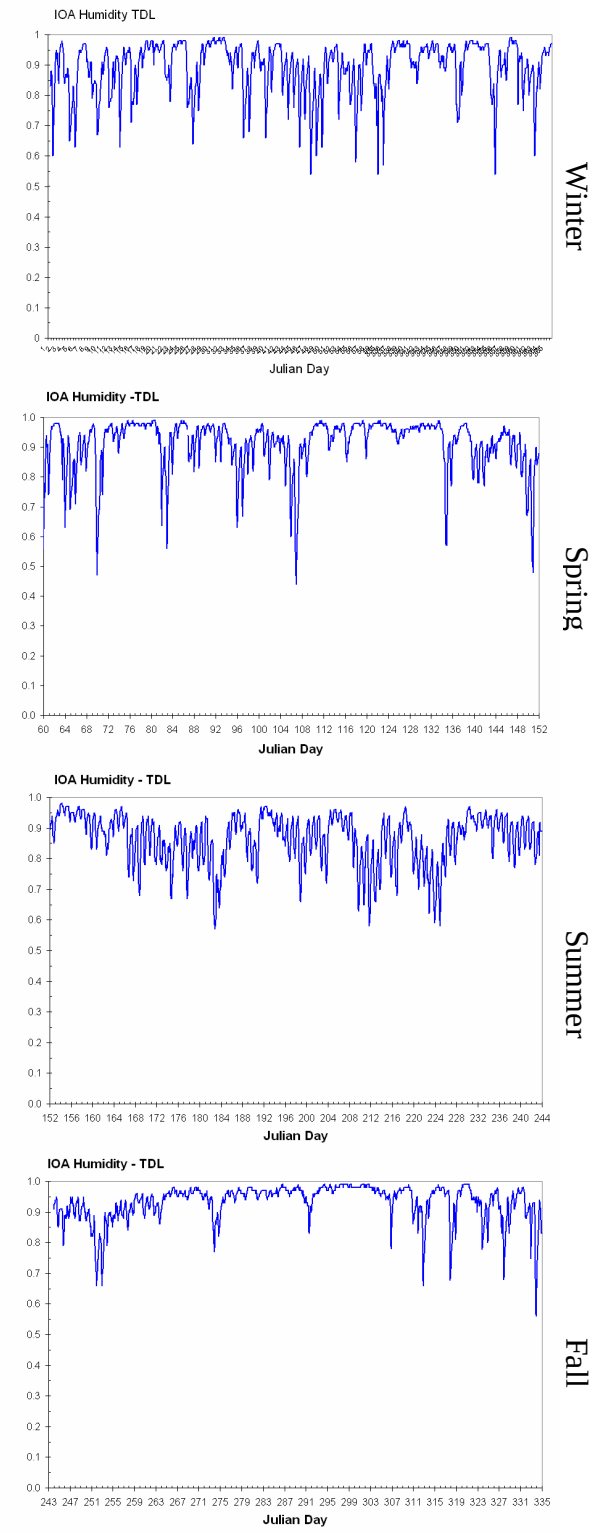
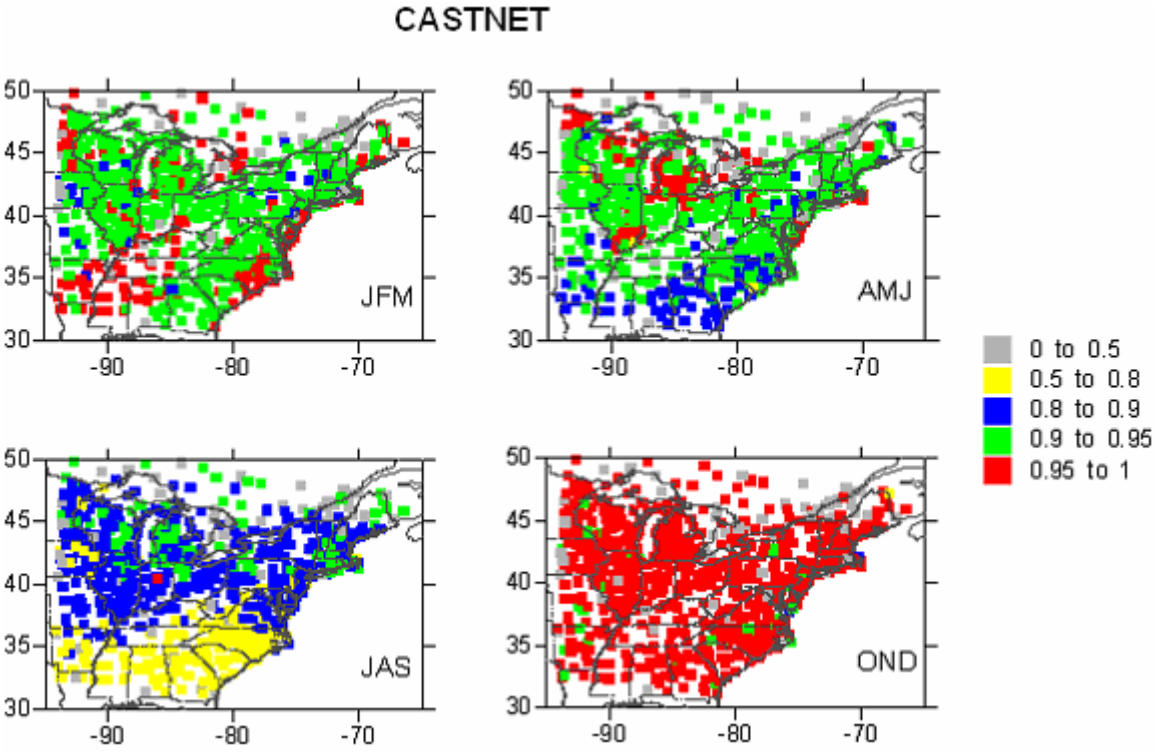


Figure C-14. Quarterly correlation coefficient (r) of hourly humidity between modeling and measurement for each observation site in 2002



C.2.1.2. CMAQ Emissions

Emission scenarios are simulated using the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System. SMOKE is primarily an emissions processing system designed to create gridded, speciated, hourly emissions for input into a variety of air quality models such as CMAQ, REMSAD, the Comprehensive Air quality Model with extensions (CAM_X) and the Urban Airshed Model (UAM). SMOKE supports area, biogenic, mobile (both onroad and nonroad), and point source emissions processing for criteria, particulate, and toxic pollutants. For biogenic emissions modeling, SMOKE uses the Biogenic Emission Inventory System, version 2.3 (BEIS2) and version 3.09 and 3.12 (BEIS3). SMOKE is also integrated with the on-road emissions model MOBILE6. The sparse matrix approach used throughout SMOKE permits rapid and flexible processing of emissions data. Flexible processing comes from splitting the processing steps of inventory growth, controls, chemical speciation, temporal allocation, and spatial allocation into independent steps whenever possible. The results from these steps are merged together in the final stage of processing using vector-matrix multiplication. It allows individual steps (such as adding a new control strategy, or processing for a different grid) to be performed and merged without having to redo all of the other processing steps (http://cf.unc.edu/cep/empd/products/smoke/version2.1/html/).

The emission processing for CMAQ for the 36km national domain and 12km Eastern domain (Domain 2) has been performed by New York State Department of Environmental Conservation (NYS DEC) (for base year 2002 and future year 2009) and by NESCAUM (for future year 2018) using SMOKE v2.1 compiled on a Red Hat 9.0

Linux operating system with the Portland group Fortran compiler version 5.1. They use the 2002 static emission inventory, CEM data, and surrogates data based on the 2002 RPO data. Biogenic emissions are calculated using BEIS3 with BELD3 data. Mobile source emissions are processed using MOBILE6. The updated Canada inventory for 2000 and 1999 Mexico inventory were used for processing.

The emissions processing was performed on a month-by-month and RPO-by-RPO basis, i.e. SMOKE processing was performed for each of the RPOs (MANE-VU, VISTAS, CENRAP, MRPO, WRAP) individually as well as for Canada and Mexico. Note the processing of WRAP and Mexican emissions was necessary for use with the 36 km grid modeling only. For each month/RPO combination, a separate SMOKE ASSIGNS file was created, and the length of the episode in each of these ASSIGNS files was set to the entire month. Specific data sources for individual source categories are listed below and the examples of processed emissions outputs are shown in Figure C-15.

Emissions Inventory

Area

MANE-VU:

MANEVU_AREA_SMOKE_INPUT_ANNUAL_SUMMERDAY_011705.txt
Fugitive dust correction: This was applied as county-specific correction factors for SCC's listed at http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; the correction factors were obtained from http://www.epa.gov/ttn/chief/emch/invent/transportfractions.xls; this adjustment was performed outside of SMOKE with in-house Fortran programs

VISTAS:

ida_ar_2002_rev_29sep04.vistas.emis, arinv_2002_ncnox_01apr05.emis, ida_ar_fire_typ_29nov04.vistas.emis, ida_ar_dust_2002_wfac_27nov04.vistas.emis
All files were obtained from Greg Stella (VISTAS) and were processed to extract VISTAS-only emissions as described above. Note: the header lines of these files indicate that the fugitive dust correction was already applied, so no further correction was performed.

MRPO:

ida_ar_2002_rev_29sep04.mrpo.emis,
ida_ar_fire_typ_29nov04.mrpo.emis,
ida_ar_dust_2002_wfac_27nov04.mrpo.emis
All files were obtained from Greg Stella (VISTAS) and processed to extract MRPO-only emissions as described above. Note: the header lines of these files indicate that the fugitive dust correction was already applied, so no further correction was performed.

CENRAP:

CENRAP_AREA_SMOKE_INPUT_ANN_STATES_120704.txt,
CENRAP_AREA_MISC_SMOKE_INPUT_ANN_STATE_120704.txt,
CENRAP_AREA_BURNING_SMOKE_INPUT_ANN_TX_AR_NELI_1
20704.txt,

CENRAP_AREA_MISC_SMOKE_INPUT_NH3_MONTH_{MMM}_120304.txt, where {MMM} is MAY, JUN, JUL, AUG, or SEP
CENRAP_AREA_SMOKE_INPUT_NH3_MONTH_{MMM}_120304.txt where {MMM} is MAY, JUN, JUL, AUG, or SEP
All files were downloaded from the CENRAP website http://www.cenrap.org/emission_document.asp. Fugitive dust correction: This was applied as county-specific correction factors for SCC's listed at: http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; the correction factors were obtained from http://www.epa.gov/ttn/chief/emch/invent/transportfractions.xls; this adjustment was performed outside of SMOKE with in-house Fortran programs

CANADA:

AS2000_SMOKEready.txt obtained from ftp://ftp.epa.gov/EmisInventory/canada_2000inventory . Fugitive dust correction: We applied "divide-by-four" correction for SCC's listed at http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; this adjustment was performed outside SMOKE with in-house Fortran programs. No county/province-specific correction factors were available for Canada

Nonroad

MANE-VU:

CT_NRD2002.IDA, DC_NRD2002.IDA, DE_NRD2002.IDA, MA_NRD2002.IDA, MD_NRD2002.IDA, ME_NRD2002.IDA, NH_NRD2002.IDA, NJ_NRD2002.IDA, NY_NRD2002.IDA, PA_NRD2002.IDA, RI_NRD2002.IDA, and VT_NRD2002.IDA contained in the "MANE-VU Nrd SMOKE files.zip" file prepared by PECHAN

VISTAS:

ida_nr_2002_rev_01oct.vistas.emis was obtained from Greg Stella (VISTAS) and processed to extract VISTAS-only emissions as described above.

MRPO:

ida_nr_2002_rev_01oct.wrap.emis, obtained from Greg Stella (VISTAS) and processed to extract WRAP-only emissions as described above.

CENRAP:

CENRAP_NONROAD_SMOKE_INPUT_ANN_120704.txt downloaded from the CENRAP website http://www.cenrap.org/emission_document.asp

CANADA:

File: NONROAD2000_SMOKEready.txt obtained from ftp://ftp.epa.gov/EmisInventory/canada_2000inventory

On-road

MANE-VU:

VMT/Speed: MANEVU_2002_mbinv.txt prepared by PECHAN

VISTAS:

VMT/Speed file: mbinv_2002_ida_vmt_22sep04.vistas.txt, obtained from Greg Stella (VISTAS) and was processed to extract VISTAS-only VMT as described above.

MRPO:

VMT/Speed file: mbinv_2002_ida_vmt_22sep04.mrpo.txt, obtained from Greg Stella (VISTAS) and were processed to extract MRPO-only VMT as described above in the VISTAS section. Note: Per email exchange between Gopal Sistla, Mark Janssen and Jeff Vukovich, it was determined that the VMT information used by VISTAS for their revised Phase II modeling reflects the latest MRPO information. Therefore, the MRPO-portion of the mobile source files obtained from Greg Stella (VISTAS), were used in this work.

CENRAP:

VMT/Speed files: mbinv02_vmt_cenrap_ce.ida, mbinv02_vmt_cenrap_no.ida, mbinv02_vmt_cenrap_so.ida, and mbinv02_vmt_cenrap_we.ida, downloaded from the CENRAP website http://www.cenrap.org/emission_document.asp

CANADA:

MOBILE2000_SMOKEready.txt obtained from ftp://ftp.epa.gov/EmisInventory/canada_2000inventory . Fugitive dust correction: applied "divide-by-four" correction for SCC's listed at http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; this adjustment was performed outside of SMOKE with in-house Fortran programs. No county/province-specific correction factors were available for Canada.

Point

MANE-VU:

MANEVU_Point_SMOKE_INPUT_ANNUAL_SUMMERDAY_122004.txt prepared by PECHAN was downloaded from ftp.marama.org (username mane-vu, password exchange). Fugitive dust correction: This was applied as county-specific correction factors for SCC's listed at http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; the correction factors were obtained from http://www.epa.gov/ttn/chief/emch/invent/transportfractions.xls; this adjustment was performed outside of SMOKE with in-house Fortran programs. Emission corrections were made for "THE HARTFORD STEAM CO" in Connecticut, Plant ID P0250.

VISTAS:

Annual: ptinv_2002typ_28nov04.vistas.ida and ptinv_fires_{MM}_typ.vistas.txt where {MM} is 01, 02, 03, etc. depending on the month.

Hour-specific: pthour_rev2002typ_{MMM}_08nov04.vistas.ems and pthour_fires_{MM}_typ.vistas.ida where {MMM} is jan, feb, mar, etc. and {MM} is 01, 02, 03, etc. depending on the month
All files were obtained from Greg Stella (VISTAS) and were processed to extract VISTAS-only emissions as described above.
Note: the header lines of these files indicate that the fugitive dust correction was already applied, so no further correction was performed.

MRPO:

Annual: ptinv_2002typ_28nov04.mrpo.ida and ptinv_fires_{MM}_typ.mrpo.txt where {MM} is 01, 02, 03, etc. depending on the month. Hour-specific: pthour_rev2002typ_{MMM}_08nov04.mrpo.ems and pthour_fires_{MM}_typ.mrpo.ida where {MMM} is jan, feb, mar, etc. and {MM} is 01, 02, 03, etc. depending on the month. All files were obtained from Greg Stella (VISTAS) and processed to extract MRPO-only emissions as described above. Note: the header lines of these files indicate that the fugitive dust correction was already applied, so no further correction was performed

CENRAP:

Annual: CENRAP_Point_SMOKE_INPUT_ANN_121004.txt. Hour-specific CEM: pthour.{QQ}.{ST}.txt where {QQ} is the quarter (q1, q2, q3, or q4) and {ST} is the state (AR, IA, KS, LA, MN, MO, NE, OK, TX) All files were downloaded from the CENRAP website http://www.cenrap.org/emission_document.asp . Fugitive dust correction: This was applied as county-specific correction factors for SCC's listed at http://www.epa.gov/ttn/chief/emch/invent/index.html#dust; the correction factors were obtained from http://www.epa.gov/ttn/chief/emch/invent/transportfractions.xls; this adjustment was performed outside of SMOKE with in-house Fortran programs

CANADA :

There has long been difficulty in obtaining an up-to-date Canadian criteria emissions inventory for point sources. This is due largely to confidentiality rights afforded to Canadian facilities. Thus far, the most recent inventory of Canadian point sources is rooted in the 1985 NAPAP data and is close to two decades old. Because there are a number of high emitting industrial facilities in southern Canada it is of particular importance to have a reasonably accurate inventory of these sources especially when modeling air quality over the Northeast and Midwest United States. Toward this end, an effort was made to obtain more recent Canadian point source data and incorporate it into an inventory database.

Perhaps the most accurate and publicly accessible source of Canadian pollutant data is now available from the National Pollutant Release Inventory (NPRI) database. This database contains 268 substances. Facilities that manufacture, process or otherwise use one of these

substances and that meet reporting thresholds are required to report these emissions to Environment Canada on an annual basis. The NPRI data are available at Environment Canada's website and can be found at the link http://www.ec.gc.ca/pdb/npri/npri_home_e.cfm. The page hosts an on-line search engine where one can locate emissions by pollutant or location. In addition, the entire database is available for download as an MS Access or Excel file. The NPRI database contains numerous pages with a rather comprehensive list of information. Detailed information is available about each facility, including location, activity and annual emissions. In addition, facilities having stacks with a height of 50 meters or more are required to report stack parameters.

