APPENDIX G

Evaluation of the 2002 GEOS-CHEM Global Chemistry Model Used for Boundary Conditions in the VISTAS Regional Haze Modeling

Application of GEOS CHEM for 2002 Boundary Conditions for CMAQ Modeling of the VISTAS Region

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CMAQ modeling of regional haze in the VISTAS region for 2002 and 2018 was carried out on a grid of 12x12 km cells that covers the VISTAS states (AL, GA, FL, MS, SC, NC, TN, KY, VA, WV) and states adjacent to them. This grid is nested within a larger national CMAQ modeling grid of 36x36 km grid cells that covers the continental United States, portions of Canada and Mexico, and portions of the Atlantic and Pacific Oceans along the east and west coasts. Transport of pollution from the national domain into the VISTAS modeling domain is reflected in hourly-varying boundary conditions around the VISTAS domain, which are derived from the CMAQ simulations in the 36 km national domain. The national domain incorporates emissions from northern Mexico and southern Canada, and therefore explicitly calculates the effects of these international emissions on concentrations at the boundaries of the VISTAS modeling domain.

Effects of emissions from more distant sources that are outside of the 36 km national domain were estimated using outputs from a global air pollution model. The Big Bend Regional Aerosol and Visibility Observational (BRAVO) study had found that the performance of national-scale modeling was enhanced by using monthly averages of outputs from a global air pollution model, GOCART, to generate SO_2 and sulfate boundary conditions for its 36 km national modeling grid (Schichtel, et al., 2005), instead of using the EPA default concentrations that are normally used. VISTAS elected to use a similar approach, using the outputs of the GEOS-CHEM global air pollution model, which also contains a detailed representation of ozone-NO_x-VOC-PM chemistry.

Under EPRI sponsorship, Harvard University had already used the GEOS-CHEM model, with 2°x2.5° horizontal resolution, to estimate natural conditions and transboundary influences on elemental and organic carbon aerosols in the United States in 1998 (Park et al., 2003). After some emission inventory and model improvements, similar information was developed for sulfate and nitrate aerosols in 2001 (Park et al., 2004).

Under contract to VISTAS (which was acting on behalf of all the RPOs), Harvard then simulated global air pollution for the year 2002 (the VISTAS modeling year) with 4°x5° horizontal resolution and 3-hour temporal resolution (Jacob, 2005). The coarser grid scale was considered sufficient to provide boundary conditions outside of North America, largely over the oceans. Although the VISTAS simulation was generally similar to the 2001 EPRI simulation mentioned above, the VISTAS modeling incorporated the following substantive changes in addition to the change in grid scale: (1) use of U.S. anthropogenic emissions from the EPA NEI 1999 inventory, except for ammonia; (2) use of forest fire information specific to 2002; (3) inclusion of the secondary organic aerosol (SOA) formation mechanism from Chung and Seinfeld (2002); and (4) inclusion of prototype soil dust and sea salt simulations. An additional model improvement, which has since been implemented in the standard version of GEOS-CHEM, was the application of surface emissions and dry deposition to the entire mixed layer column diagnosed by GEOS

(the Global Earth Observation System of NASA) rather than to just the surface layer of the model. Details of the emissions, meteorology, model configuration, and model performance evaluation for the VISTAS simulations are described in the report to VISTAS (Jacob, 2005).

For North America, the VISTAS GEOS-CHEM modeling used monthly-average emissions, so the model results do not reflect diurnal or weekday-weekend variability in emissions, and also diminish the impacts of episodic emissions, such as the July 2002 wildfire in Ontario, Canada. The Mexican emissions were from the inventory of the BRAVO Study, while Canadian emissions for the year 2000 were used. Emissions from Caribbean islands were not included, which could impact model estimates at the Everglades.

The surface meteorological data inputs to GEOS-CHEM had 3-hr temporal resolution and the upper air meteorological inputs had 6-hr resolution, with a horizontal resolution of 1°x1° or better and 48 layers vertically.

The output of the GEOS-CHEM modeling, as delivered to VISTAS, contained concentrations of all visibility-related components of particulate matter with 3-hr temporal resolution and 4°x5° horizontal resolution over multiple layers of the atmosphere.

Even though the coarse spatial resolution degraded the performance of the model over North America, relative to results from prior modeling for other years using finer grids, the concentrations simulated by GEOS-CHEM maintained the synoptic-scale structure and did not have a continental-scale bias. At a finer scale, the concentrations simulated by GEOS-CHEM for the surface layer in 2002 were generally within a factor of two of regional-averages (~500 km scale) of measurements of the various chemical components of particulate matter in the United States and, except for organic carbon, satisfactorily simulated the seasonal variability in the observations.

