4.11 REGIONAL MODELING OF PARTICULATE MATTER (PM) USING THE CMAQ AND CAMx PHOTOCHEMICAL GRID MODELS

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1. INTRODUCTION

The 1977 Clean Air Act Amendments and recent Regional Haze Rule (RHR) are aimed, in part, at returning 156 national parks and wilderness areas (Federal Class I areas) to their natural visibility conditions by 2064. Since visibility impairment is often a regional issue as relevant pollutants can be originated from sources located across broad geographic areas, the US Environmental Protection Agency (EPA) has created five regional planning organizations (RPOs) to address regional haze and related issues. Two state-of-science 'one-atmosphere' regional modeling platforms are widely used for ozone, fine particulate, and regional haze applications in the U.S. and abroad. One is CMAQ, EPA's Community Multiscale Air Quality (CMAQ) modeling system (Byun and Ching, 1999) while the Comprehensive Air Quality Model with Extensions (CAMx) is another open-source photochemical-aerosol model (ENVIRON, 2004). Most current 8-hr ozone, fine particulate and/or regional haze modeling being carried out in the U.S. by the States, RPOs, or stakeholder groups entail the use of one or both of these modeling systems: The Midwest RPO (MRPO) is currently using CAMx as the primary model; The Visibility Improvement State and Tribal Association of Southeast (VISTAS) selected CMAQ as the primary modeling framework and employed CAMx in parallel with CMAQ as a corroborative and diagnostic tool; The Central States Regional Air Partnership (CENRAP) is currently applying both CMAQ and CAMx models in parallel. There are many advantages in applying and evaluating alternative models. All models have uncertainties and limitations. By applying multiple models, we obtain more confidence in the results and also obtain an estimate of the uncertainties in the modeling. The benefits of employing a pair of complementary state-of-the-science air quality models include:

Diagnosis: To serve as an efficient diagnostic tool for addressing model performance issues. CMAQ and CAMx both include process analysis that can help diagnose model performance. CAMx's suite of diagnostic probing tools and its flexi-nesting algorithms make it an attractive tool for assisting in the diagnosis of model performance.

Model Evaluation Corroboration: To provide corroboration of the base case model performance evaluation and help identify any compensatory errors in the modeling systems.

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Emission Control Response Corroboration: To provide corroboration of the response of a modeling system to generic and specific future-year emissions changes on modeled gas-phase and particulate aerosol concentrations and resultant regional haze impacts.

Quantification of Model Uncertainty: To provide one estimate of the range of uncertainty in the annual and episodic base case simulations, and in the estimates of $PM_{2.5}$ and visibility reductions associated with future emissions-change scenarios.

Alternative Science: CAMx and CMAQ contain alternative science algorithms that may elucidate model performance issues with one model or the other or provide an alternative approach for simulating aerosols.

Backup Contingency: To provide a "backstop" model in the event that unforeseen difficulties with the primary model occur.

In this paper, a brief description of the two modeling systems and their features are presented. We also discuss the extension of the CAMx source apportionment probing tools to the PM portions of the code. In addition, the two models are applied to US continental modeling domain for February and July 2002 and their results are intercompared.

2. CMAQ AND CAMx MODELING SYSTEM

CMAQ (v4.4) is a well documented model and has been progressively updated by the user community. Key science features in the CMAQ's aerosol component include modal representation where the PM size distribution is represented as the superposition of three lognormal subdistributions (Aitken, accumulation, and coarse modes), equilibrium mass transfer between the aerosol and gas phases, inorganic aerosol composition equilibrium modeled by ISORROPIA, and the inclusion of RADM aqueous phase chemistry. CMAQ treats Secondary Organic Aerosol (SOA) with the Secondary Organic Aerosol Model (SORGAM) update consisting of a reversible semi-volatile scheme whereby VOCs may be converted to condensable gases (CG). These condensable gases may then form SOA and later evaporate back to CG depending on thermodynamic conditions. Since CMAQ aerosol module emphasized fine PM, the coarse mode was implemented in a noninteractive way. CMAQ also assumes that all secondary PM is in the fine (Aitken and accumulation) modes.