Unfortunately, one of the limitations of the NPRI database for modeling purposes is that the data are only available at the facility level. Emissions models require process level information, so in order to use this data, a few generalizations had to be made. Each facility has a Standard Industrial Classification (SIC) code associated with it; however, emissions models require Source Classification Codes (SCC's). SCC's are of critical importance as the emissions models use these codes for assignment of temporal and speciation profiles. SIC codes describe the general activity of a facility while SCC codes describe specific processes taking place at each facility. While no direct relationship exists between these two codes, a general albeit subjective association can be made.

For the purposes of creating a model-ready inventory file it was necessary to obtain the whole NPRI database. After merging all the necessary components from the NPRI database required in the SMOKE inventory file, the SIC code from each facility was examined and assigned an SCC code. In most cases, only a SCC3 level code was assigned with confidence. While this is admittedly a less than desirable process, it does allow for the use of the most recent emissions from the NPRI database to be used in modeling. Furthermore, having some level of SCC associated with these emissions will ensure that they will be assigned a temporal and speciation profile by the model, other than the default. Once the model-ready inventory file was developed, it was processed through SMOKE.

Emissions Processing Files

Temporal Allocation

MANE-VU:

Area and nonroad sources: amptpro.m3.us+can.manevu.030205.txt and amptref.m3.manevu.012405.txt
Mobile source: MANEVU_2002_mtpro.txt and MANEVU_2002_mtref.txt
Point sources: Based on the same files as for the MANE-VU area and nonroad temporal files listed above, but added the VISTAS-generated CEM-based 2002 state-specific temporal profiles and cross-references for EGU sources for the MANE-VU states
No CEM-based hour-specific EGU emissions were used.

CENRAP:

The following temporal profiles and cross-reference files were used for all source categories: amptpro.m3.us_can.cenrap.010605.txt, amptref.m3.cenrap.010605.txt
These files were downloaded from the CENRAP website http://www.cenrap.org/emission_document.asp
For point sources, the CEM-based hour-specific EGU emissions described in Section 2.2.4 were utilized to override the annual-total based emissions whenever a match could be established by SMOKE

VISTAS, WRAP and MRPO:

The following month-specific temporal profiles and cross-reference files were used for all source categories:
amptpro_typ_us_can_{MMM}_vistas_27nov04.txt where {MMM} is jan, feb, mar, etc., amptref_2002_us_can_vistas_17dec04.txt
These files were obtained from Greg Stella (VISTAS)
For point sources (EGU and fires), the hour-specific emission files described in Sections 2.3.4 and 2.5.4 were utilized for the VISTAS and WRAP states to override the annual-total based emissions whenever a match could be established by SMOKE

Canada and Mexico:

For Canada and Mexico, the SMOKE2.1 default temporal profiles and cross-reference files (amptpro.m3.us+can.txt and amptref.m3.us+can.txt) were utilized.

Chemical speciation

The same speciation profiles (gspro.cmaq.cb4p25.txt) and cross-references (gsref.cmaq.cb4p25.txt) were utilized for all regions and all source categories. Different versions of these files were obtained (SMOKE2.1 default, EPA-CAIR modeling, VISTAS, CENRAP and MANE-VU) and compared. After comparing the creation dates and header lines of these files, it was determined that the EPA-CAIR and MANE-VU files had the most recent updates, and consequently the final speciation profile and cross-reference files used for all regions and source categories was based on the EPA-CAIR files with the addition of MANE-VU specific updates.

Spatial Allocation

U.S.

The spatial surrogates for the 12 km and 36 km domains were extracted from the national grid 12 km and 36 km U.S. gridding surrogates posted at EPA's website at http://www.epa.gov/ttn/chief/emch/spatial/newsurrogate.html The gridding cross-references were also obtained from this website, but for the processing of MANE-VU area source emissions, MANE-VU specific cross-reference entries posted on the MARAMA ftp site were added.

Canada

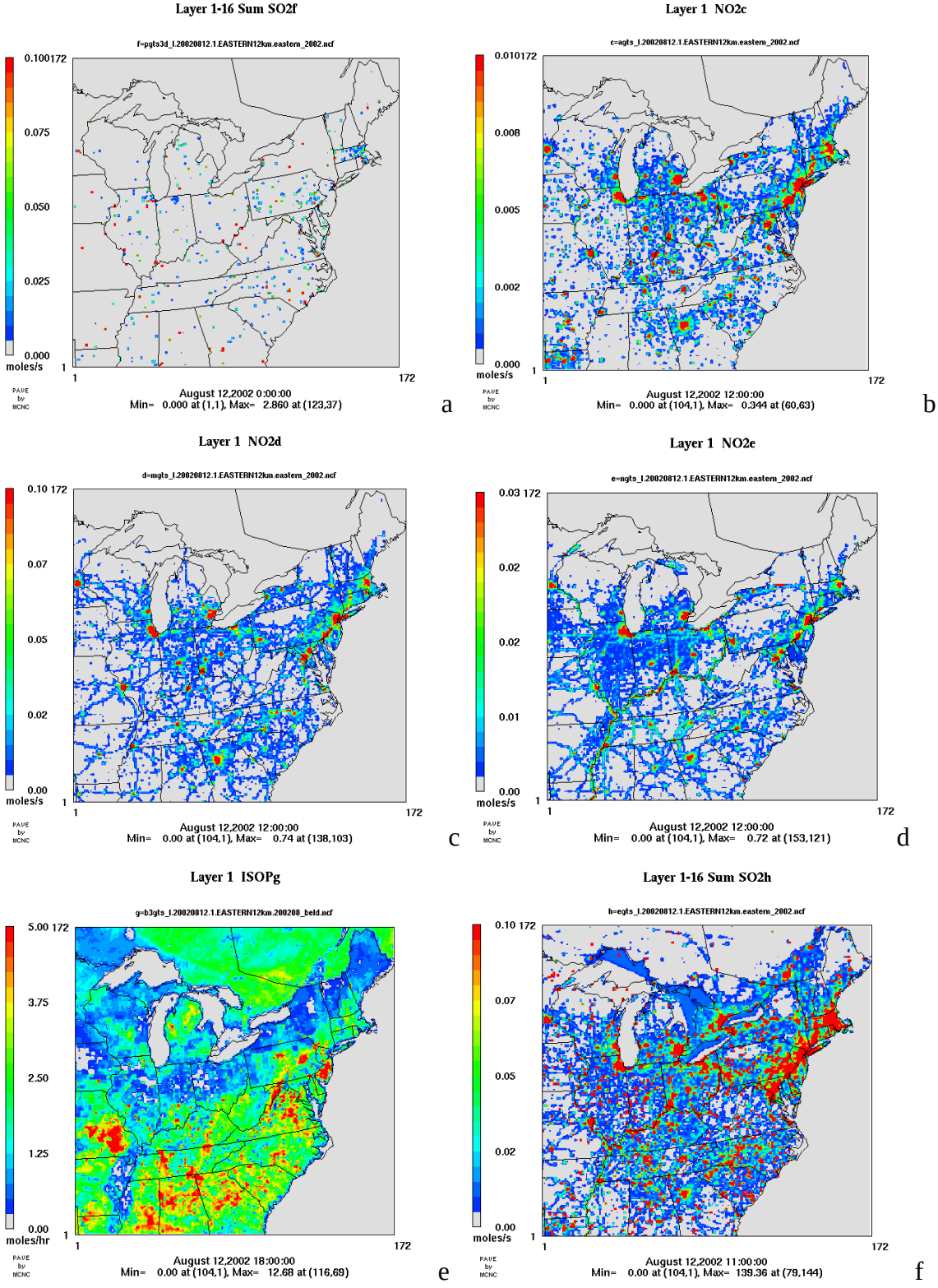
The spatial surrogates for Canadian emissions for the 12 km and 36 km domains were extracted from the national grid 12 km and 36 km Canadian gridding surrogates posted at EPA's website at http://www.epa.gov/ttn/chief/emch/spatial/newsurrogate.html The gridding cross-references were also obtained from this website.

Mexico

The spatial surrogates for Mexican emissions the 36 km domain were extracted from the national 36 km gridding surrogates used by EPA in the CAIR modeling. These files were obtained from EPA's CAIR NODA ftp site http://www.airmodelingftp.com . The gridding cross-references were also obtained from this ftp site.

Figure C-15. Examples of processed model-ready emissions

(a): SO2 from Point; (b): NO2 from Area; (c): NO2 from On-road; (d): NO2 from Non-Road; (e): ISOP from Biogenic; (f): SO2 from all source categories)



C.2.2. REMSAD

The Regional Modeling System for Aerosols and Deposition (REMSAD) is also a three-dimensional Eulerian model designed to support a better understanding of the distributions, sources, and removal processes relevant to fine particles and other airborne pollutants. It calculates the concentrations of both inert and chemically reactive pollutants by simulating the physical and chemical processes in the atmosphere that affect pollutant concentrations. The basis for the model is the atmospheric diffusion equation representing a mass balance in which all of the relevant emissions, transport, diffusion, chemical reactions, and removal processes are expressed in mathematical terms. The REMSAD model performs a four-step solution procedure: emissions, horizontal advection/diffusion, vertical advection/diffusion and deposition, and chemical transformations during one half of each advective time step, and then reverses the order for the following half time step. The maximum advective time step for stability is a function of the grid size and the maximum wind velocity or horizontal diffusion coefficient. Vertical diffusion is solved on fractions of the advective time step to keep their individual numerical schemes stable.

REMSAD uses a flexible horizontal and vertical coordinate system with nested-grid capabilities and user-defined vertical layers. It accepts a geodetic (latitude/longitude) horizontal coordinate system or a Cartesian horizontal coordinate system measured in kilometers. REMSAD uses a simplified version of CB-IV chemistry mechanism which is based on a reduction in the number of different organic compound species and also includes radical-radical termination reactions. The organic portion of the chemistry is based on three primary organic compound species and one carbonyl species. The model parameterizes aerosol chemistry and dynamics for PM and calculates SOA yields from emitted hydrocarbons. REMSAD V7.12 and newer versions have capabilities that allow model tags of sulfur species (up to 11 tags), nitrogen (4 tags), mercury (up to 24 tags), and cadmium (up to 10 tags) to identify the impact of specific tagged species.

Unlike CMAQ, REMSAD provides no choice of chemical and physical mechanisms. The modeling configuration for future work with REMSAD will be similar to the CMAQ modeling setup. The initial concentrations and boundary conditions will be generated using the same concentration profile used by CMAQ. The approach is to use similar model inputs to allow comparison of REMSAD with CMAQ to better understand differences between the two models. Due to the simplified chemistry mechanism, REMSAD may not simulate atmospheric processes as well as CMAQ. However, advantages such as the tagging feature for sulfur, more efficient modeling, and reasonable correspondence with measurements for many species, make REMSAD an important source apportionment tool for MANE-VU.

In our present REMSAD modeling, the same 12km domain (i.e. domain2) presented in the previous section is used for a three full annual runs for the base year (2002). Multiple runs are necessary to permit tagging of sulfur emissions for all of the states in the domain, Canada and the boundary conditions.

C.2.2.1. REMSAD Meteorology

For the regional haze modeling study, an entire year of MM5 generated meteorological data for 2002 was provided by UMD and used to drive annual CMAQ and REMSAD modeling as described in the previous section. Meteorological inputs for these runs were developed by applying a simple translation script (MM52REMSAD) provided by the University of Maryland (Jeff Stehr, personal communication) to generate hourly meteorological field required by REMSAD.

C.2.2.2. REMSAD Emissions

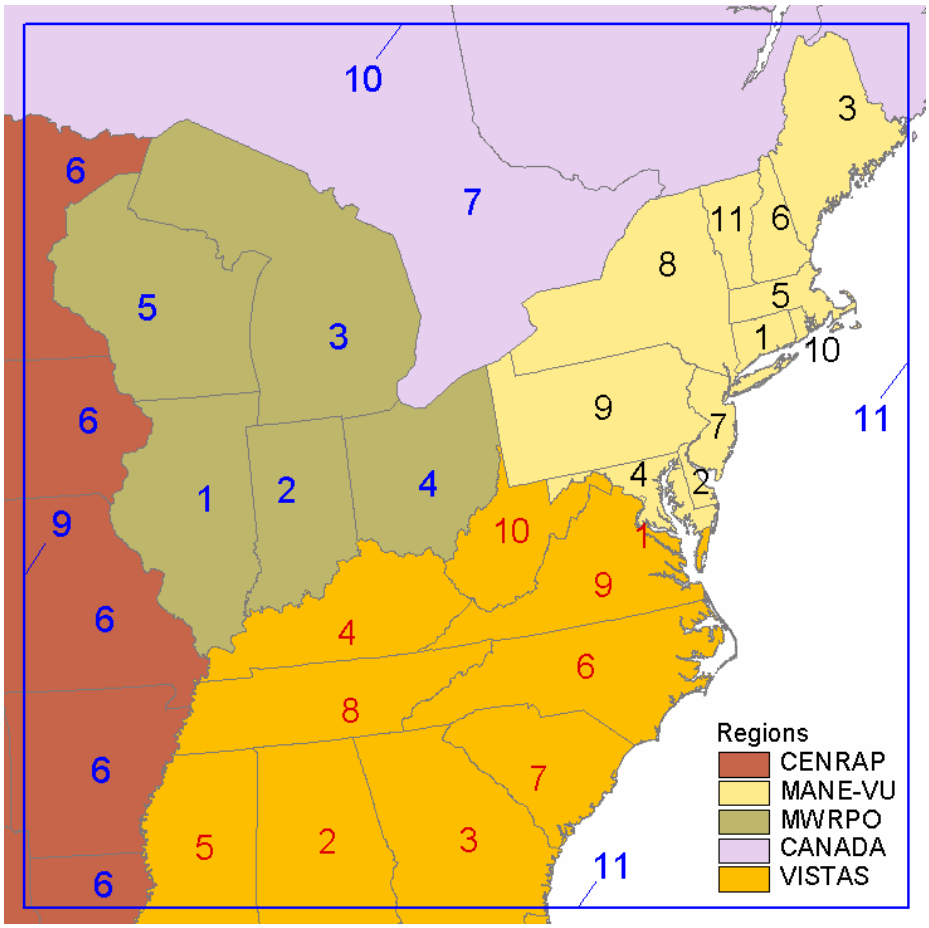
Emissions processed for REMSAD take advantage of the SO_2 tagging capabilities, whereas CMAQ treats all SO_2 as indistinguishable. NESCAUM has developed emissions tagging techniques for application in air quality impact analyses using the REMSAD tagging scheme incorporated in REMSAD version 7.10 and higher. In general, these emissions tagging schemes can be used to assess source contributions in various ways including: (1) by size and susceptibility to transport (e.g., as between large elevated sources vs. small, low-level sources); (2) by sectors/types (e.g. by SCCs or by point, area, or mobile source categories); (3) by regions (e.g. by country/state/county); or (4) by combinations (e.g. largest electricity generating unit (EGU) in a specific state).

The emissions inventory used for emissions tagging for REMSAD processing was primarily the same as the inventory described in the previous section (i.e. CMAQ platform). Some differences in emissions, however, do exist because of emissions updates (MANE-VU) and the necessity of simplified emissions processing to compensate for the added complexity introduced by tagging. Since REMSAD is a simpler model than CMAQ and is used mostly for longer term (> monthly) impact analysis, using a simplified emissions processing approach is reasonable. The major differences are:

1) The point source emissions for MANE-VU were updated from ver.1 to ver. 2.
2) CEM data were not used
3) RPO-by-RPO emissions files were merged first, prior to processing in SMOKE
4) Anthropogenic emissions were tagged

MANE-VU had already conducted some preliminary runs to inform the early stages of regional haze planning. NESCAUM has taken the additional step of processing source emission files such that the model input is formatted to take advantage of REMSAD's tagging capabilities. Thus, all combustion and industrial process emissions sources in over 30 Eastern states in the modeling domain have been tagged according to their state of origin, providing an estimate of the contribution those sources in each state make toward simulated sulfate concentrations at Eastern receptor sites. Moreover, boundary conditions were tagged to assess out-of-domain impact. The tagging scheme employed for this analysis is shown in Figure C-16.