In the meantime, Harvard University had also carried out new finer-grid simulations for 2001 for EPRI, using a 1°x1° grid over North America, nested in a coarser grid that covered the globe. The final version of these simulations included modifications to the modeling grid to improve delineation between domestic US emissions and those of foreign sources at the national boundaries, and revision (reduction) of the Canadian and Mexican sulfur emissions. These GEOS-CHEM 1°x1° grid scale simulations for 2001 and their results are described in the paper by Park et al. (2006). Over appropriate geographic and temporal scales, the results of these simulations for 2001were similar to those of the 2002 VISTAS simulations, which gave confidence in the VISTAS GEOS-CHEM results.

The concentration fields that resulted from the GEOS-CHEM simulations were not directly usable as boundary conditions for the 36 km grid scale CMAQ simulations. GEOS-CHEM and CMAQ use different horizontal and vertical grid scales and different map projections, and have different labels for chemical constituents. The GEOS-CHEM outputs were processed by Dr. Daewon Byun at the University of Houston to adjust for these discrepancies and to develop boundary conditions for the 36 km CMAQ modeling. Since the CMAQ modeling has temporal resolution of one hour, the boundary conditions were given for every hour. However, since the GEOS-CHEM outputs had 3-hr resolution, the hourly boundary conditions were the same for three consecutive hours. An example of the resulting boundary conditions for sulfate at the surface is shown in Figure G-1. As indicated by the colors along the boundary of the modeling

domain, GEOS-CHEM projects higher sulfate values for the southern boundary, indicating emissions from Mexico, and the eastern boundary, indicating recirculation of emissions from the eastern US.

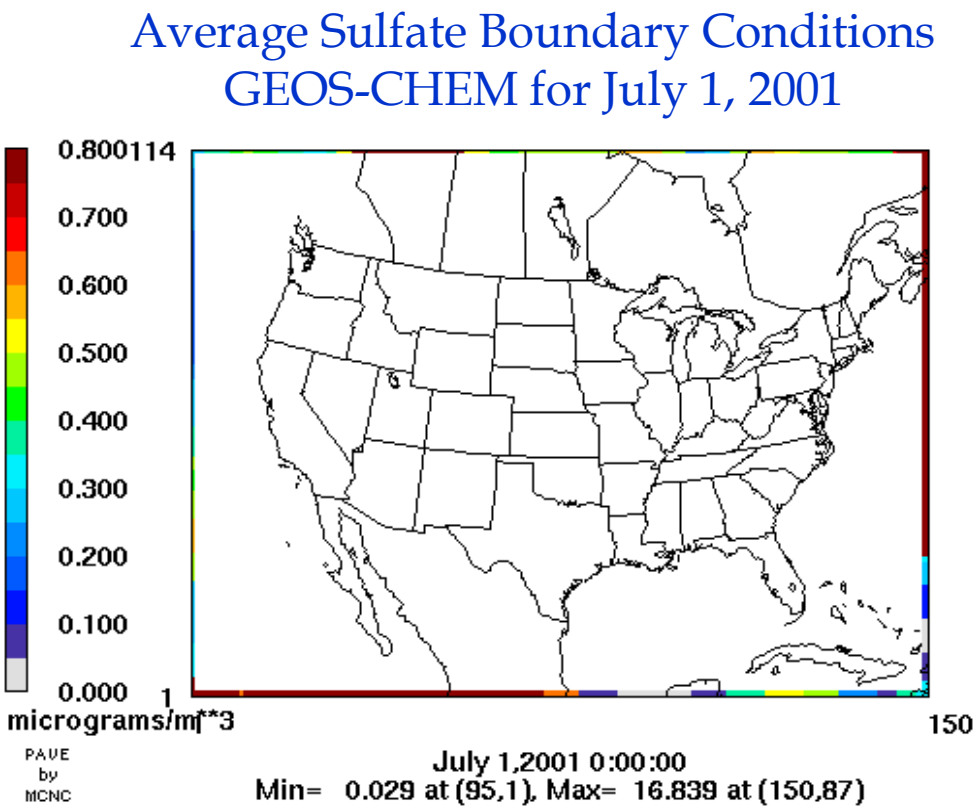

Figure G-1. GEOS-CHEM projections of sulfate boundary conditions for the national CMAQ modeling grid on July 1, 2001.

Model performance for the 2002 4x4 degree annual GEOS-CHEM run was compared to observations at 145 IMPROVE sites and CASTNET sites across the continental US. Example plots from the model performance evaluation are illustrated in Figures G-2 through G-9. Additional information is provided in the final GEOS-CHEM report (Jacobs, 2005). Performance at the boundaries is driven by accuracy of international emission estimates; performance at US continental monitors is driven by US emissions. Given the large grid size, projections are approximations that show the correct order magnitude. Performance for 2002 boundary conditions is acceptable compared to observations and an improvement over the CMAQ default single annual value for all boundaries. Sulfate projections reproduce observed values fairly well, particularly for boundary conditions. Sulfate is slightly over predicted for winter months. Summer sulfate levels are under predicted, particularly on high sulfate days, but mean fraction bias for monthly average sulfate is within performance criteria set for CMAQ (+/-30%).