CAMx is also a publicly available, three-dimensional, multi-scale photochemical aerosol grid modeling system designed to treat variety of air quality issues including ozone, PM, visibility, acid deposition, mercury and air toxics (www.camx.com). The flexible CAMx architecture makes it a convenient, robust host model for implementation of several 'probing tool' including process analysis, decoupled direct method (DDM), and the ozone source apportionment technology (OSAT). Key attributes of the CAMx system include the following:

- Two-way grid nesting that supports multiple levels of fully interactive grid nesting;
- Carbon Bond IV (CB-IV) or Statewide Air Pollution Research Center (SAPRC-99) chemical mechanisms;
- Two chemical solvers, the CAMx Chemical Mechanism Compiler (CMC) Fast Solver or the highly accurate Implicit-Explicit Hybrid (IEH) solver;
- Multiple numerical algorithms for horizontal transport, including the Piecewise Parabolic Method (PPM) and Bott advection solvers;
- A sub-grid-scale Plume-in-Grid algorithm to treat the near-source plume dynamics and chemistry from large NO_x point-source plumes;
- The ability to interface with a variety of meteorological models, including the MM5 and RAMS prognostic hydrostatic meteorological models and the CALMET diagnostic meteorological model (others are also compatible);
- The ozone source apportionment technology (OSAT), which identifies the ozone contribution due to geographic source regions and source categories (e.g., mobile, point, biogenic);
- The DDM sensitivity method, which is implemented for emissions and ICs/BCs to obtain first-order sensitivity coefficients for all gas-phase species; and
- Two separate treatments of PM. The Coarse/Fine (CF) scheme uses two size sections and assumes that all secondary PM is in the fine mode like CMAQ. The multi-section "full-science" approach uses aerosol modules developed at Carnegie Mellon University (CMU) where a sectional approach is used to represent the PM size distribution.

In addition to the probing tools described above, the PM source apportionment technology (PSAT) was recently developed and will be discussed in the following section. Also the DDM implementation in CAMx is currently being extended to PM species.

3. CAMx PSAT

PSAT has been developed for CAMx to provide geographic and source-category-specific PM source apportionment (Yarwood et al., 2004). PM source apportionment information from PSAT is useful for:

- understanding model performance and thereby improving model inputs/formulation,
- performing culpability assessments to identify sources that contribute significantly to PM pollution, and
- designing the most effective and cost-effective PM control strategies.

Source apportionment for primary PM is relatively simple to obtain from any air pollution model, because source-receptor relationships are essentially linear for primary pollutants. Gaussian steady-state models and Lagrangian puff models have been used extensively to model primary PM pollution from specific sources, which provides source apportionment. The Gaussian and Lagrangian approaches work for primary PM because the models can assume that emissions from separate sources do not interact. This assumption breaks down for secondary PM pollutants (e.g., sulfate, nitrate, ammonium, secondary organic aerosol), so puff models may dramatically simplify the chemistry (to eliminate interactions between sources) when they are applied to secondary PM. Eulerian photochemical grid models are better suited to modeling secondary pollutants because they account for chemical interactions between sources. However, these models do not naturally provide source apportionment because the impact of all sources has been combined in the total pollutant concentration. PSAT has been developed to retain the advantage of using a grid model to describe the chemistry of secondary PM formation and also provide source apportionment.