Figure C-16. REMSAD modeling tagging schemes.
(black: group 1, red: group 2, and blue: group 3)



Note: Sulfur species from anthropogenic emission sources are tagged by states for three sets of tags. Tag group 3 also includes boundary conditions. The color of the numbers represents tag groups (black: group 1, red: group 2, and blue: group 3)

C.3. Model Evaluation and Results

C.3.1. CMAQ

C.3.1.1. CMAQ Performance evaluation of $PM_{2.5}$ species

CMAQ modeling has been conducted for the year 2002 (completed by cooperative modeling efforts from NYDEC, UMD, NJDEP, Rutgers, VADEP, and NESCAUM) under Base A emission scenario and for 5-month summer period of 2002 (completed by NYDEC) under Base A1 (refined from Base A) emission scenario. CMAQ performance for $PM_{2.5}$ species and visibility is examined based on these two CMAQ runs on a 12km resolution domain. Measurements from IMPROVE and STN networks are used to pair with model predictions by location and time for evaluation. Figure C-17 presents the domain wide paired comparison of $PM_{2.5}$ species (Sulfate,

Nitrate, OC, EC, Fine Soil, and $PM_{2.5}$) daily average concentration between two sets of simulations (BaseA & BaseA1) and two sets of observations (STN & IMPROVE). It shows that predicted $PM_{2.5}$ Sulfate (top row left panel) and measured Sulfate are in a good 1:1 linear relationship with r^2 varying from 0.6 to 0.7. $PM_{2.5}$ Nitrate (top row right panel) also has close to a 1:1 linear relationship between the model and observations, although the r^2 values are much lower (from ~0.2 to ~0.5) than for Sulfate. Paired OC (mid row left panel) concentrations have a scattered distribution with over- and under-estimating and a very weak linear relationship (r^2 of ~0.1). CMAQ tends to overestimate EC (mid row right panel) and fine soil (bottom row left panel) concentration. EC and soil are inert species not involved in chemical transformation. Poor emission inventory data may be the main cause for the weak linear relationships between prediction and measurement. In addition, there is no fire emission considered in CMAQ modeling. The wild fire in Quebec, Canada in early July of 2002 led to high concentrations of observed OC, EC, and Fine Soil that are not predicted by CMAQ. Since Sulfate is the dominant $PM_{2.5}$ species, modeled $PM_{2.5}$ (bottom row right panel) also shows a relatively strong near 1:1 linear relationship (slope between 0.7~0.8 with r^2 of 0.4~0.5).

Similar comparisons of $PM_{2.5}$ species between prediction and observation from other RPOs' CMAQ modeling work (*Ku et al.*, 2005) are shown in Figure C-18. Results from WRAP CMAQ run and CENRAP CMAQ run paired at 21 IMPROVE sites in July 2002 are on the left panel; while results from VISTAS CMAQ run and CENRAP CAMx run at 17 IMPROVE sites in July 2002 are on the right panel. The rows from top to bottom are Sulfate, Nitrate, OC, EC, and Fine Soil. For $PM_{2.5}$ Sulfate, other RPOs' prediction show a rather strong 1:1 linear relation with measurement (r^2 of 0.6~0.7), similar to our CMAQ performance. Other $PM_{2.5}$ species all show poor linear relationships between model prediction and observations. Values of r^2 are ~0.1 for Nitrate; 0.2~0.4 for OC; 0.2~0.4 for EC; <0.02 for Fine Soil. Therefore, our CMAQ performance of $PM_{2.5}$ is in the same range as other RPOs.

Figure C-19 describes the spatial distribution of the correlation coefficient of sulfate between CMAQ prediction and observations (STN data on the top row and IMPROVE data on the bottom row) at network sites. CMAQ predictions show a similar spatial pattern of correlation with both networks. Generally, the north region of the domain has stronger correlations than do the south region. Correlation coefficients within MANE-VU region are highest (~0.9 in average) compared to other RPO regions. The fact that summer time correlation coefficients are higher than annual values indicates CMAQ performs better for summer than for other seasons. No significant improvement in correlation is observed for the BaseA1 case over the BaseA case. The spatial distribution of correlation coefficient for $PM_{2.5}$ is presented in Figure C-20. The $PM_{2.5}$ correlation coefficient spatial pattern follows $PM_{2.5}$ sulfate correlation coefficient, although at the same observation site coefficient values are ~0.1 lower than the sulfate coefficient value. Like $PM_{2.5}$ sulfate, CMAQ also performs the best for $PM_{2.5}$ in the MANE-VU region with ~0.7 annual average and ~0.8 summer average of the correlation coefficient.

In 2004 James Boylan from VISTAS suggested the goal and the criteria for $PM_{2.5}$ evaluation (*Boylan,* 2004). This standard has been adopted by every RPO for SIP modeling. The proposed performance goals are: Mean Fractional Error (MFE) <=

+50%, and Mean Fraction Bias (MFB) <= +-30%; while the criteria is proposed as: MFE <= +75%, and MFB <= +-60%.

CMAQ prediction of $PM_{2.5}$ species from 40 STN sites and 17 IMPROVE sites within MANE-VU region are paired with measurements and statistically analyzed to generate MFE and MFB values. Figure C-21 presents MFE of $PM_{2.5}$ Sulfate, Nitrate, OC, EC, Fine Soil, and $PM_{2.5}$, and curves of the goal and criteria. MFB values are shown in Figure C-22. Considering CMAQ performance in terms of both MFE and MFB, Sulfate, Nitrate, OC, EC, and $PM_{2.5}$ all have the majority of data points within the goal curve, some are between the goal and acceptable criteria, and only a few outside the criteria curve. Only fine soil has the majority of points outside the criteria curve, but there are some sites still within the goal. For the MANE-VU region, CMAQ performs best for $PM_{2.5}$ Sulfate, followed by $PM_{2.5}$, EC, Nitrate, OC, and finally the fine soil.

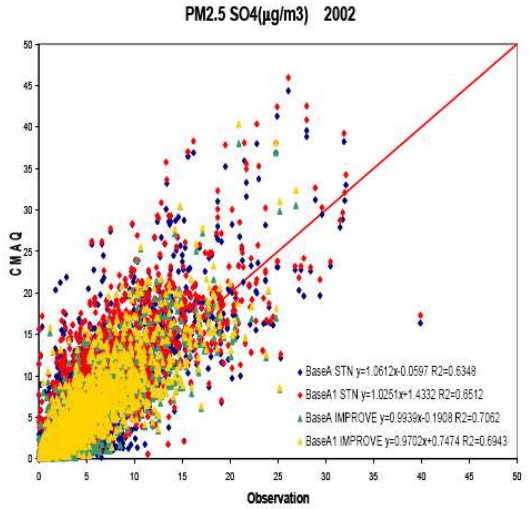
Regional haze modeling also requires a CMAQ performance evaluation for aerosol extinction coefficient (Bext) and the haze index. Modeled daily aerosol extinction at each IMPROVE site is calculated following the IMPROVE formula with modeled daily $PM_{2.5}$ species concentration and relative humidity factor from IMPROVE. Figure C-23 shows the paired comparison of domain wide daily aerosol extinction coefficient for 2002 between prediction and measurement with 1:1 line and ±20% lines. The modeled Bext shows a near 1:1 linear relationship (slope of 0.74 and r^2 of 0.53) with IMPROVE observed Bext. Mean bias is -6.31 Mm^{-1}, which is less than 1% compared to mean Bext of either observation (76.54 Mm^{-1}) or prediction (70.22 Mm^{-1}). MFE of 35% and MFB of -13% both meet the standard goal. CMAQ prediction of the aerosol extinction coefficient agrees well with IMPROVE observation because CMAQ performs well on sulfate, which dominates aerosol extinction. Further, the modeled haze index (HI) is calculated based on modeled Bext. Figure C-24 presents the paired comparison of HI values at 4 Class I sites in the Eastern US between CMAQ prediction and IMPROVE measurement for 2002 with 1:1 line and +-20% lines. The majority of the data points are within the ±20% regime. Acadia shows the best model performance with a slope of 0.96 and r^2 of 0.64, mean bias of 0.05 compared to mean HI of ~15. Next is Brigantine, with a slope of 0.86 and r2 of 0.5, mean bias of 0.2 compared to an HI of ~20. Then is Lye Brook with a slope of 0.77 and r^2 of 0.6, mean bias of 1.55 compared to an HI of ~14. Finally is Shenandoah with a slope of 0.6 and r^2 of 0.4, mean bias of 1.62 compared to an HI of ~21. MFE and MFB of all 4 sites meet the performance goal.

Overall, NESCAUM CMAQ modeling on the 12km resolution domain for 2002 accurately portrays sulfate, $PM_{2.5}$, aerosol extinction coefficient and the Haze Index. It provides reasonable performance for $PM_{2.5}$ Nitrate, OC, and EC. The model performs better for summertime than for wintertime, and better in the MANE-VU region than in others regions.

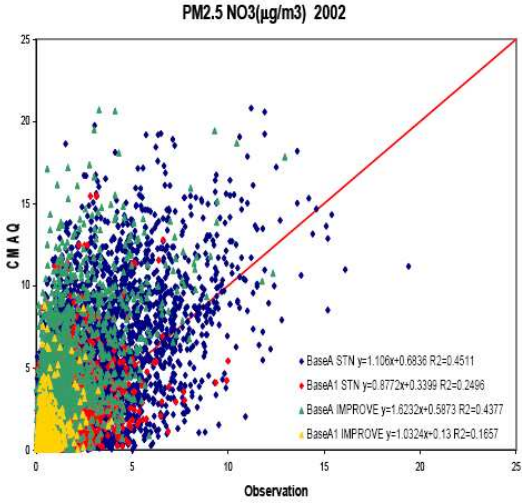
In late 2005, CMAQ V4.5 was released to the public. Although our current modeling results are acceptable, MANE-VU modelers have been considering a shift from CMAQ V4.4 to V4.5. CMAQ V4.5 is reported to significantly improve the PM science in the model. The more recent model version has added sea salt (fine equilibrium; non-interactive coarse mode) and updated the module of PM treatment (AERO4). It revised the aerosol dry deposition algorithm and corrected inconsistencies. It improved ISORROPIA and fixed discontinuities. It also added a new sub-grid cloud mixing

algorithm/module. In addition, CMAQ V4.5 updated the planetary boundary layer (PBL) module to use urban fraction for setting minimum vertical diffusivity (K_Z). Comparison studies (Appel et al., 2005) between CMAQ V4.5 and V4.4 show significantly improved performance on $PM_{2.5}$ Sulfate and Nitrate using V4.5 over V4.4, while V4.5 maintains the same acceptable ozone performance as V4.4. While other RPOs have already used CMAQ V4.5 in their SIP modeling, MANE-VU recently decided to switch to CMAQ V4.5 based on results of comparison study conducted by NJ DEP. MANE-VU believes that V4.5 would improve PM performance for Regional Haze and $PM_{2.5}$ SIP modeling for MANE-VU.

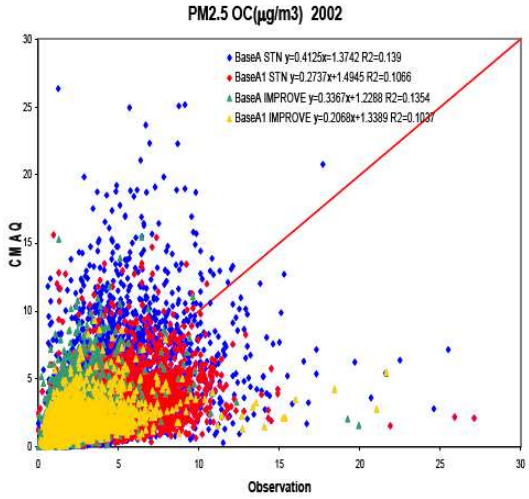
Figure C-17. Domain wide paired comparison of daily average $PM_{2.5}$ species between CMAQ predictions and measurements from IMPROVE and STN networks



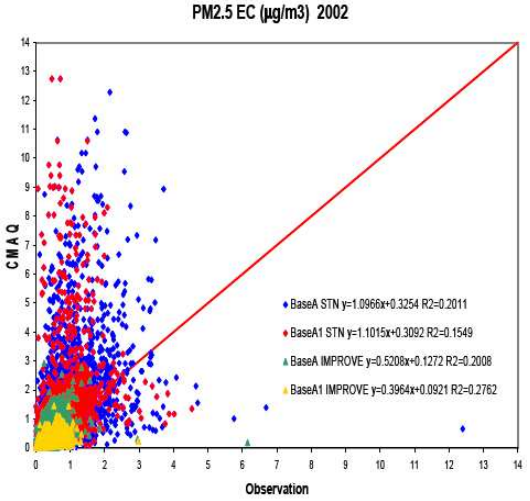
PM2.5 Sulfate



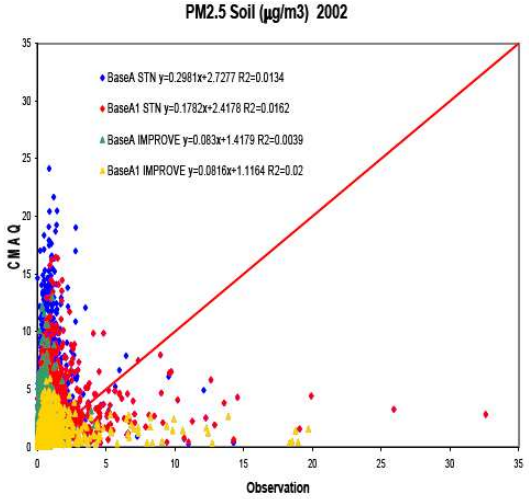
PM2.5 Nitrate



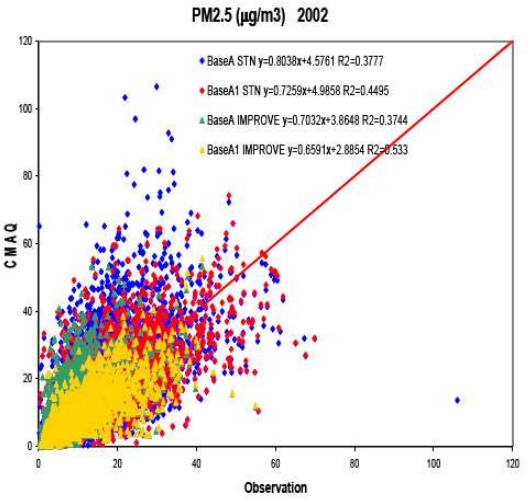
PM2.5 Organic Carbon



PM2.5 Elemental Carbon



PM2.5 Soil



PM2.5

Figure C-18. Paired comparison of $PM_{2.5}$ species from other RPOs' modeling work (*Ku et al.,2005)*

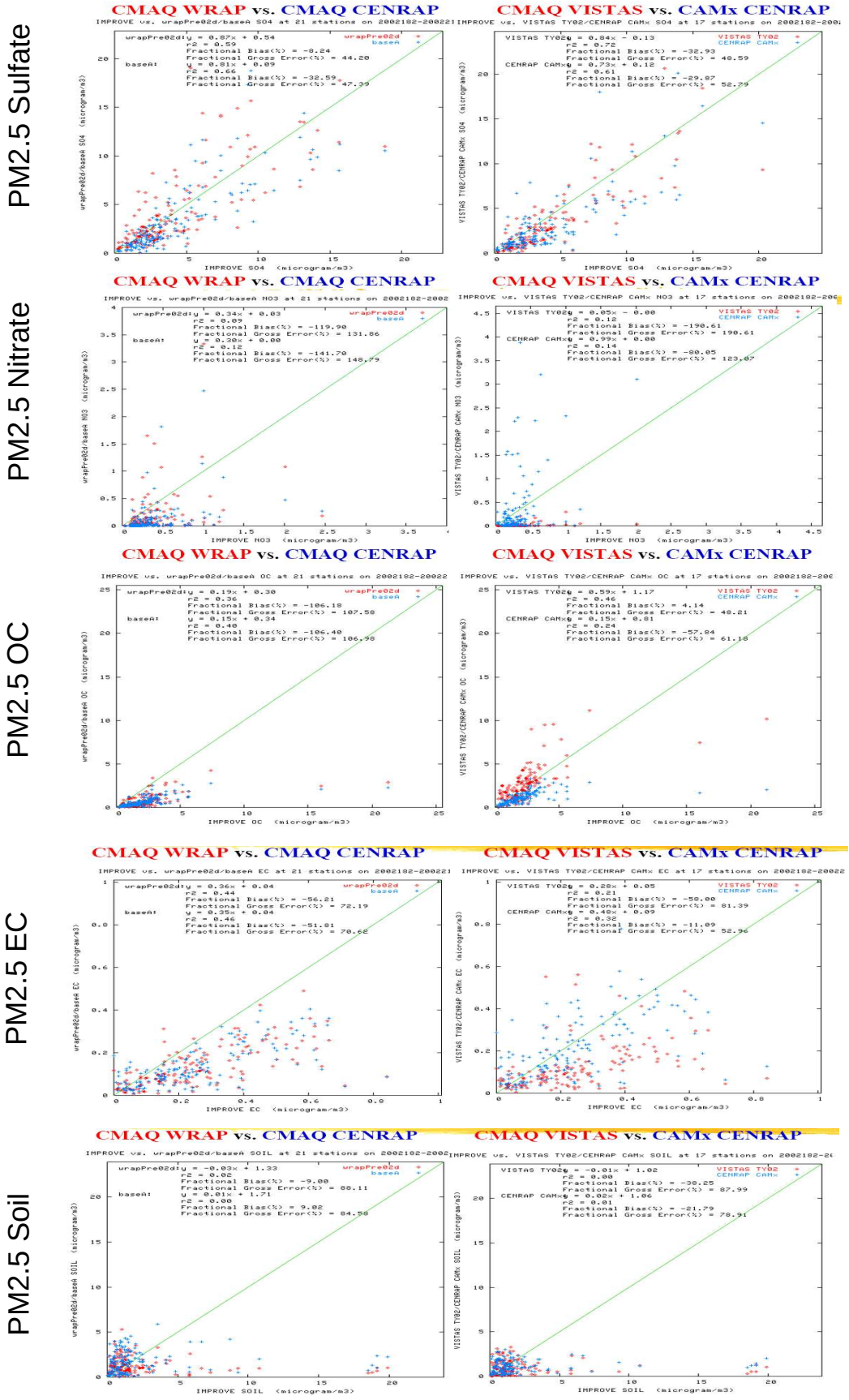


Figure C-19. Spatial distribution of correlation coefficient between $PM_{2.5}$ Sulfate and measurement