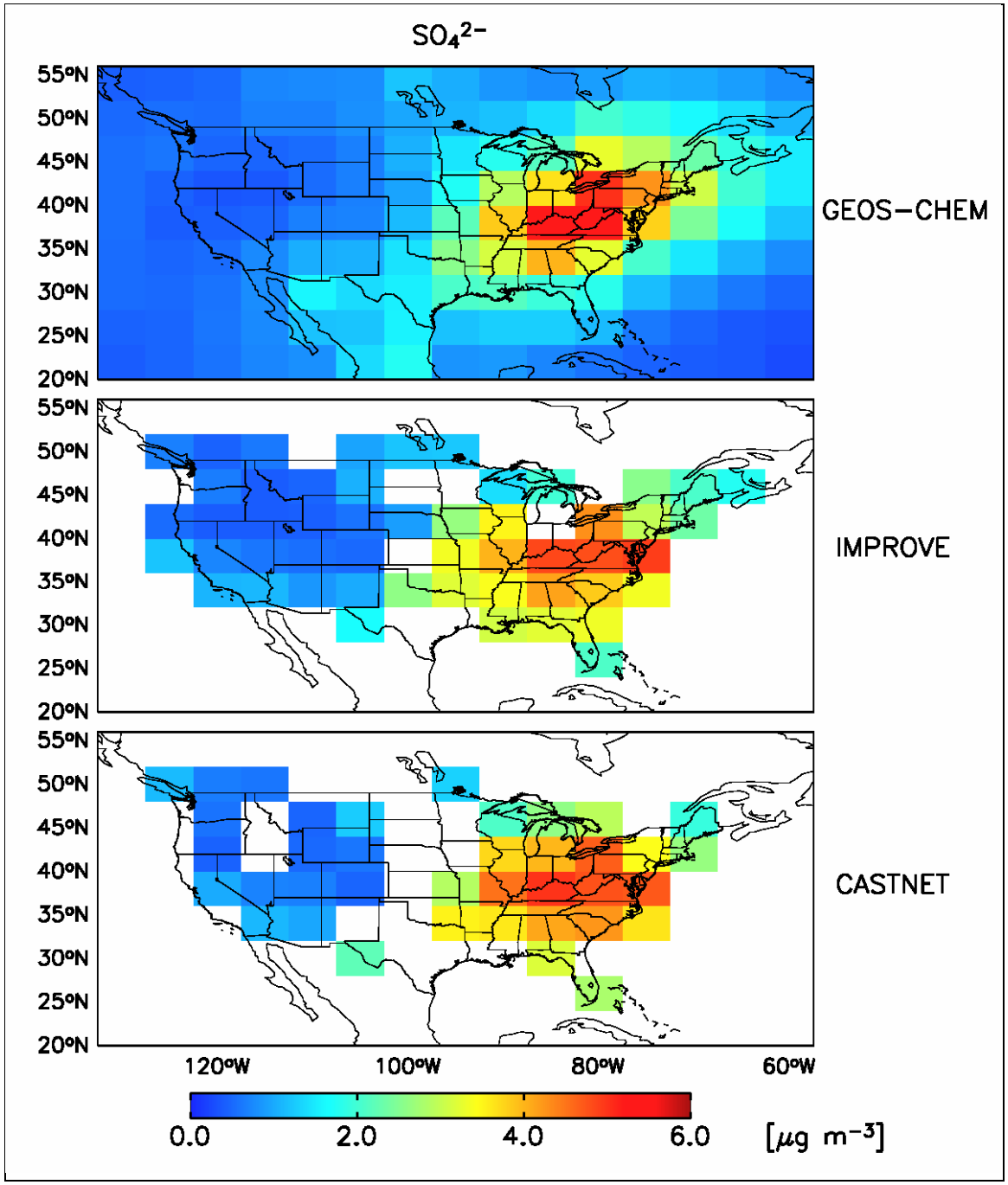

Figure G-2. GEOS-CHEM 2002 SO4 annual performance compared to IMPROVE (145 sites) and CASTNET monitoring networks.