The CAMx PSAT uses reactive tracers, which are extra species added to a grid model to track pollutants from specific sources. For example, a standard grid model calculates concentrations for a species *X* that has many sources and so the concentration of *X* is the total concentration due to all sources. A reactive tracer (x_i) is assigned to for each source (*i*) with the intention that the sum of the reactive tracers will equal total concentration

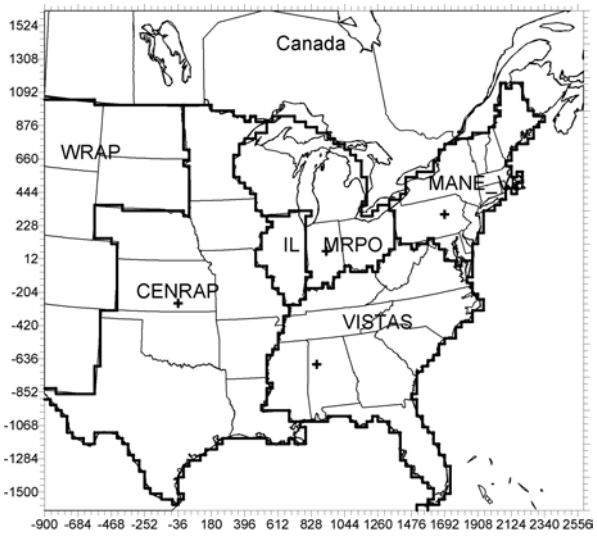


Figure 1. The CAMx modeling domain for PSAT testing showing sub-division to geographic areas and locations of four hypothetical point sources (+ symbols)

($X=\sum x_i$). The challenge is to develop numerical algorithms for solving the reactive tracer concentrations that ensure that this equality is maintained. Depending upon the formulation of the tracer algorithms, it may be possible to model tracers for a single source of interest and omit tracers for all other sources, or it may be necessary to include tracers for all sources (as is the case for PSAT). Reactive tracers can potentially provide true source apportionment ($X=\sum x_i$), however the numerical value of the source apportionment will depend upon assumptions within the reactive tracer formulation. In particular, for any process that is non-linear in species concentrations (e.g., chemistry) there is no unique way to assign the total concentration change to the reactive tracers.

3.1 *Testing of PSAT*

Initial testing of PSAT for sulfate and nitrate is shown here. Sulfate was tested by comparing PSAT to zero out results in full 3-D CAMx simulations. Zero-out method is a type of sensitivity analysis where a specific emissions input is set to zero to reveal the source's impact. This method is not expected to agree perfectly with PSAT because it does not give true source apportionment in non-linear systems. That is, the sum of zero-out impacts over all sources will not equal the total concentration. However, non-linearity in sulfate formulation chemistry is expected to be less important than for other secondary PM species. The most complex PSAT chemistry algorithm is for nitrate. Zero out tests were not used for nitrate because the relationship between NO emissions and nitric acid may be highly non-linear. Therefore, 1-D (box model) tests were used to evaluate PSAT results for nitrate by comparing against the method called Source Oriented