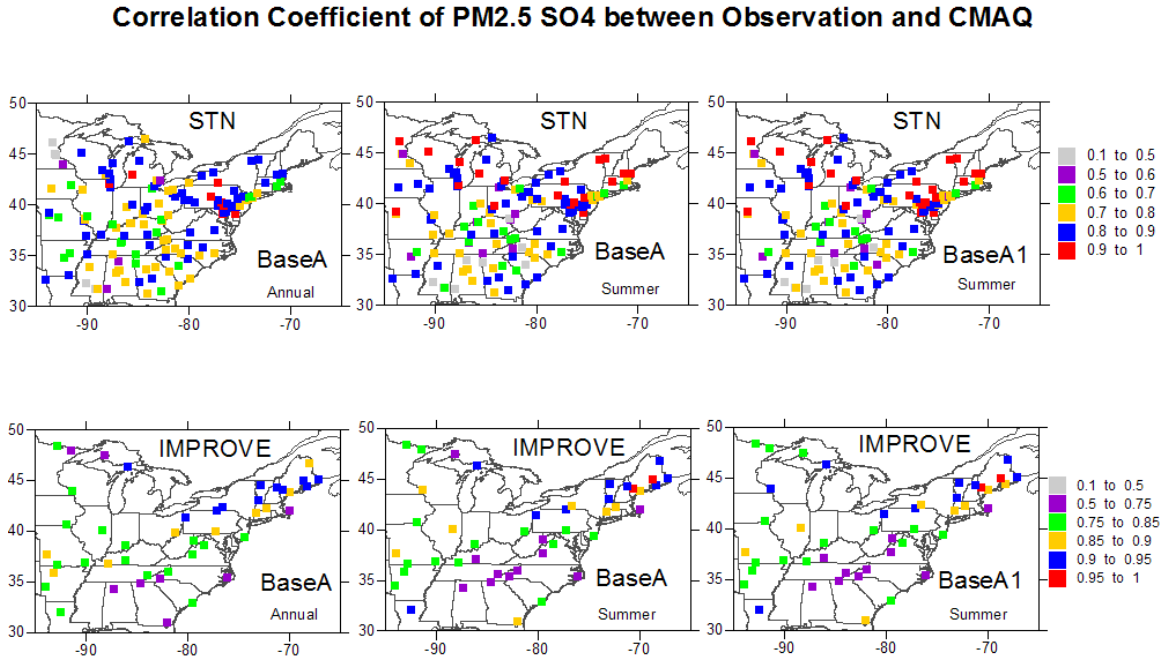


Figure C-20. Spatial distribution of correlation coefficient between $PM_{2.5}$ and measurement

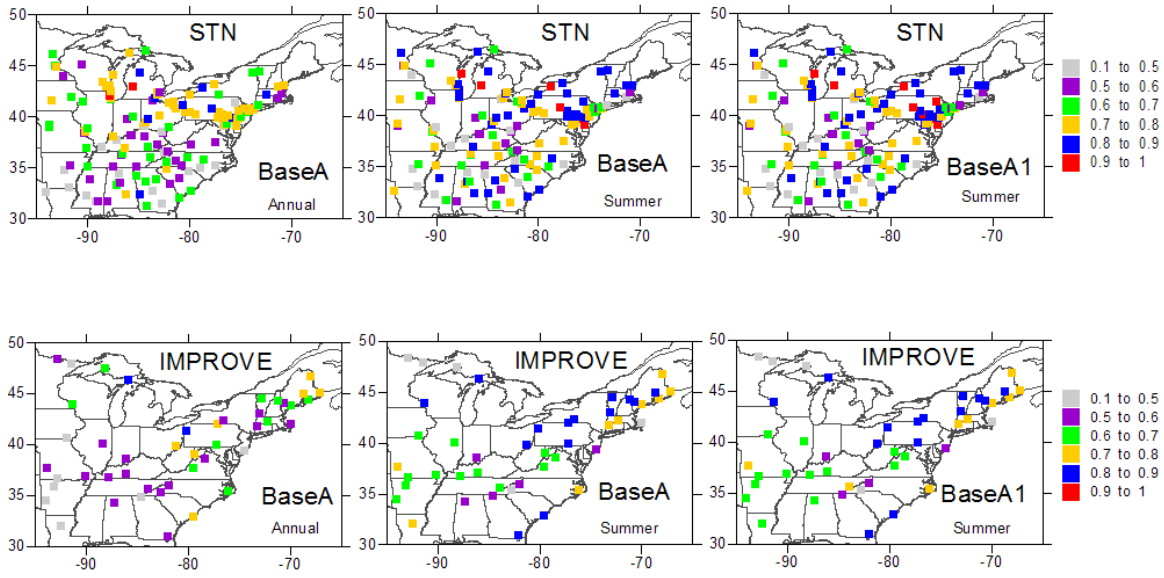
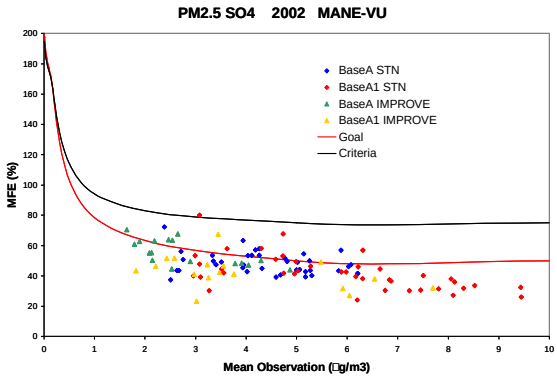
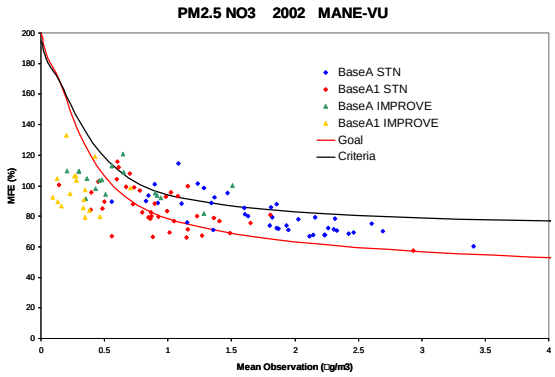


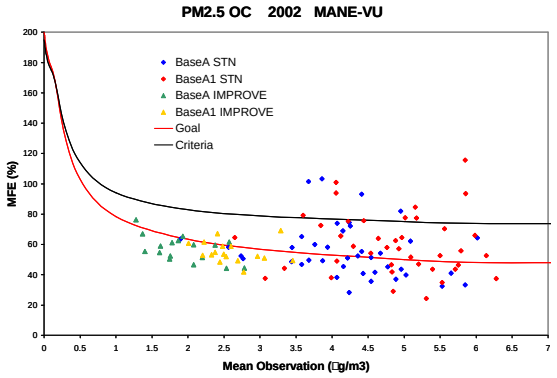
Figure C-21. Mean Fractional Error of $PM_{2.5}$ species within MANE-VU region



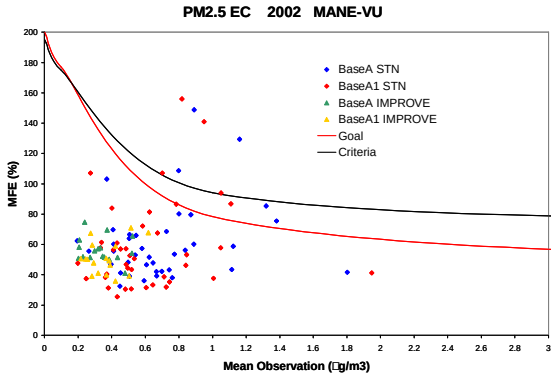
PM2.5 Sulfate



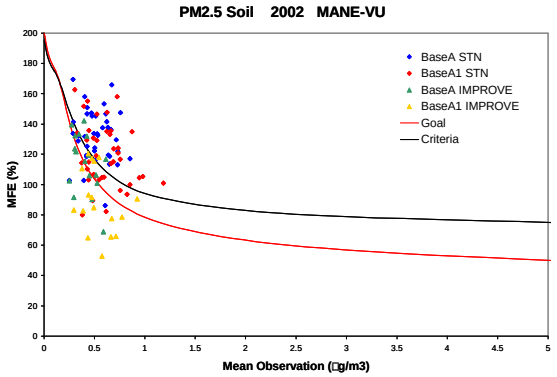
PM2.5 Nitrate



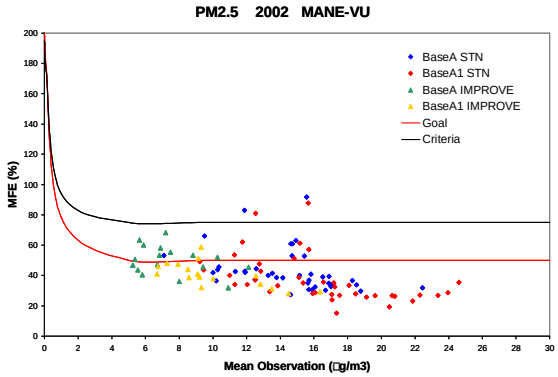
PM2.5 Organic Carbon



PM2.5 Elemental Carbon

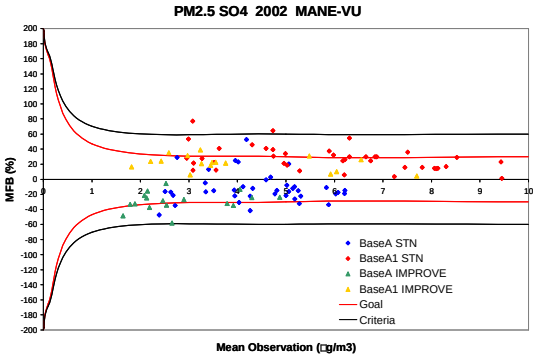


PM2.5 Fine Soil

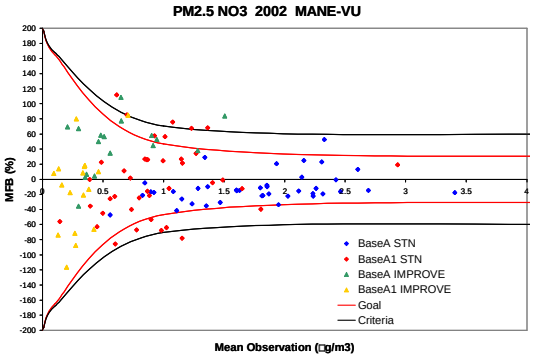


PM2.5

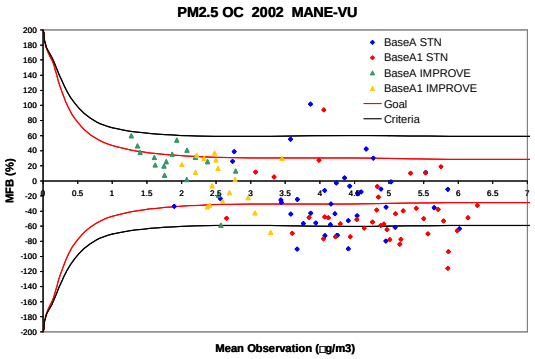
Figure C-22. Mean Fraction Bias of $PM_{2.5}$ species within MANE-VU region



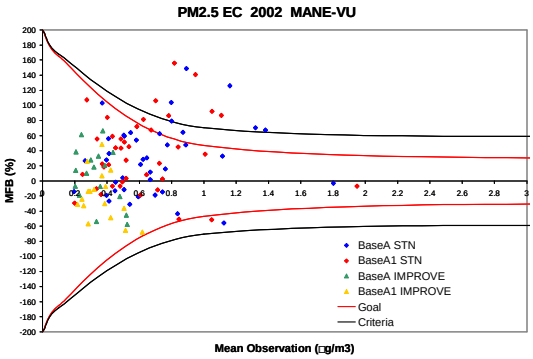
PM2.5 Sulfate



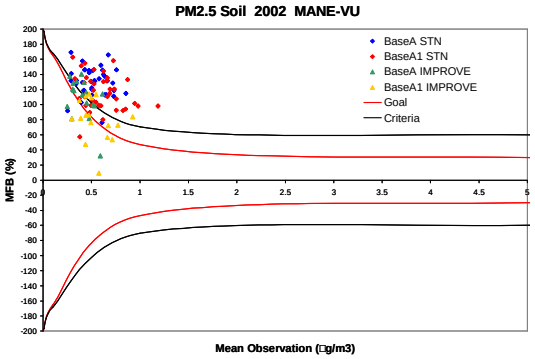
PM2.5 Nitrate



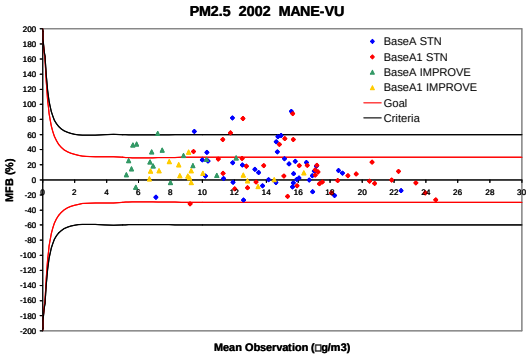
PM2.5 Organic Carbon



PM2.5 Elemental Carbon

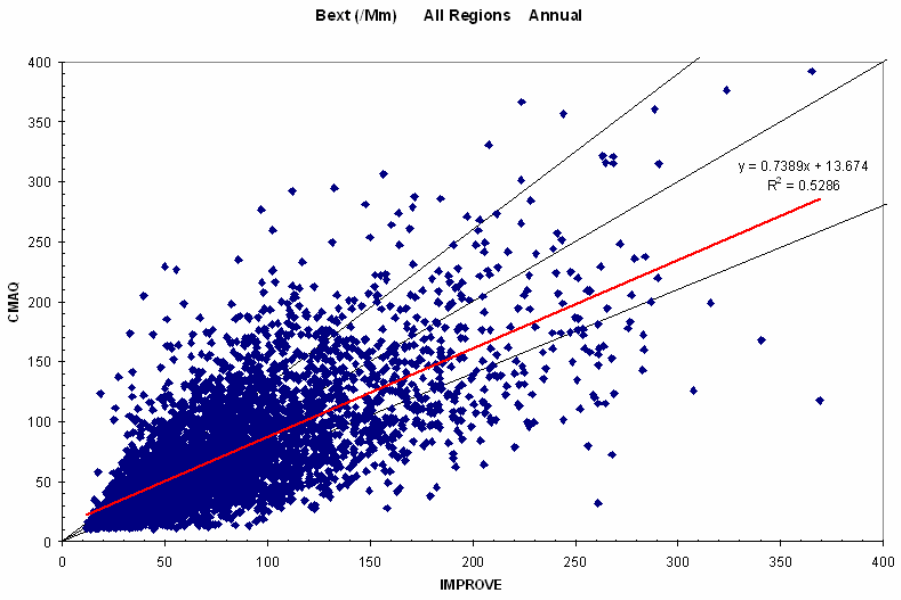


PM2.5 Fine Soil



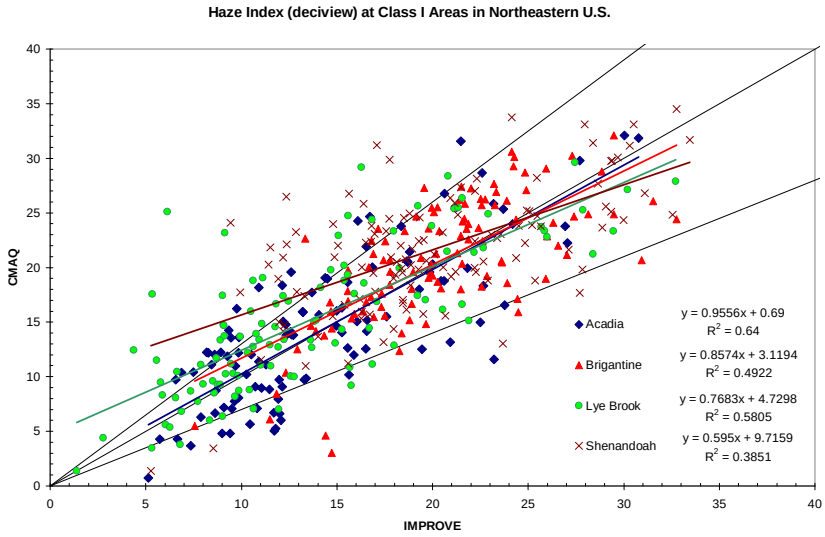
PM2.5

Figure C-23. Paired comparison of extinction coefficient between CMAQ prediction and IMPROVE measurement



Mobs	Mpre	MB	ME	FB(%)	FE(%)	NMB(%)	NME(%)	MNB(%)	MNE(%)
76.54	70.22	-6.31	25.67	-12.64	35.34	-8.25	33.54	-3.25	34.22

Figure C-24. Paired Comparison of Haze Index between CMAQ prediction and IMPROVE measurement at Class I sites within Northeastern US



Class I area	Mobs	Mpre	MAXobs	MAXpre	MB	ME	NMB(%)	NME(%)	FB(%)	FE(%)	MNB(%)	MNE(%)
Acadia	14.50	14.54	30.77	32.08	0.05	3.39	0.32	23.35	-4.58	27.28	0.71	26.20
Brigantine	20.47	20.67	32.75	32.08	0.20	3.28	0.98	16.02	-1.37	17.15	1.22	16.45
Lye Brook	13.71	15.26	32.72	29.64	1.55	3.59	11.34	26.22	11.01	26.07	19.68	32.75
Shenandoah	20.00	21.61	33.46	34.53	1.62	4.05	8.08	20.25	7.28	21.58	12.41	24.36

C.3.1.2. CMAQ Control Scenario Results

The results from preliminary annual simulations of the 2002 baseline, 2009 and 2018 future case scenarios have been reviewed. These model runs provide insight into the current and expected ambient levels of fine particles and haze causing constituents. The general character should remain consistent with future model runs that may use updated model code (CMAQ V4.5) and revised emissions (MANE-VU v3.0 inventory).