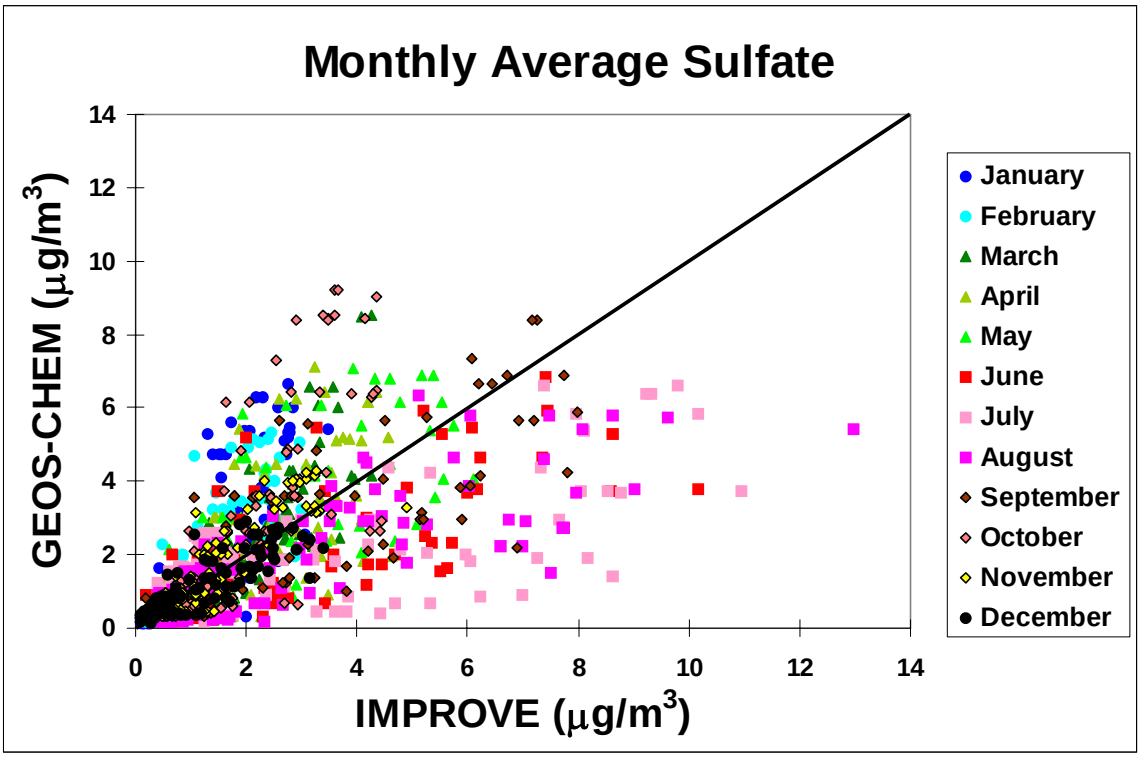

Figure G-3. GEOS-CHEM 2002 Monthly SO4 Performance compared to 145 IMPROVE sites in the continental US.

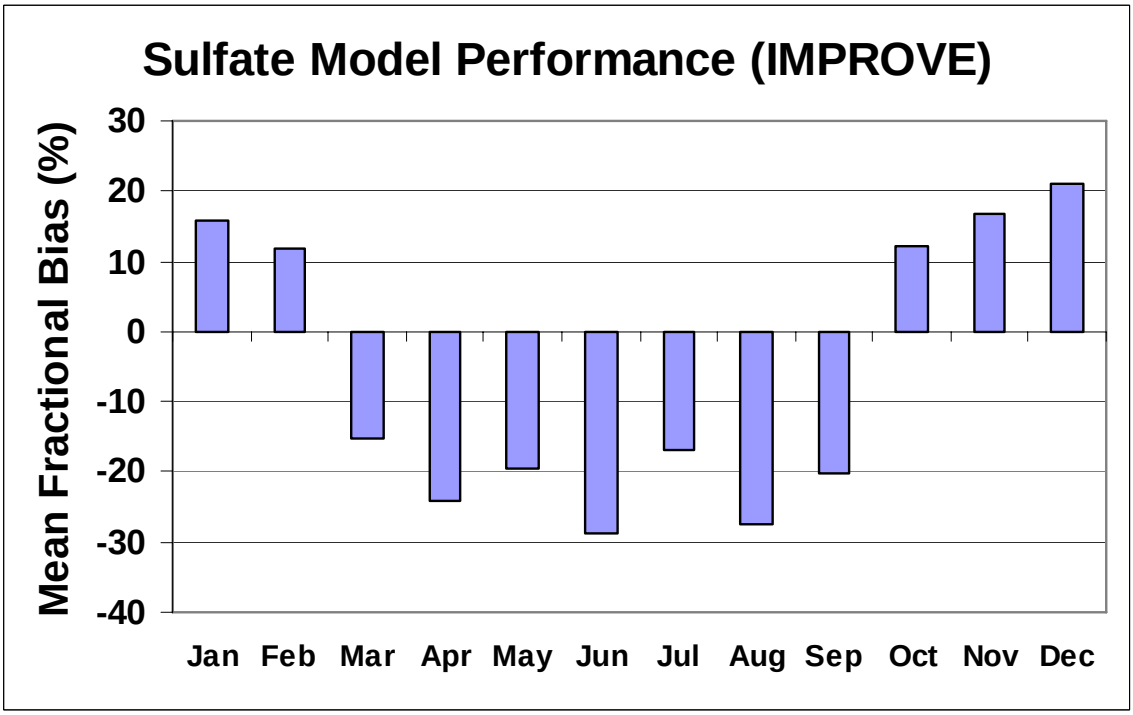

Figure G-4. GEOS-CHEM 2002 Monthly SO4 Performance compared to 145 IMPROVE sites in the continental US.