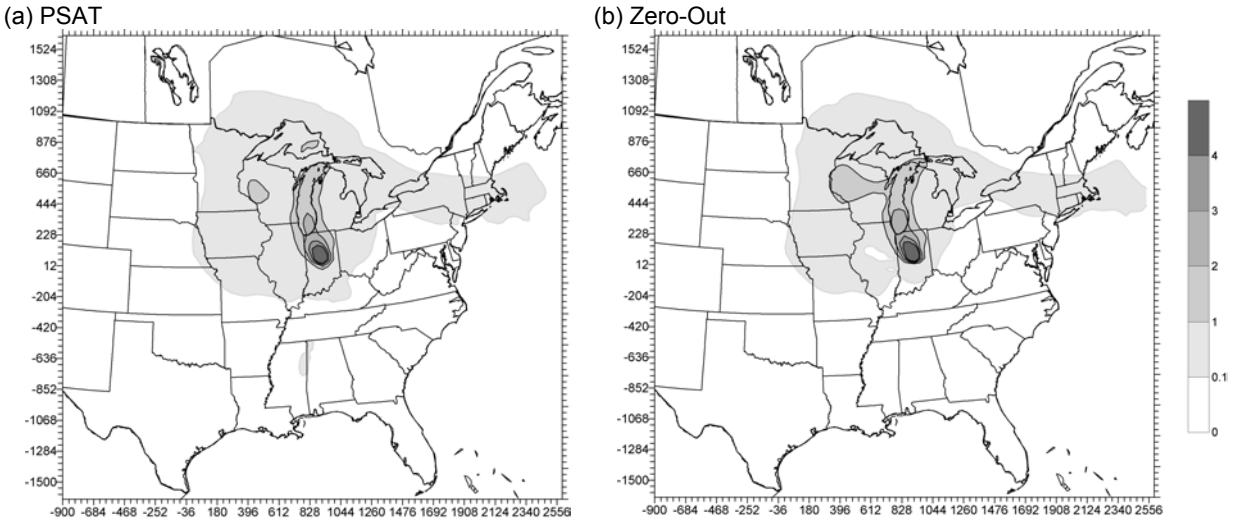


Figure 2. Comparison of sulfate impacts (μg/m^3) from the hypothetical MRPO point source on 28 June 2001 at hour 15: (a) PSAT result; (b) Zero-Out result

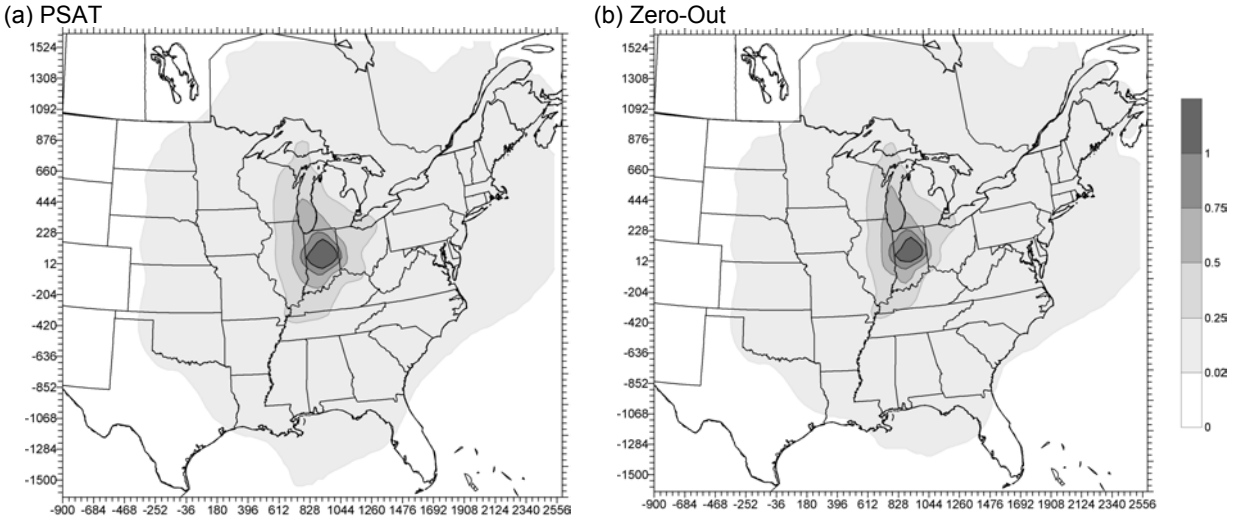


Figure 3. Comparison of episode average (June 18 to July 21, 2001) sulfate impacts (μg/m^3) from the hypothetical MRPO point source: (a) PSAT result; (b) Zero-Out result

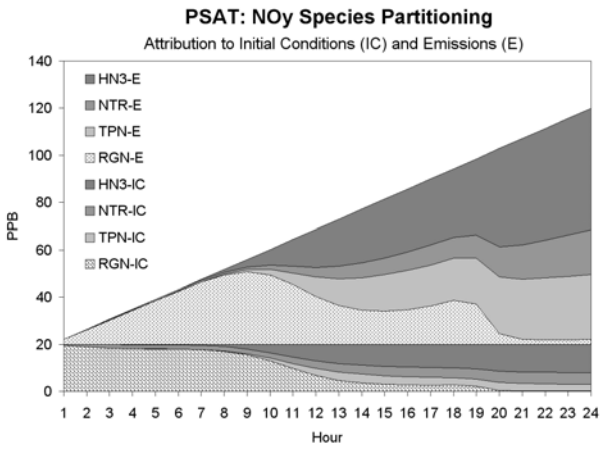


Figure 4. PSAT apportionment of reactive nitrogen species to initial conditions and emissions during a 24-hour box model simulation