The six maps in

Figure C-25 show annual average results for $PM_{2.5}$ and sulfate for three modeling runs (2002/2009/2018). The other PM25 constituents are of lower concern for regional haze in MANE-VU, as sulfate dominates visibility degradation in the region. The total PM25 maps on the left provide the spatial distribution throughout the modeling domain, with levels in urban centers highest for all three model runs. The spatial distribution of annual sulfate levels is somewhat different from the $PM_{2.5}$. Specifically, sulfate levels along the Ohio River valley are greater than surrounding areas for the baseline run, with the gradients becoming much less for future scenarios.

Sulfate results are investigated further in Figure C-26. The set of six maps in this figure show relative reduction factors for sulfate. Model results are used in a relative sense to address potential uncertainties in the absolute results; uncertainty in relative changes is believed to be smaller than the absolute uncertainty. The top two figures display the ratio of sulfate results from future case runs to the base case for 2009 and 2018 respectively. For regional haze purposes, site specific reduction factors were generated for the best and worst 20% days. The annual average results are spatially consistent with the seven Class 1 site factors derived for the worst 20% days

[3]. These model results predict the greatest percentage sulfate reduction to occur in West Virginia and its immediate surrounds, with minor reductions calculated for areas west of the Mississippi River and moderate declines in the more northeastern section of MANE-VU.

The bottom four panels of Figure C-26 provide quarterly estimates of relative reduction in sulfate for 2009. For $PM_{2.5}$ mass, EPA guidance recommends the use of a quarterly reduction factor, unlike the 20% best and worst factors used in haze calculations. These results are instructive and improve the understanding of the seasonal impacts to be expected in the future due to emission changes. The broadest reductions occur in the third quarter, followed closely by the second quarter. The pattern for the first and fourth quarters differs substantially from the warmer months, with much of the domain predicted to exhibit increases in sulfate during the colder months of 2009 relative to the base year 2002. For the most-part, the changes modeled in the colder months are modest, falling within 10% of the base-year calculations.

[3] To project future haze levels, relative reduction factors were determined for best and worst days for all six haze relevant constituents (sulfate, nitrate, organic carbon, elemental carbon, fine soil and coarse particulate). The seven sites investigated were Acadia, Brigantine, Lye Brook, Moosehorn, Great Gulf, Dolly Sods and Shenandoah.

Figure C-25. CMAQ results for $PM_{2.5}$ and Sulfate for 2002, 2009 and 2018

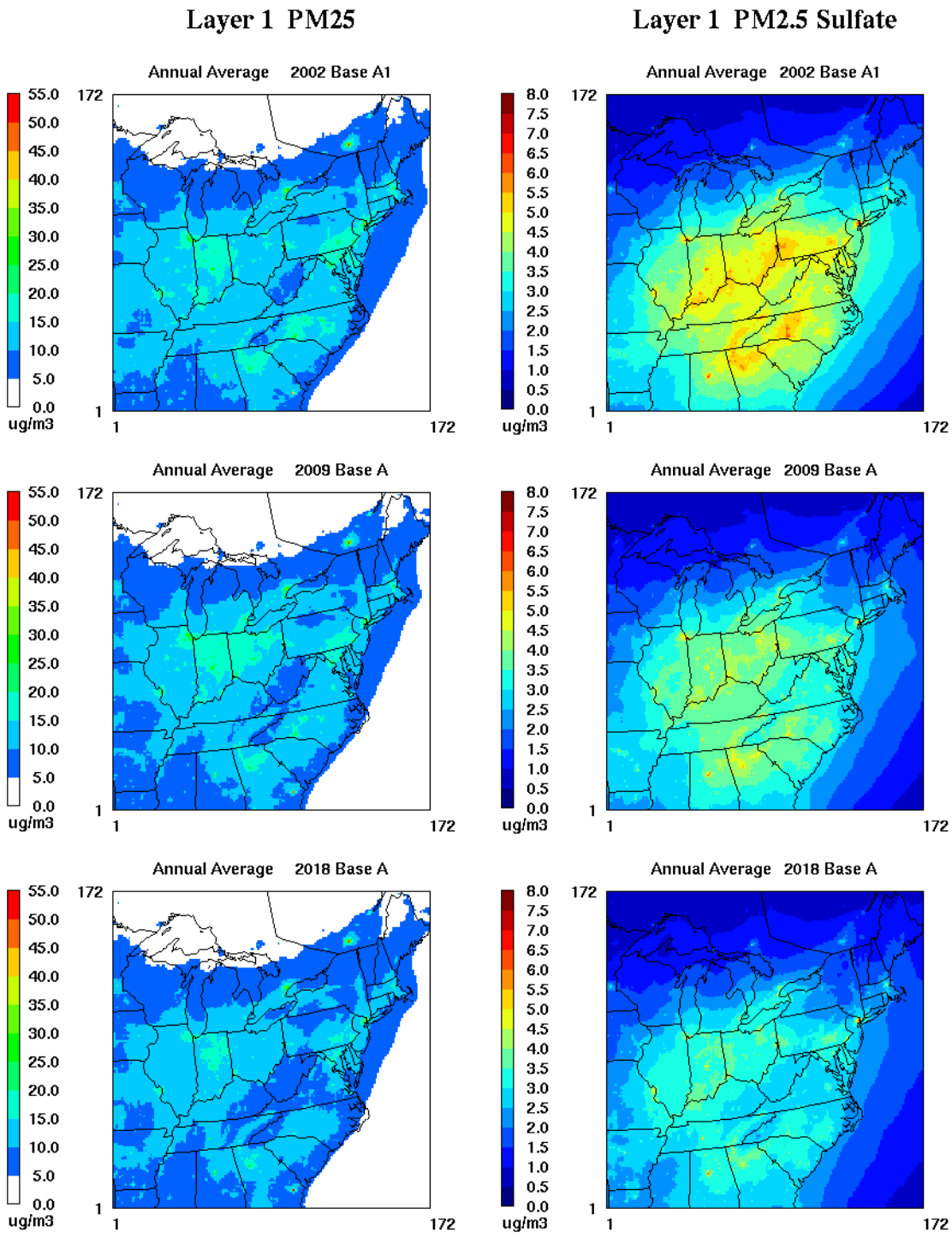
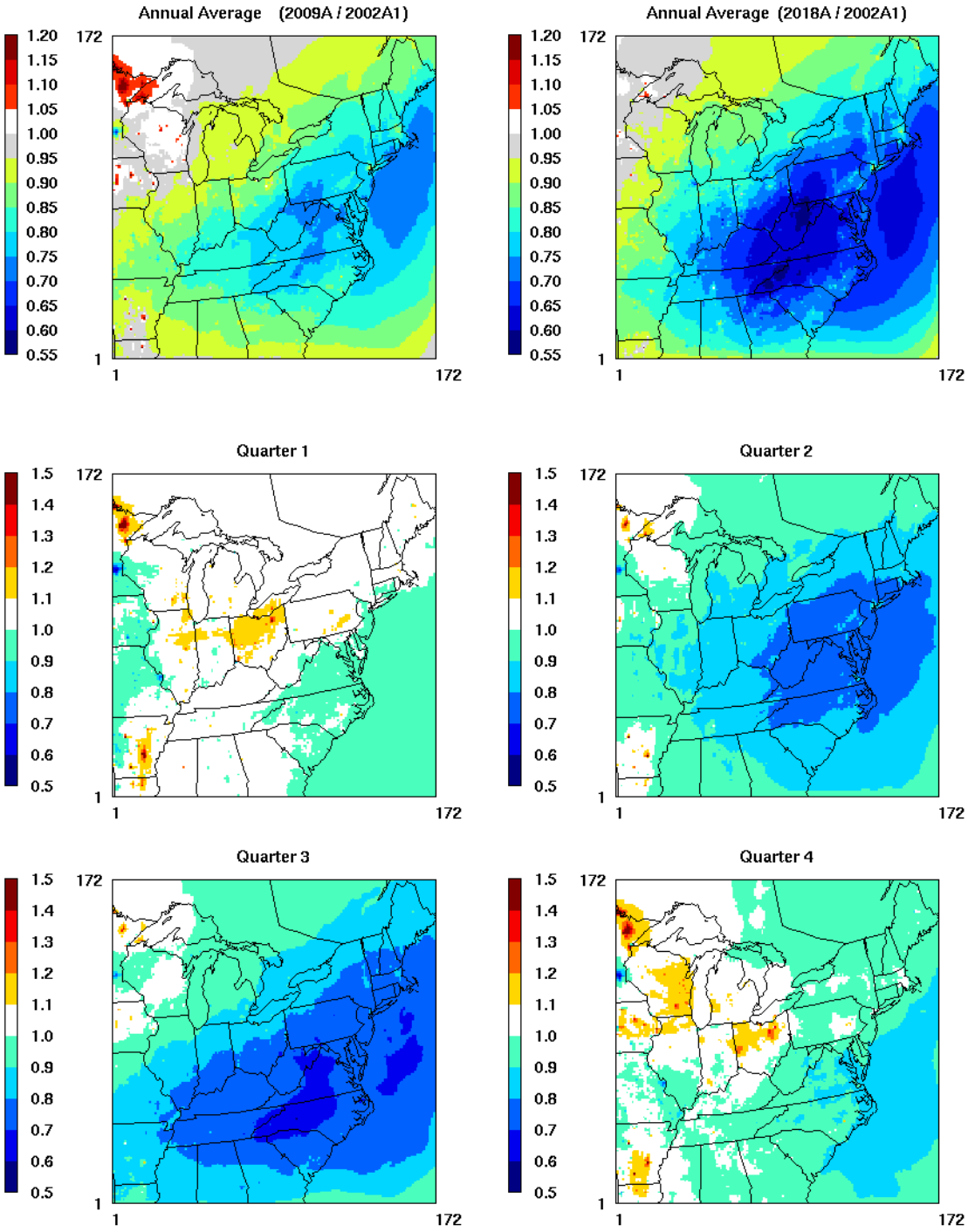


Figure C-26. Sulfate Relative Reduction Factors for 2009 and 2018. Quarterly factors are shown for 2009 in the bottom four panels



Relative reduction factors for all haze relevant PM constituents at seven Class 1 sites are displayed in Table C-1. For each site and specie, the modeled change relative to the baseline year 2002 is shown. Therefore, negative values imply a modeled decrease while a positive value represents an increase. The values for 2009 and 2018 are additive,

such that when added, the sum corresponds to the overall change from the baseline year to 2018. Using Lye Brook as an example, the table reveals a 28% decrease in concentrations from 2002 to 2009, followed by another 10% decline (relative to 2002) between 2009 and 2018, yielding a total reduction of 38% between 2002 and 2018. For fine soil, levels increase 17% in the first modeled period, then decline by 4% in the second period. The net change between 2002 and 2018 sums these two values (17 + (-4) = 13), implying an overall rise in fine soil concentrations of 13 percent. As a point of interest, the model results do not provide evidence of nitrate replacement at these sites between the period of 2002 and 2018, despite the substantial reductions in predicted ambient sulfate levels.

The results for modeled sulfate in Figure C-27 show the application of the reduction factors from Table C-1 to the baseline measured ammonium sulfate on the 20% worst days. The yellow bar gives the five-year average sulfate levels on the worst days for each site. Using the reduction factors for 2009 yields sulfate concentrations shown at the red bar, while applying the 2018 factor predicts mass values given by the blue bar. These bars clearly indicate more substantial reductions in sulfate levels are expected to occur by 2009 with smaller reduction in the latter modeled timeframe.

The final set of CMAQ results are graphed in Figure C-28(a) and (b). These plot the modeled progress combining all six species' reduction factors. Based on the modeling, all sites except one are shown to meet their uniform progress goal by 2018. Brigantine Wilderness Area in New Jersey is projected to fall about a half deciview shy of the uniform rate under existing emission reduction plans.

Table C-1. Relative Reduction factors by site and specie. Change is relative to baseline modeled year 2002 and overall change from 2002 to 2018 is additive

	YEAR	Sulfate	Nitrate	Organic Carbon	Elemental Carbon	Fine Soil	Coarse
Acadia	2009	-31%	0%	-7%	-19%	5%	6%
	2018	-7%	-5%	-6%	-17%	-1%	8%
Brigantine	2009	-29%	-1%	-8%	-23%	13%	11%
	2018	-10%	-11%	-9%	-20%	-2%	6%
Great Gulf	2009	-24%	-3%	-5%	-15%	16%	15%
	2018	-9%	-2%	-8%	-16%	-4%	7%
Lye Brook	2009	-28%	4%	0%	-16%	17%	10%
	2018	-10%	-3%	-8%	-19%	-4%	5%
Moosehorn	2009	-27%	-2%	-3%	-13%	9%	6%
	2018	-6%	-4%	-5%	-14%	-1%	6%
Dolly Sods	2009	-33%	-15%	4%	-10%	29%	34%
	2018	-16%	-11%	-11%	-22%	0%	11%
Shenandoah	2009	-29%	-24%	2%	-13%	23%	15%
	2018	-14%	-17%	-16%	-29%	-2%	8%