GEOS-CHEM nitrate performance is acceptable for boundary conditions. Nitrate is over predicted at some sites on some winter days; overall nitrate is under predicted in all months, and particularly in summer months. Since summer nitrate values are generally low, this under prediction does not affect overall model performance for CMAQ.

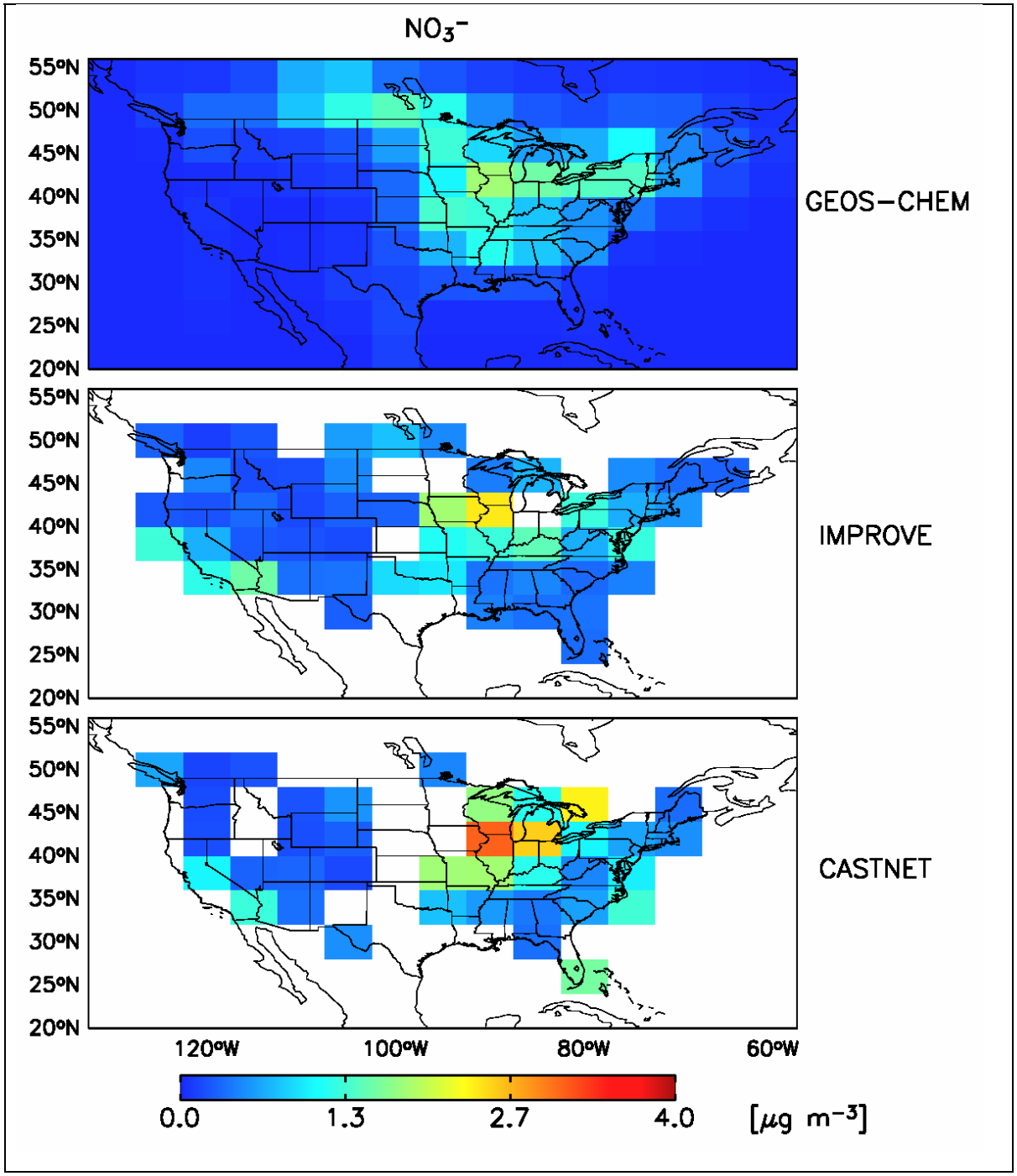

Figure G-5. GEOS-CHEM 2002 NO3 annual average performance compared to IMPROVE (145 sites) and CASTNET monitoring networks.