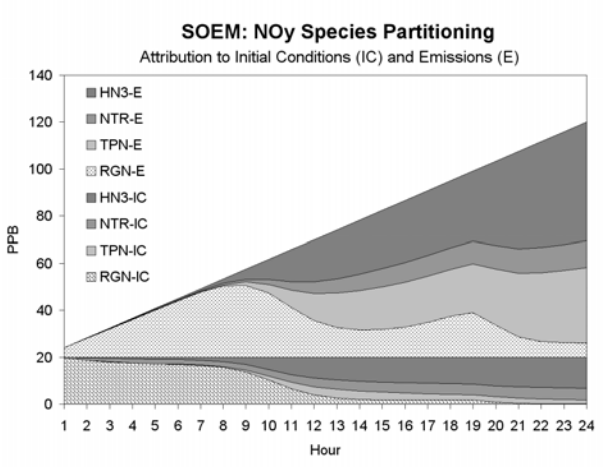


Figure 5. Source-Oriented External Mixture (SOEM) apportionment of reactive nitrogen species to initial conditions and emissions during a 24-hour box model simulation

External Mixture (SOEM) method developed by Kleeman and Cass (2001).

The PSAT performance for sulfate was tested using a CAMx database developed by the Midwest RPO (MPRO) for PM and visibility modeling of the Eastern US (Baker, 2004). The model was exercised from June 18, 2001 to July 21, 2001 and used a 36-km modeling grid with meteorology developed using the mesoscale model version 5 (MM5). The modeling domain was sub-divided to geographic areas according to RPOs (Figure 1) and the RPOs are labeled by their respective acronyms (MRPO, MANE-VU, VISTAS, CERAP and WRAP). The state of Illinois (IL) was split out from the Midwest RPO (MPRO) to test the ability of PSAT to apportion the contribution from a single state. Four hypothetical point sources were added near the middle of the MRPO, MANE-VU, VISTAS and CENRAP areas (shown by the + symbols in Figure 1) to test the ability of PSAT to track contributions from singe sources. The hypothetical point sources were chosen to be generally representative of a large coal-fired electrical generating units, but do not represent actual sources at these locations. In total, sulfate was apportioned to 11 source regions including a remainder area for Canada, Mexico and over water, for 13 source groups in all when initial concentrations (IC) and boundary conditions (BC) are included. The sulfate impacts from the hypothetical MRPO point source are compared in Figure 2 at a single hour (hour 15) on 28 June 2001. The spatial distribution of sulfate impacts is very similar in the PSAT and zero-out results as shown by the edge of plume is impact (0.1 $\mu g/m^3$ level). There are differences in the areas of larger impacts (e.g., the 1 and 2 $\mu g/m^3$ levels) and these are due to the effects of non-linear chemistry in the zero-out test. As discussed above, sensitivity methods such as zero-out do not provide accurate source apportionments for non-linear processes. Sulfate formation can be limited by the availability of oxidants, especially hydrogen peroxide in aqueous-phase chemistry, which will tend to depress the maximum impact levels in zero-out runs as well as shift impacts further downwind (to where oxidant availability is no longer limiting). The oxidant limiting effect on zero-out sulfate impacts is most easily seen from the 2 $\mu g/m^3$ level extending further downwind over Lake Michigan in the zero-out result than the PSAT source apportionment result. The episode average sulfate impacts from the hypothetical MRPO point source are compared in Figure 3 for the entire 28 June to July 21, 2001 modeling period. The spatial distribution of sulfate impacts is very similar in the PSAT and zero-out results. The maximum impact occurs very close to the source and is higher in the PSAT result (2.2 $\mu g/m^3$) than the zero-out result (1.8 $\mu g/m^3$) due to the effect of oxidant limitation on sulfate impacts determined by the zero-out method. The PSAT sulfate tests provided a comparison of the efficiency of the PSAT method compared to zero-out modeling. Zero-out modeling requires a new full model simulation for each source contribution determined, so the incremental time for each “apportionment” is the same as for the model base case. In contrast, the marginal cost for each PSAT source apportionment was about 2% of the time required for the base case. That is, PSAT sulfate PM source apportionment is 50 times more efficient than zero-out runs.