Figure C-27. Ammonium Sulfate mass predicted reduction for 20% worst days

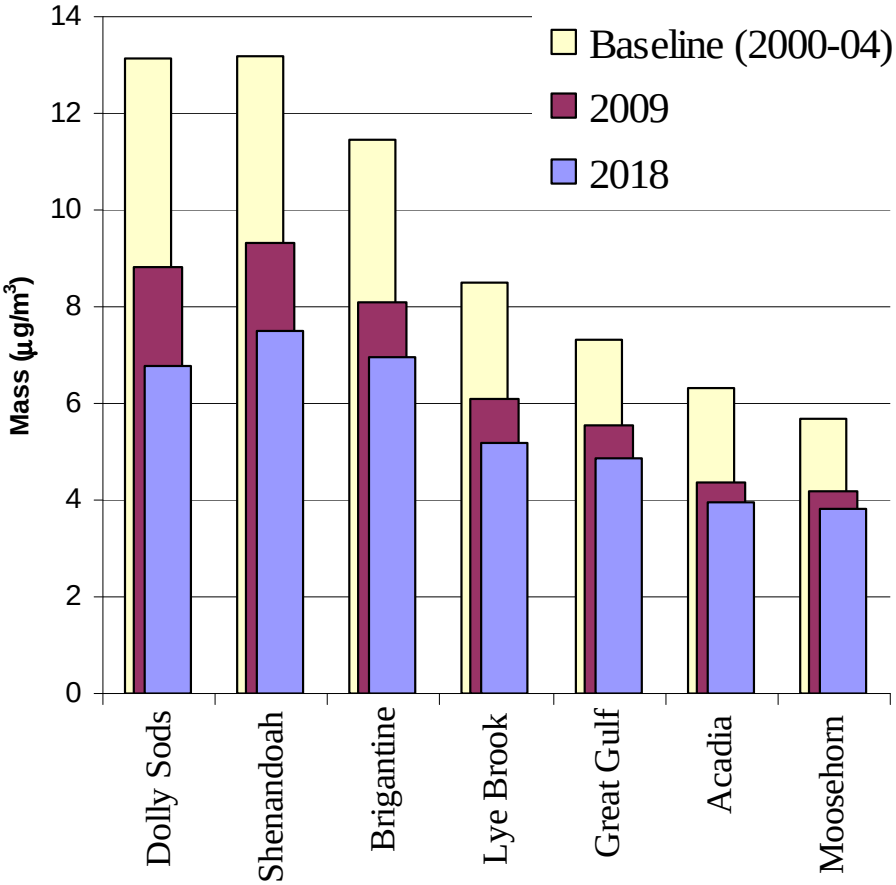
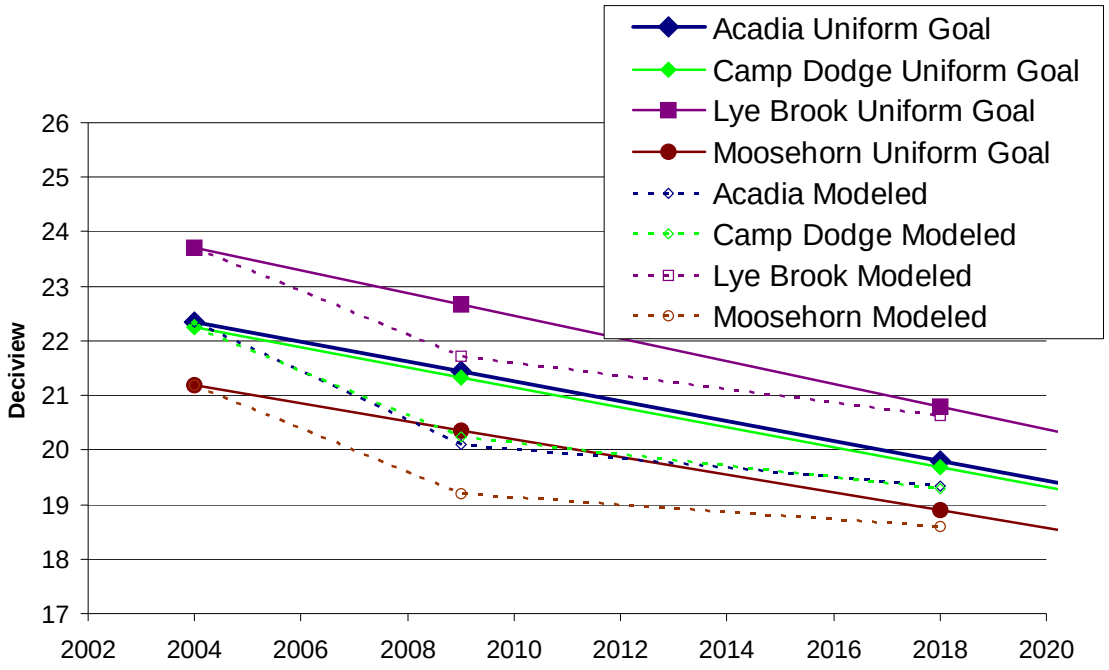
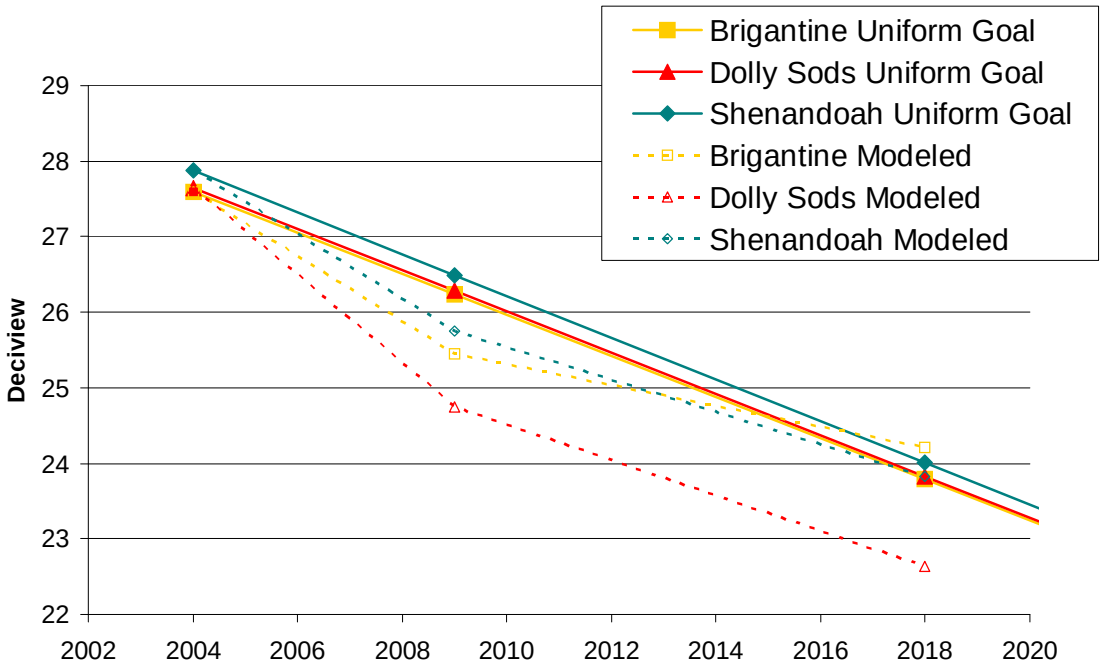


Figure C-28 (a) and (b). CMAQ Integrated SIP Modeling Platform simulation results for 2002, 2009 and 2018 relative to Uniform Progress Goals calculated according to current USEPA Guidance for (a) Northeast Class I sites in MANE-VU and (b) Mid-Atlantic Class I sites in or near MANE-VU.

(a)



(b)



C.3.2. REMSAD

C.3.2.1. Model performance

REMSAD has been evaluated by EPA OAQPS for their CSA base case study using 1996 meteorology and 1996 NET Inventory.[4] Modeling results were compared with IMPROVE measurement as summarized in Table C-2. It shows that REMSAD performs better in the Eastern US than in the Western US on PM sulfate and $PM_{2.5}$, although it underestimates ambient levels countrywide. Emissions may contribute to poor performance on soil, carbonaceous aerosols and PM nitrate.

Table C-2. Normalized error of annual mean model prediction to annual mean observation on PM species between IMPROVE measurements and REMSAD 1996 annual simulation (after Timin, B. et al., 2002)

IMPROVE PM Species	National	East	West
$PM_{2.5}$	-32%	-15%	-49%
Sulfate	-19%	-10%	-39%
Nitrate	5%	82%	-55%
Elemental Carbon	1%	23%	-20%
Organic Aerosols	-45%	-42%	-47%
Soil/Other	38%	225%	-18%

NESCAUM's previous REMSAD modeling exercise used 1996 meteorology along with the 2001 Proxy emission inventory, thus a direct comparison of modeling results to daily observations could not be completed. To evaluate REMSAD in that stage, NESCAUM first compared its own modeling results with EPA's CSA 2001 case modeling results, which also used 1996 meteorology. As shown in Figure C-29, NESCAUM's results exactly match with EPA's REMSAD modeling on $PM_{2.5}$ and PM sulfate distributions. In addition, NESCAUM compared the long term modeling average (annual mean) of PM species to IMPROVE annual means[5] for three sites. The results are presented in Figure C-30. It shows good agreement for REMSAD modeling of PM sulfate, NH_4, OC and EC. Emission inaccuracies may explain the model over-prediction of soil mass, while incomplete chemistry may cause observed differences for nitrate.

NESCAUM's present REMSAD modeling uses a 12km Eastern domain with 2002 RPO emissions and meteorology. Figure C-31 and Figure C-32 show the gridded SO_2 emissions with tags in our 12km modeling domain and some examples of annual average REMSAD sulfate concentrations by selected Northeast States, respectively. Figure C-32 illustrates the spatial distribution of the REMSAD simulated tagged emissions concentration fields. These fields are strongest in their own state and generally have the largest outside state impact toward the northeast.

[4] Also see Clear Skies Act Air Quality Modeling Technical Support Document at: http://www.epa.gov/air/clearskies/aq_modeling_tsd_csa2003.pdf

[5] Multi-year averages were computed from the measurements to better account for the lack of correspondence between emissions year (2001) and meteorological year (1996).

Figure C-29. Comparison of annual average PM2.5 and PM sulfate between NESCAUM REMSAD modeling and EPA REMSAD modeling

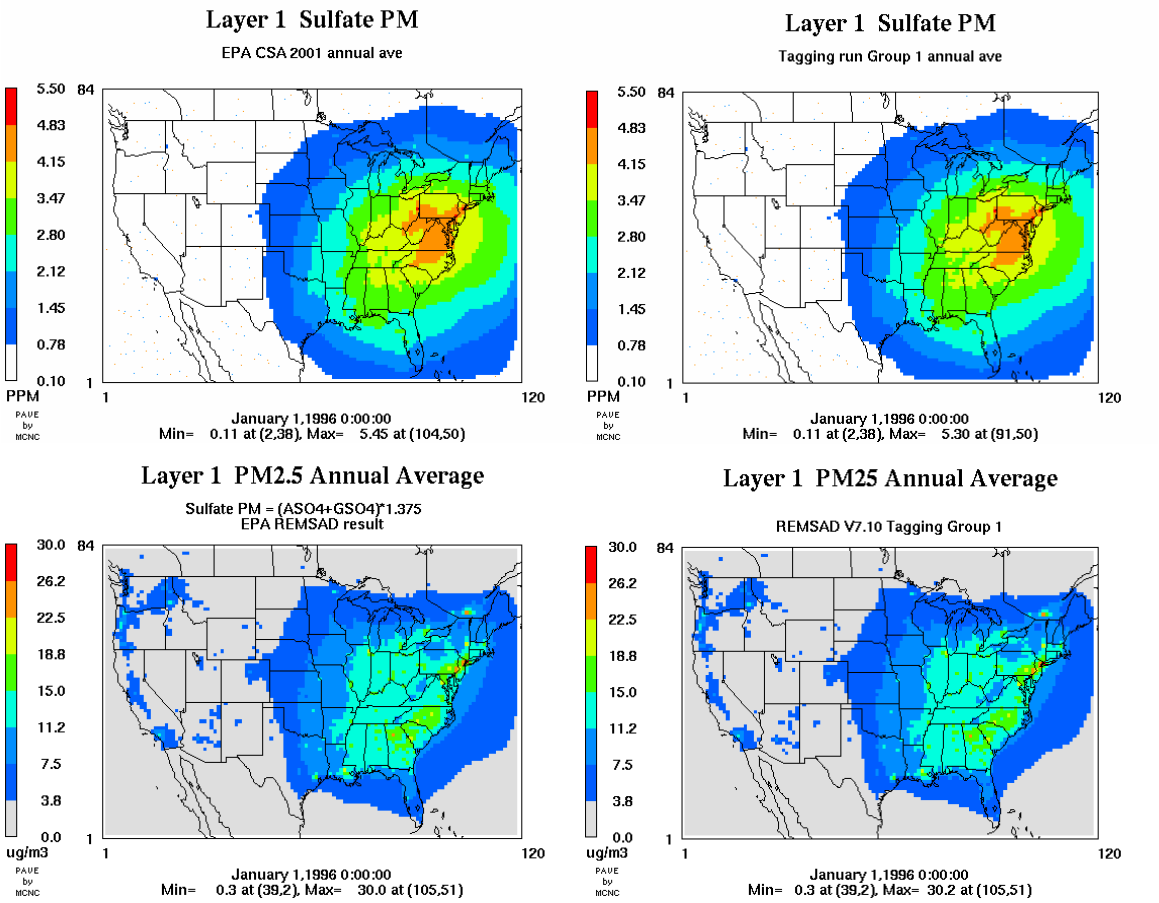


Figure C-30. Comparison of annual average PM species between NESCAUM REMSAD modeling and multi-year average IMPROVE measurements

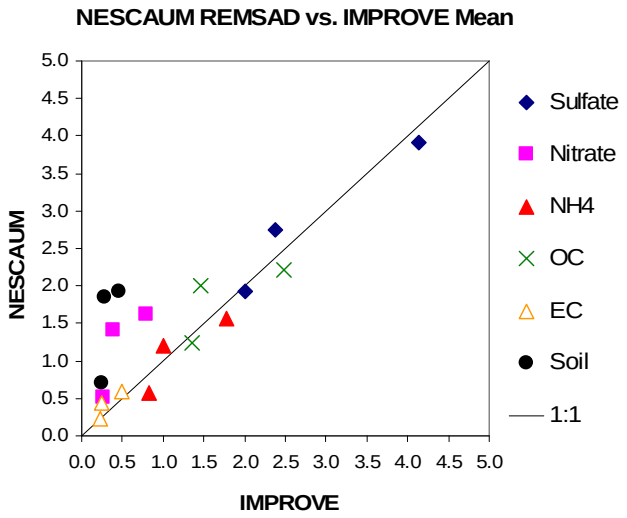


Figure C-31. Gridded SO_2 emissions distribution and tag numbers

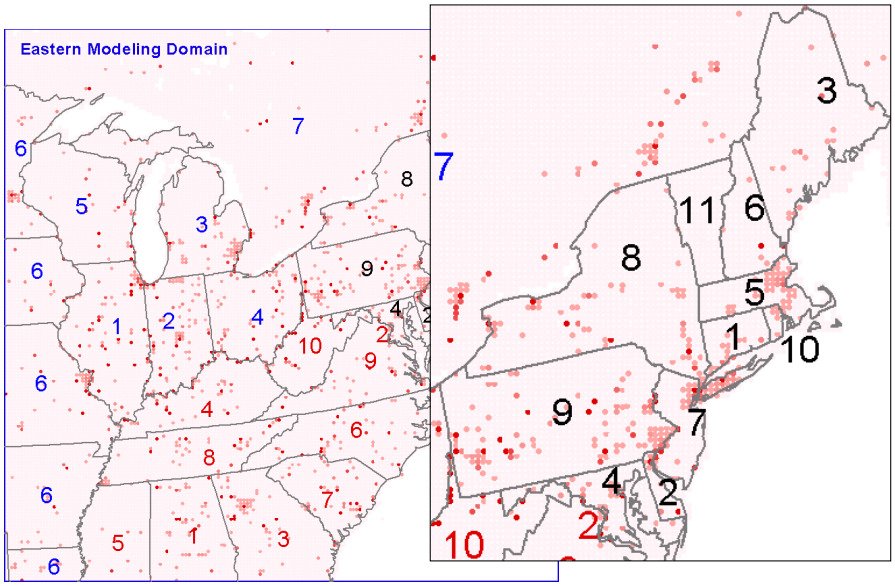
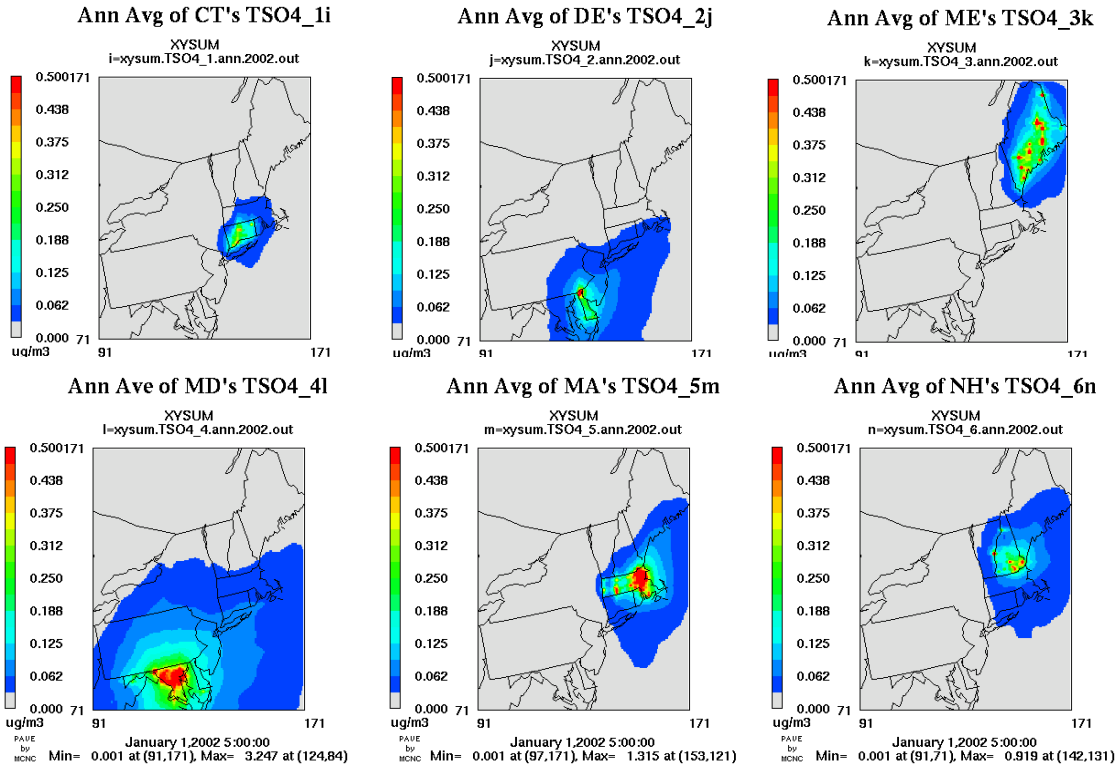


Figure C-32. Sample sulfate concentration by state



A spatial performance evaluation of REMSAD simulations for sulfate on the 12km northeast US domain for the year 2002 was conducted through comparison with IMPROVE/STN measurements, as illustrated in Figure C-33. These comparisons are inexact because the discrete measurements represent irregular areas whereas model outputs represent a uniform gridded concentration field. This approach, however, does provide a first order examination of measurement and modeling results, which is appropriate for an annual averaged analysis.