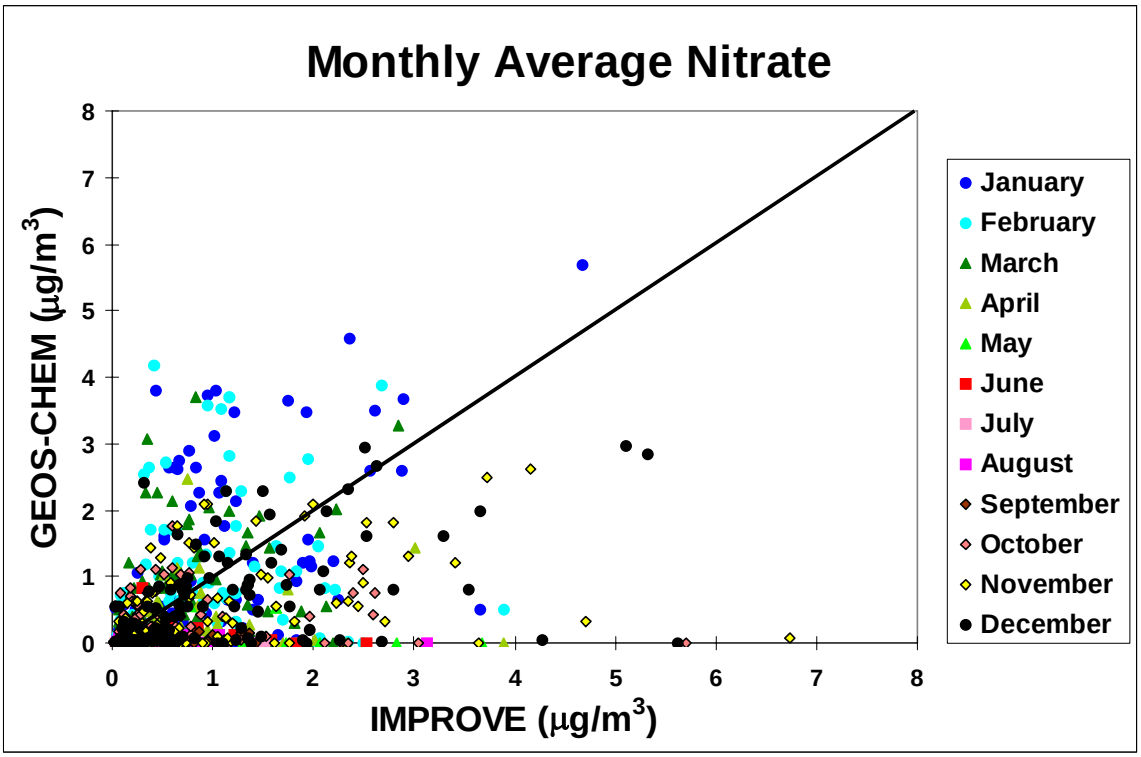

Figure G-6. GEOS-CHEM 2002 Monthly NO3 Performance compared to 145 IMPROVE sites in the continental US.