Figure C-33. Sulfate concentrations from IMPROVE/STN measurements and REMSAD model.

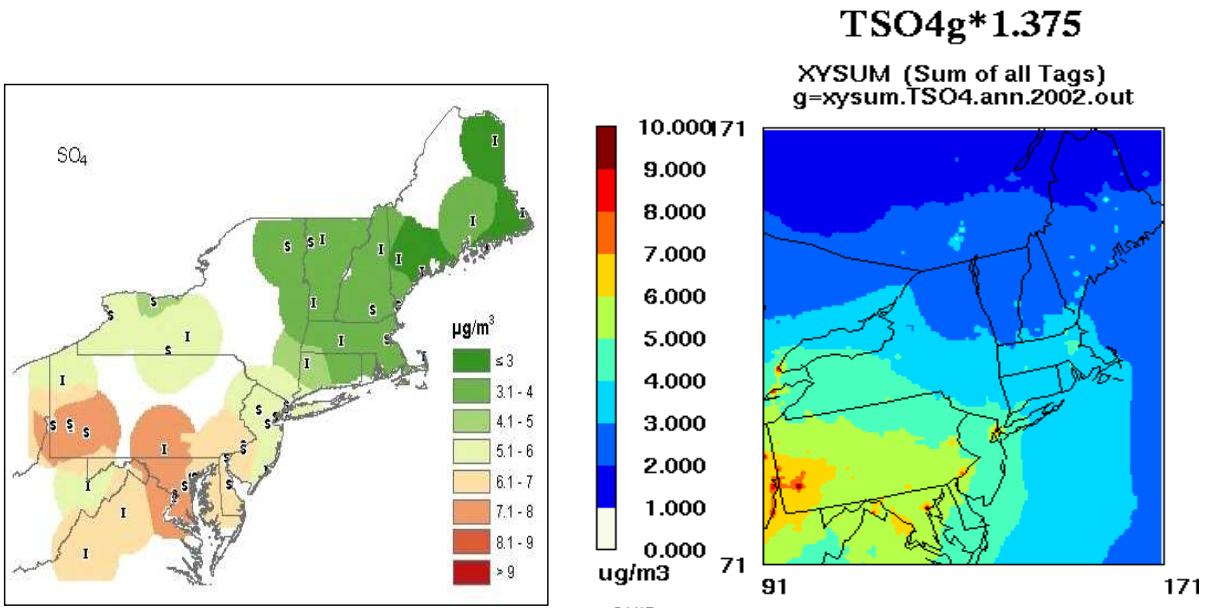
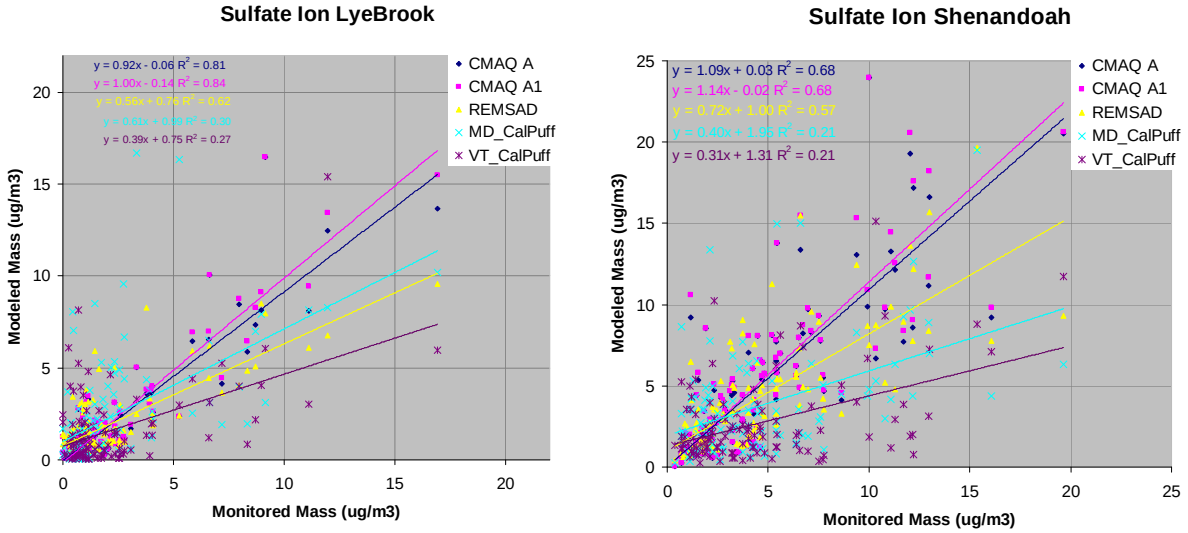


Figure C-34. Intercomparison of measurement and model data for 5 different annual model simulations.



In general, REMSAD's simulation field is well-matched with measurement data. Figure C-34 shows the comparison of paired 24-hourly surface sulfate concentrations between five different air quality model results (including REMSAD) and IMPROVE measurements during the year 2002. For Lye Brook, which is a Class I area in Vermont, the two CMAQ model runs show the best performance in terms of slope, intercept and coefficient of determination (r^2). The REMSAD result shows the 2nd best performance with the two CALPUFF results matching least to measurements. This trend remains similar for Shenandoah. Along with EPA's previous evaluation (Timin B. et al., 2002), REMSAD performs reasonably well for longer-term sulfate simulation.

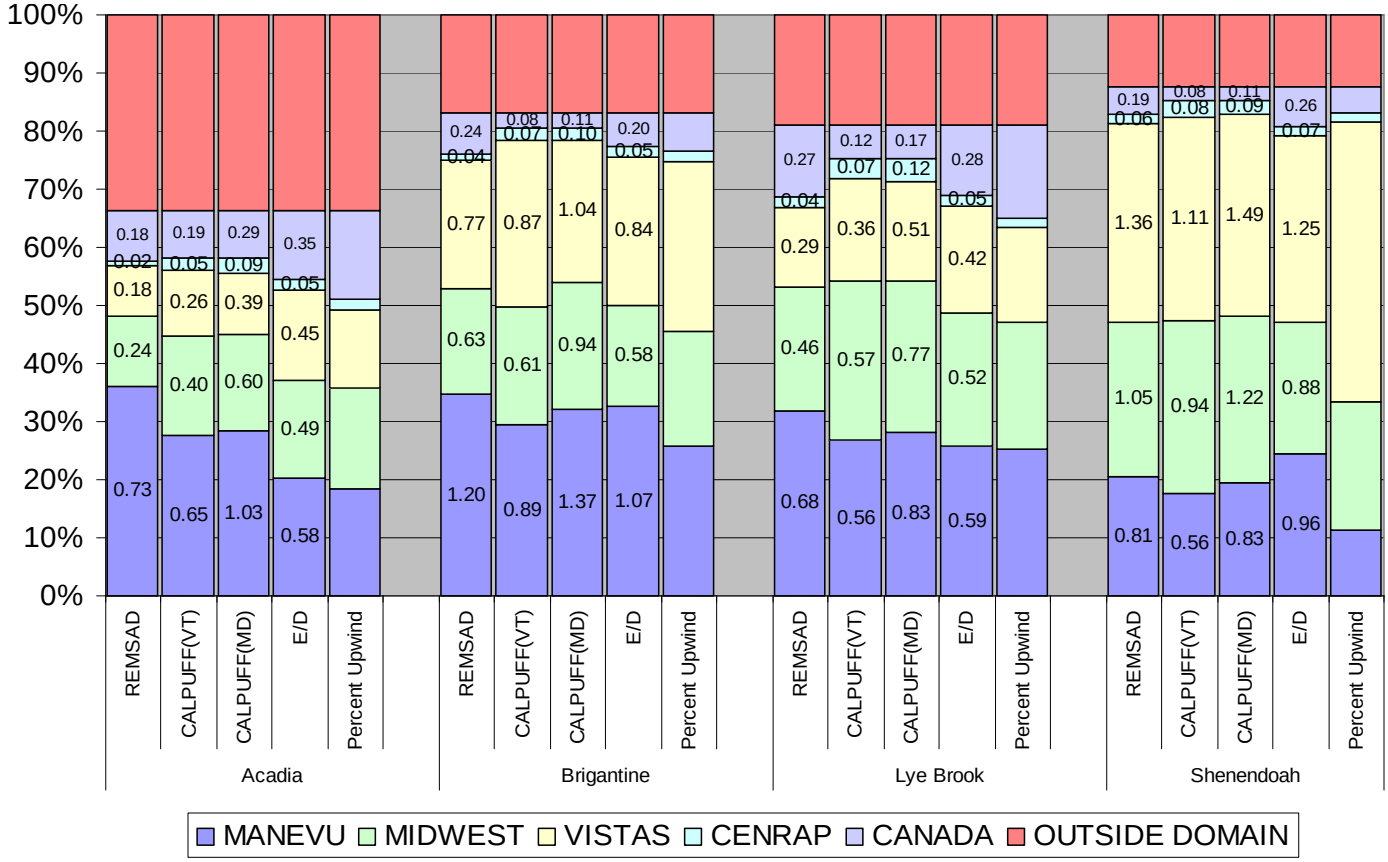
C.3.2.2. Contribution assessment

In addition to the REMSAD tagged sulfur modeling, NESCAUM and its MANE-VU partners performed other analysis techniques to assess states' impact on PM levels over the Northeast US (e.g. CALPUFF modeling, Percent-time Upwind, and E/D analysis). Figure C-35 shows modeling results of region/country specific contributions to PM sulfate in the Acadia, Brigantine, Lye Brook, and Shenendoah Class I areas from five different contribution assessment techniques. In general, the five different techniques show similar contribution of sulfate to Class I areas, but MANE-VU's contributions are estimated to be relatively higher from the REMSAD result and lower from the Percent upwind.

For Acadia, the analysis reveals about 37% of sulfate is derived from in-region sources, while 30% comes from other RPOs and Canada. The "Other" tag from REMSAD, which explains non-tagged emissions and boundary conditions (about ~33% of the total contribution to Acadia), was normalized, then included in other relative contribution analysis techniques because only REMSAD predicted those impacts. For Brigantine, about 35% of sulfate comes from in-region sources, 40% from adjacent RPO regions (i.e. MRPO and VISTAS), 10% from CENRAP (in-domain) and Canada, and the "Other" tag explains the remaining ~16 percent. The contribution to Lye Brook shows similar composition to Brigantine's case, with higher contribution of MRPO and lower contribution of VISTAS given the relative locations of sources and receptors. Shenandoah shows higher contribution from VISTAS and MRPO (about 60%) and lower contribution from MANE-VU and Others (about 20% each) due to its location (i.e. VA).

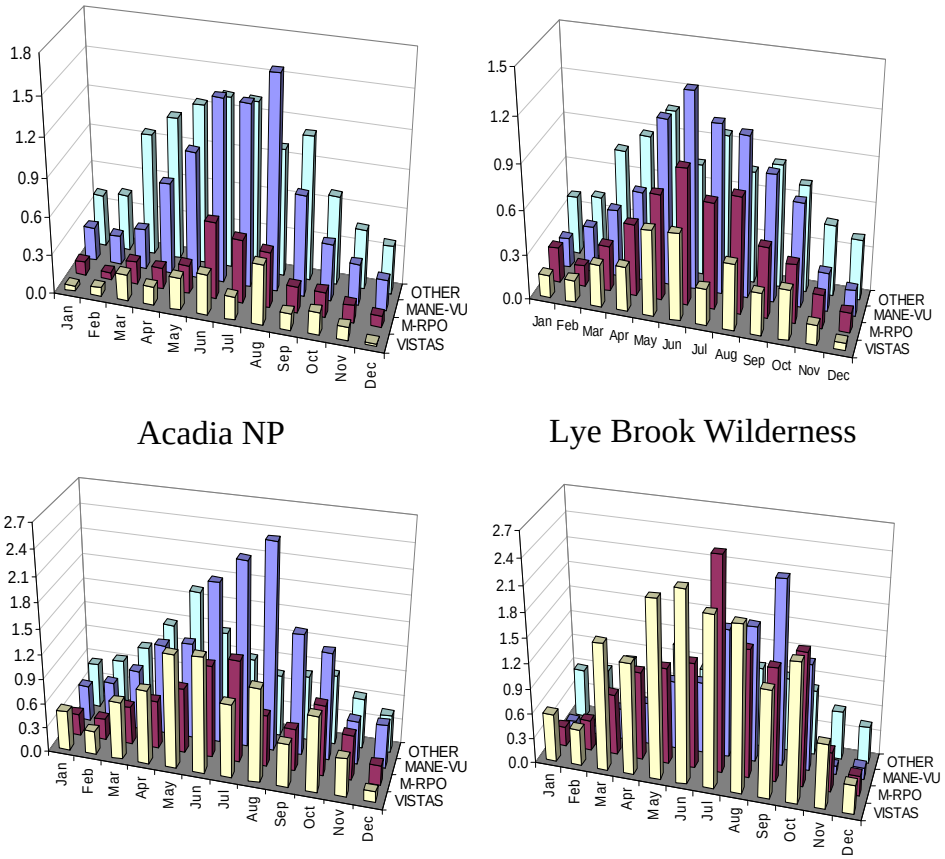
Figure C-36 shows monthly contributions by four different Class I areas in the Northeast region. The contribution of "Other" and MANE-VU region are relatively big in Acadia (in Maine) because it is located at the Northeast boundary of the modeling domain. The concentrations are generally high in the summer months (i.e. June, July, and August) for all the regions with the relative contribution of MRPO and VISTAS higher than other seasons, likely due to stronger westerly/southwesterly winds in summer. The "Other" and MANE-VU regions' contributions are still relatively big at Lye Brook, but MRPO and VISTAS' contributions are more significant than in Acadia's case because Vermont is closer to those regions. The monthly concentration shows a similar pattern to that from Acadia. For Brigantine, MANE-VU's contribution remains biggest, followed by VISTAS, "Other", and MRPO. The monthly contribution from the "Other" tag decreases after June in contrast to MRPO's. The VISTAS and MRPO's contributions are relatively large at Shenandoah due to their proximity.

Figure C-35. Contribution of tagged sources by different apportionment methods



Given the reassuring results for sulfate, NESCAUM conducted REMSAD tagged sulfur modeling to assess states' impact on PM levels over the Northeast US. Figure C-37 shows modeling results of state specific contributions to PM sulfate in the Acadia, ME Class I area. Similar plots are shown for Brigantine, Lye Brook and Shenandoah (Figure C-38, Figure C-39, and Figure C-40 respectively). For Acadia, "Other", MA, Canada, PA, ME, OH, and NY contribute more than 75% of sulfate. The higher contribution of "Other", MA, and Canada are explained by their relative location - Acadia National Park is located at the northeast boundary of our 12km modeling domain. In the case of Brigantine, PA, OH, and NY's contributions are relatively higher compared to Acadia's case because Brigantine is located nearer to those high emission states. In general, Lye Brook shows similar distribution to Brigantine, except that it shows a little higher contribution from Canada. Other than the "Other" tag, OH, PA, WV, VA, and IN show bigger contributions to Shenandoah, with Canada's contribution relatively smaller compared to other Class I areas, as it is located further south. In Shenandoah's case, the fraction of non-tagged emissions (e.g. biomass burning in VISTAS states - as opposed to boundary conditions) explains the higher contribution of "Other" tag even with smaller contribution of Canada.

Figure C-36. Contribution of tagged sources for different Class-I areas in Northeast (monthly average sulfate concentration).



Acadia NP

Lye Brook Wilderness

Brigantine NWR

Shenandoah NP

Figure C-37. Eastern states' contribution to annual PM sulfate in Acadia, ME

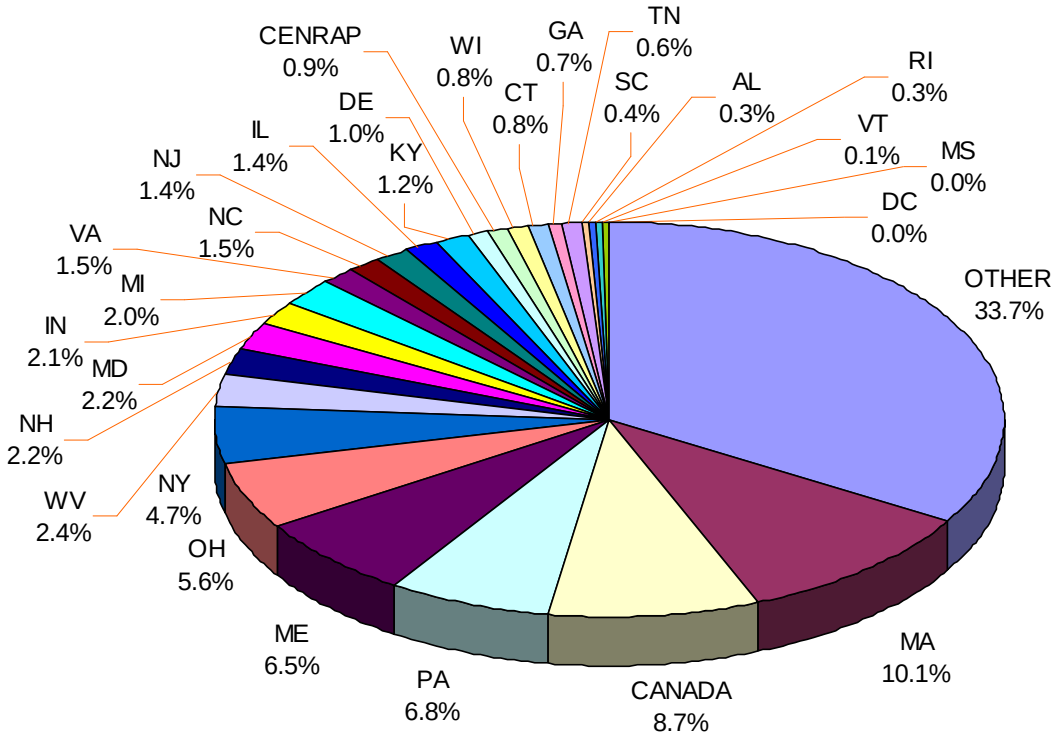


Figure C-38. Eastern states' contribution to annual PM sulfate in Brigantine, NJ

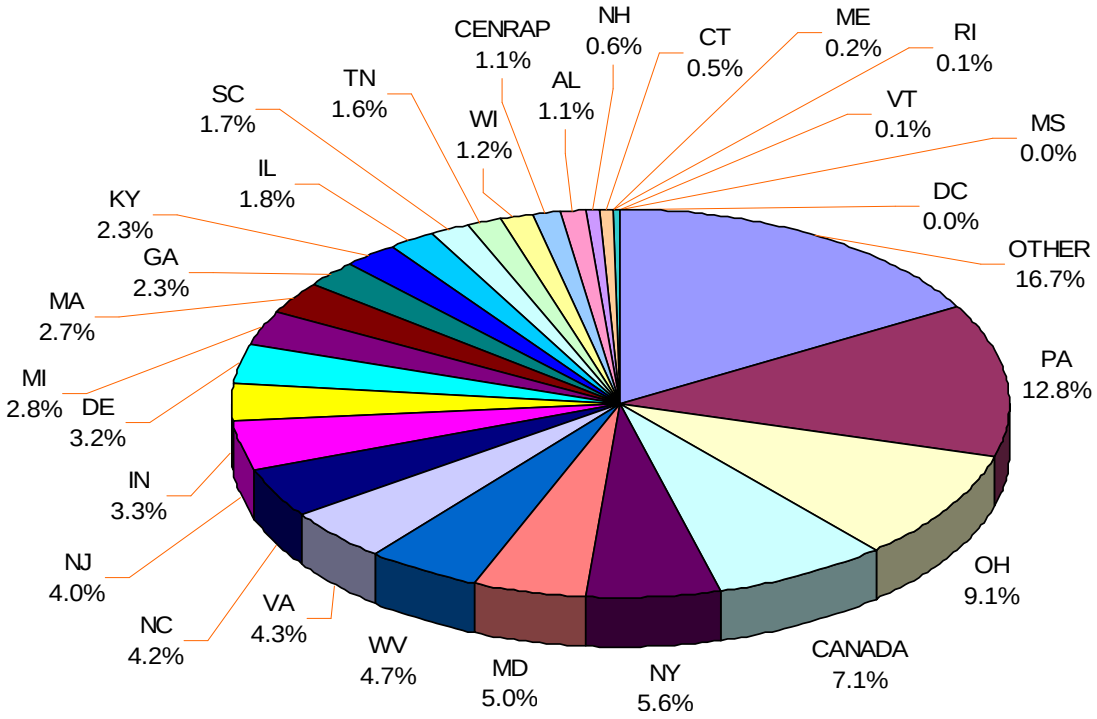


Figure C-39. Eastern states' contribution to annual PM sulfate in Lye Brook, VT

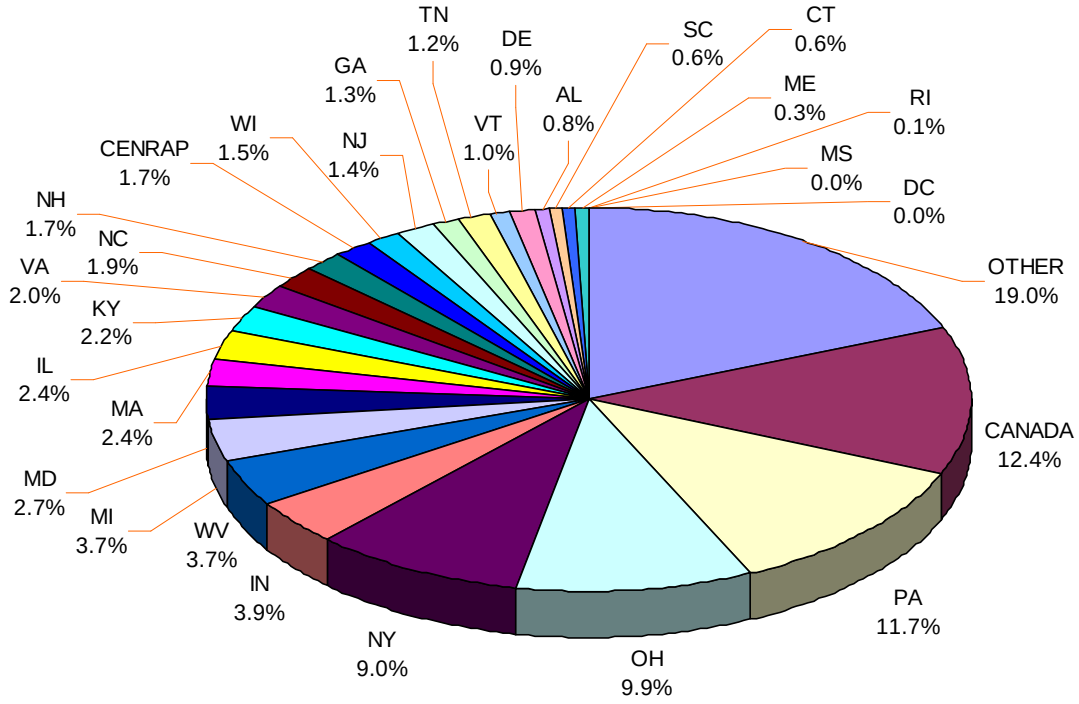
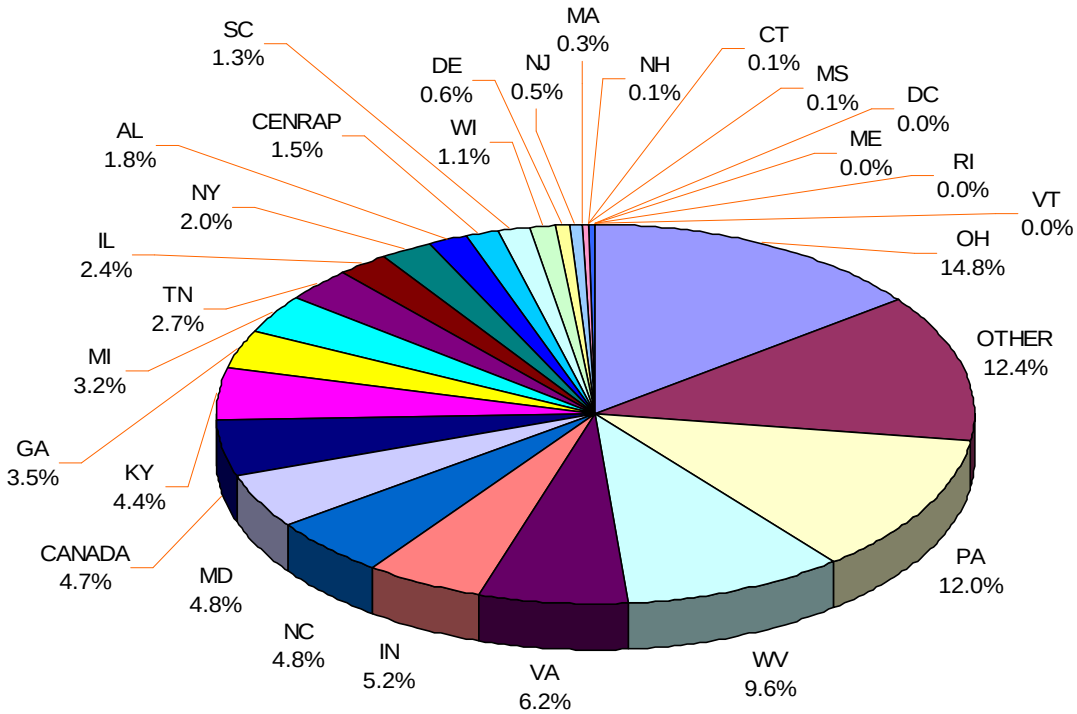


Figure C-40. Eastern states' contribution to annual PM sulfate in Shenandoah, VA



Also, the 20% worst and best visibility days results were used to evaluate contributions to PM sulfate from emission sources in each RPO region, as shown in Figure C-41 and Figure C-42. Unlike previous presentations, three more Class I areas are added (e.g. Dolly Sods Wilderness, Great Gulf Wilderness, and Moosehorn Wilderness) for analysis and all sites are arranged from the southwest to northeast. As shown in the Figure C-41, each site tends to show the greatest contribution to poor visibility from nearby regions. This tendency reveals the atmospheric transport impact that adjacent regions' strong and fresh emissions have at nearby receptor sites. The 20% best visibility days (Figure C-42) seem to occur when the contribution from boundaries are bigger unlike the behavior observed for worst case days. In comparing the annual average impact LYBR, BRIG, and SHEN show similar contribution patterns whereas Acadia shows higher contribution (45% vs. 37%) of MANE-VU on the 20% worst visibility days.

Statistical parameters used in model performance evaluation. P_i and O_i are paired model prediction and observation, respectively. MB, ME, and RMSE have the same units as P_i and O_i, while other parameters have units of percent.

Mean Bias (***MB***) $$\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)$$

Mean Error (***ME***) $$\frac{1}{N}\sum_{i=1}^{N}|P_i - O_i|$$

Root Mean Square Error (***RMSE***) $$\left[\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{1/2}$$

Normalized Mean Bias (***NMB***) $$\frac{\sum_{i=1}^{N}(P_i - O_i)}{\sum_{i=1}^{N}O_i}$$

Normalized Mean Error (***NME***) $$\frac{\sum_{i=1}^{N}|P_i - O_i|}{\sum_{i=1}^{N}O_i}$$

Mean Normalized Bias (***MNB***) $$\frac{1}{N}\sum_{i=1}^{N}\frac{(P_i - O_i)}{O_i}$$

Mean Normalized Error (***MNE***) $$\frac{1}{N}\sum_{i=1}^{N}\frac{|P_i - O_i|}{O_i}$$

Fractional Bias (***FB***) $$\frac{2}{N}\sum_{i=1}^{N}\left(\frac{P_i - O_i}{P_i + O_i}\right)$$

Fractional Error (***FE***) $$\frac{2}{N}\sum_{i=1}^{N}\left|\frac{P_i - O_i}{P_i + O_i}\right|$$

Figure C-41. Comparison of Sulfate Extinctions on 20% Worst Visibility Days

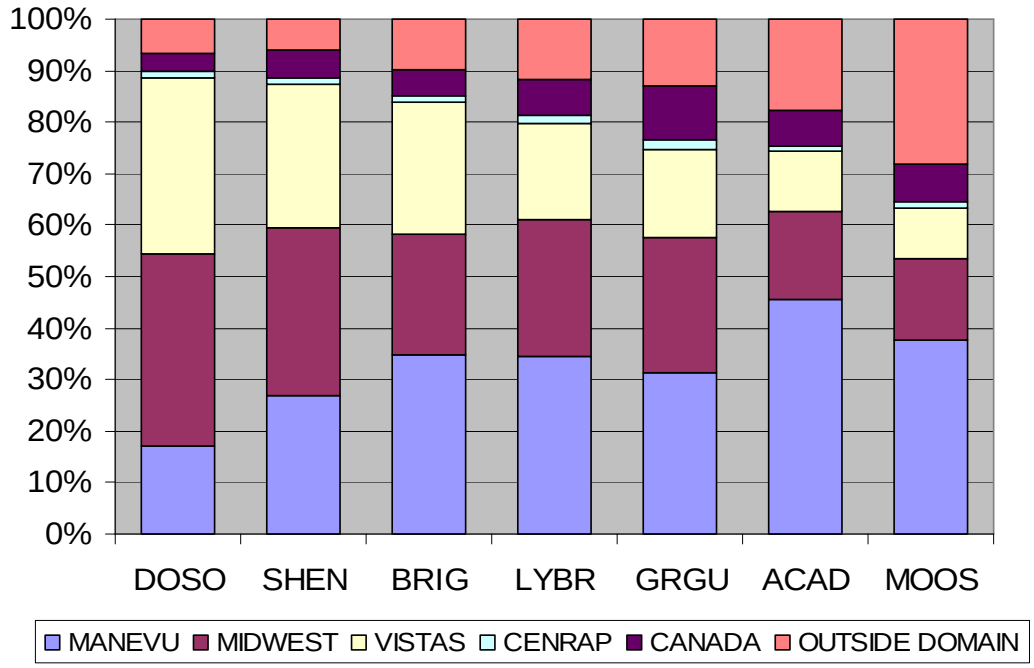
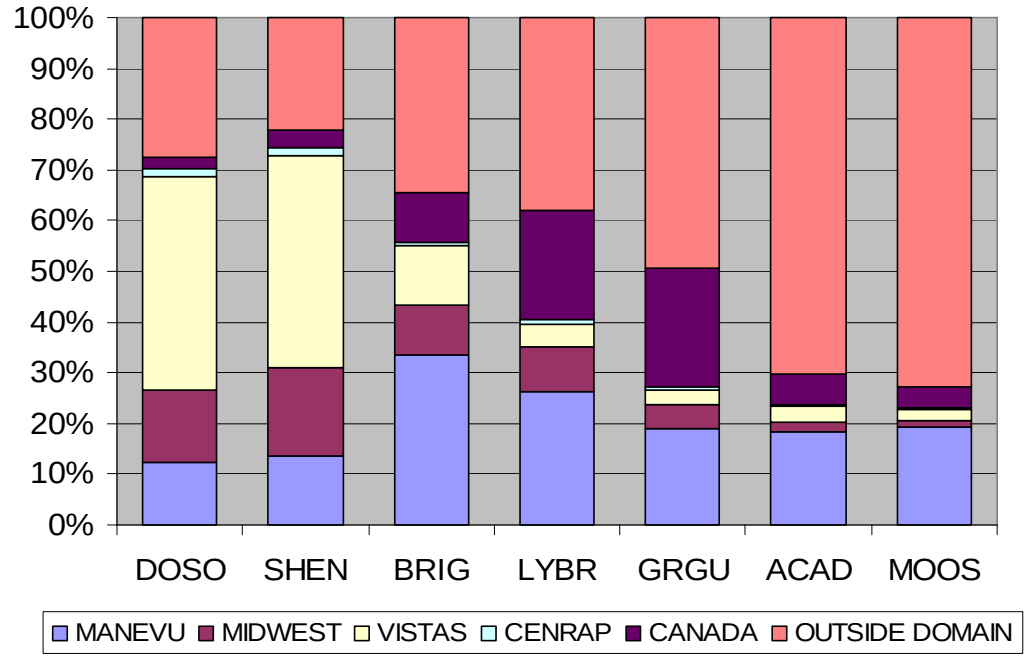


Figure C-42. Comparison of Sulfate Extinctions on 20% Best Visibility Days



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REMSAD website http://www.remsad.com