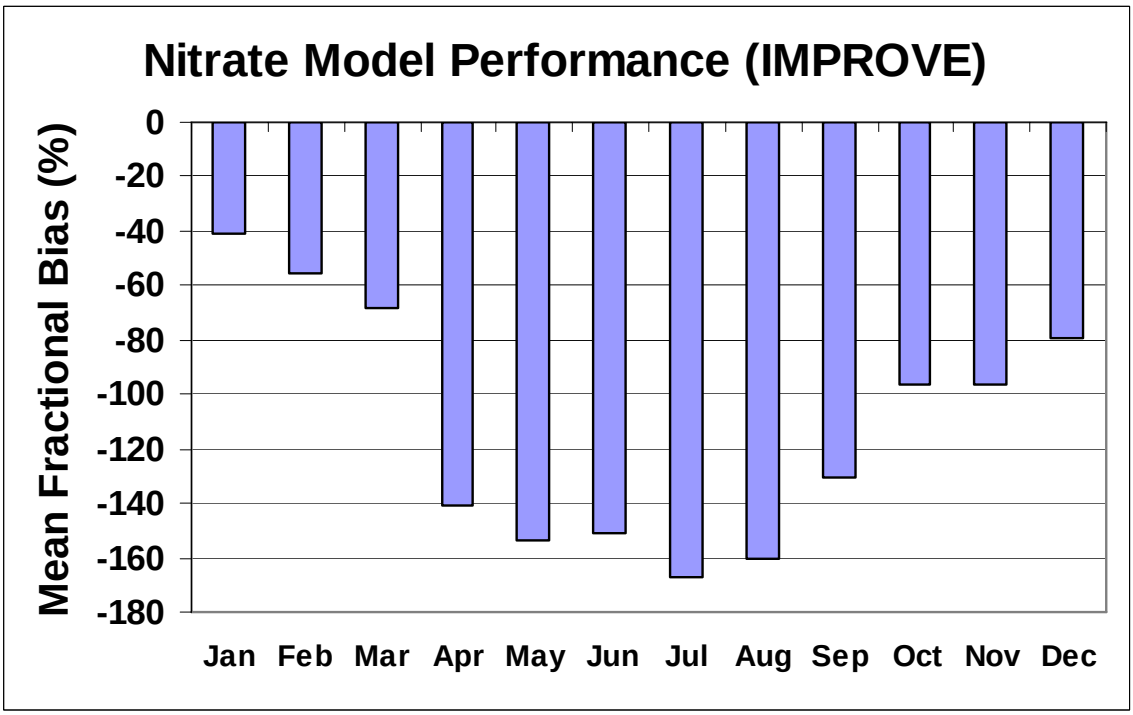

Figure G-7. GEOS-CHEM 2002 Monthly NO3 Performance compared to 145 IMPROVE sites in the continental US.

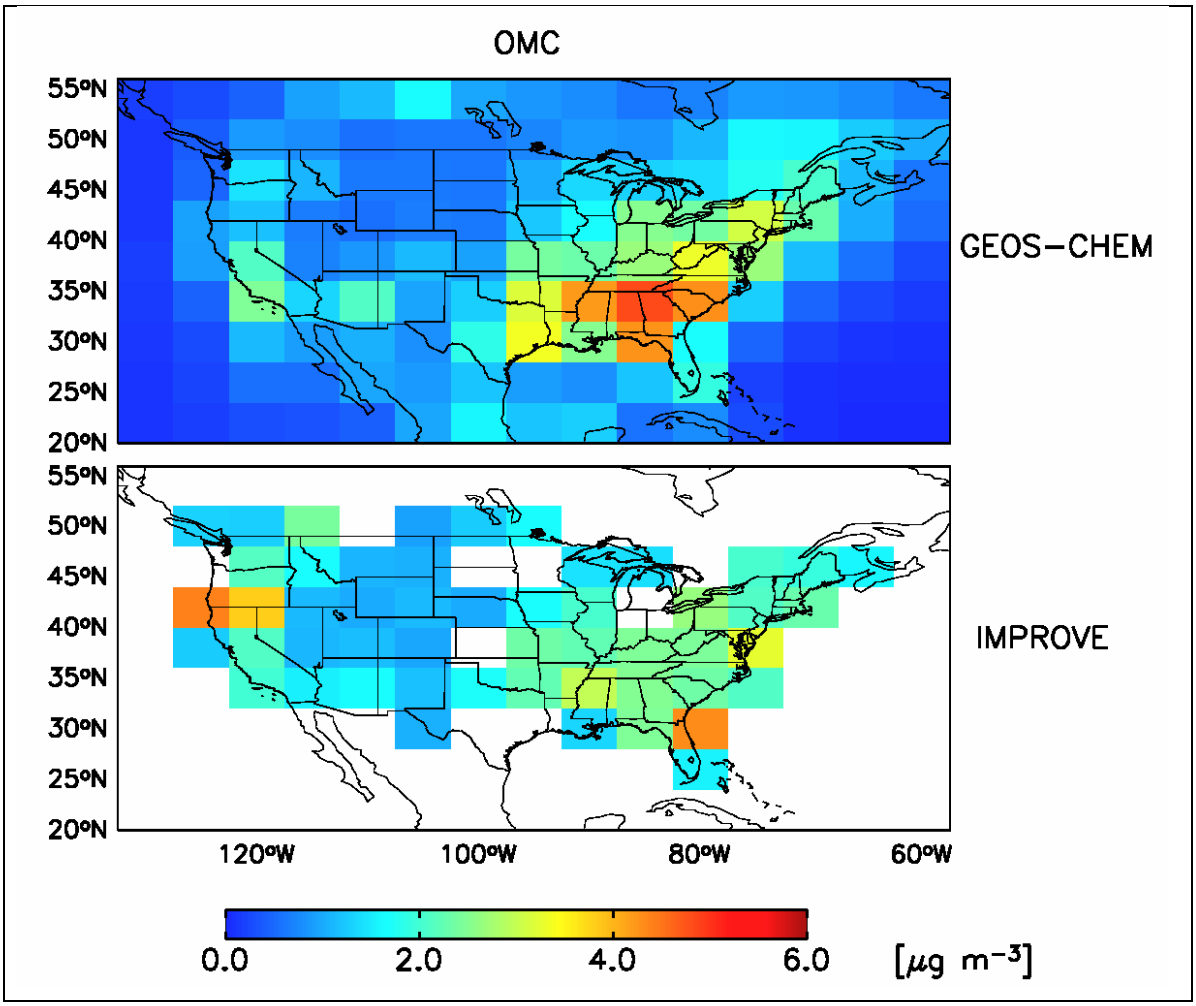

Figure G-8. GEOS-CHEM 2002 Organic Matter Carbon (OMC) annual average performance compared to IMPROVE (145 sites) and CASTNET monitoring networks.

Model performance for organic matter carbon (OMC) and elemental carbon (EC) are acceptable. Performance at the boundaries is driven by quality of the fire inventory for international emissions. While a new biomass burning inventory was developed as part of this study, monthly average fire emissions do not accurately capture the impacts from episodic fires.

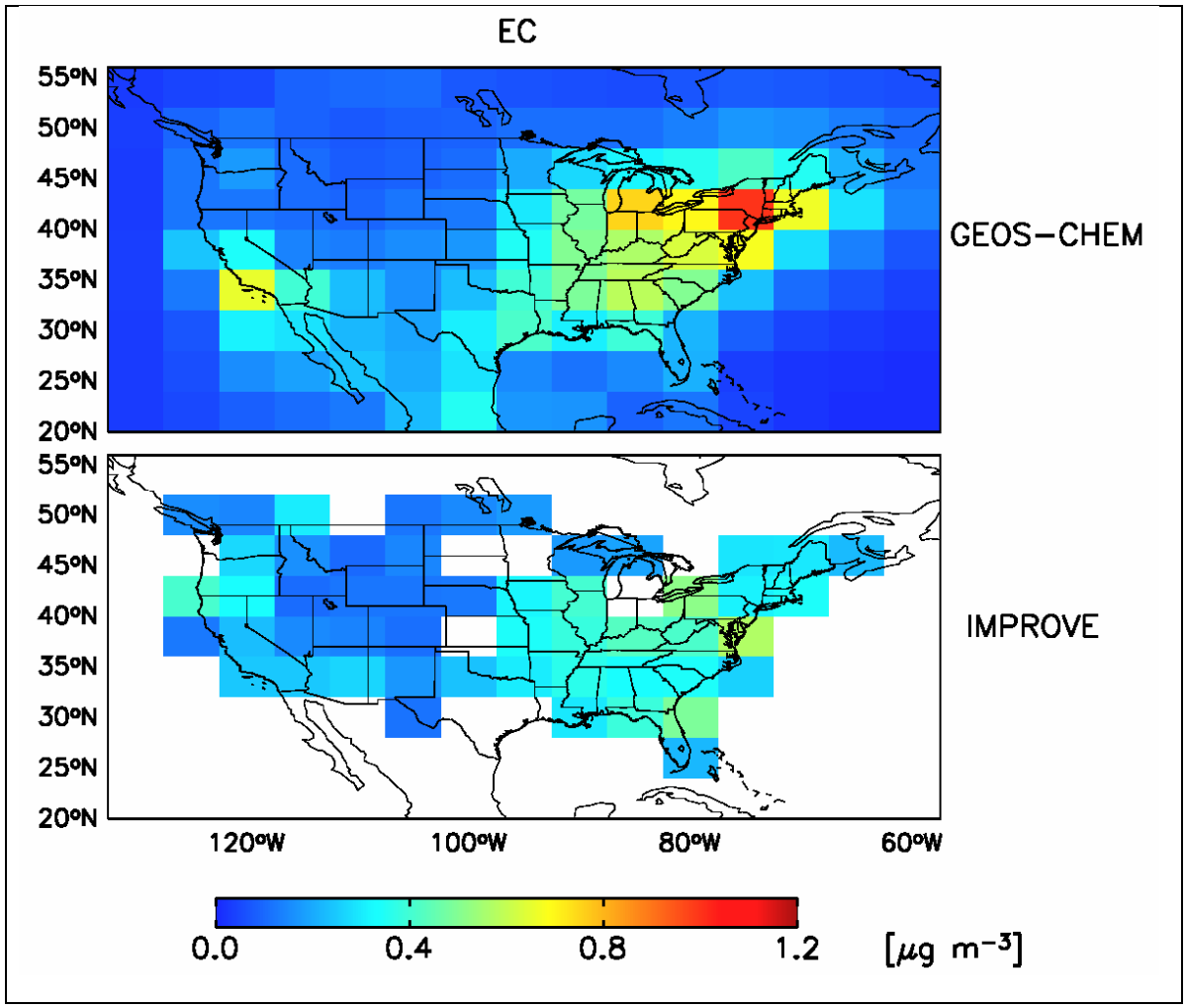

Figure G-9. GEOS-CHEM 2002 Elemental Carbon annual average performance compared to IMPROVE (145 sites) and CASTNET monitoring networks.

GEOS-CHEM 3-hr average outputs were used as inputs to CMAQ modeling for 2002 and 2018. Since VISTAS did not have information to project future changes in international emissions, VISTAS used the same boundary conditions in 2018 as developed for 2002.

References

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