The PSAT nitrate algorithms were tested using CAMx for a 1-D (box model) problem in order to focus upon the ability of the PSAT chemical algorithms to track nitrate apportionment. The box model problem was for summer conditions and PSAT was used to apportion nitrate between 20 ppb of initial NO_x and 100 ppb of NO_x emission injected continuously through the 24-hour run. There was no ammonia in the box model so that nitric acid remained in the gas phase rather than forming PM nitrate. The PSAT apportionment of NO_y to initial conditions (ICs) and emissions during the 24-hour box model simulation is shown in Figure 4. The total NO_y apportioned to ICs remains constant at 20 ppb throughout the simulation but the apportionment

changes over time from NO_x (RGN-IC) at the start to PAN (TPN-IC), organic nitrates (NTR-IC) and nitric acid (HN3-IC). The NO_y apportioned to emissions increased linearly throughout the simulation and the apportionment also evolved as the NO_x emissions (RGN-E) reacted. At the start of the simulation RGN (NO_x) is dominated by ICs whereas late in the day (hour 18) RGN is dominated by emissions. The source apportionments shown in Figure 4 are reasonable and show necessary attributes (such as conserving the total 20 ppb of ICs). The SOEM apportionment of NO_y to ICs and emissions during the 24-hour box model simulation is shown in Figure 5. The time evolution of the source apportionments for the ICs and emissions is nearly identical for SOEM (Figure 5) and PSAT (Figure 4). It is not clear that either method is more "correct," or indeed that a correct source apportionment result exists for this test, but the consistency between the PSAT and SOEM results is encouraging that these two completely different and independent techniques are providing similar nitrogen species source apportionment results.

4. COMPARATIVE EVALUATION OF THE CMAQ AND CAMx MODELS

CMAQ and CAMx were set up on the same RPO Unified Continental 36-km Modeling Grid domain for February and July 2002 (Figure 6). The meteorological inputs for both models were generated using the latest 2002 MM5 simulations. The CAMx ICs/BCs and emissions were generated from the CMAQ inputs using the CMAQ-to-CAMx IC/BC and emissions processors to provide consistent modeling inputs. The model simulations used 15-day spin up period that started on January 16, 2002 and June 15, 2002 for the February and July 2002 runs, respectively. Although approximately 45 days were simulated for each of the monthly application, only the last approximately 30 days were analyzed in the model performance evaluation. The two models were evaluated using speciated PM measurements from four separate air quality monitoring networks: IMPROVE, CASTNet, STN, and SEARCH.

4.1 *Evaluation for Sulfate*

Figure 7 compares the CMAQ and CAMx sulfate (SO_4) model performance for February and July 2002 at sites across the U.S. from the IMPROVE, CASTNet, STN and SEARCH monitoring networks. SO_4 model performance in July 2002 across the U.S. for the two models is generally quite good with fractional bias less than 10% and fractional error of 30-40%. The exception to this is the SEARCH network in the southeastern U.S. where both models exhibit a slight overestimation tendency of 17% (CMAQ) and 25% (CAMx) and errors of approximately 50% (Figure 7a). Both models generally exhibit a slight SO4 overestimation tendency for February 2002 with the CAMx overestimation tendency being greater than CMAQ. For example, across the U.S. IMPROVE monitors in February 2002, CMAQ and CAMx produce fractional bias values of 26% and 41% and fraction error values of 49% and 56%, respectively.

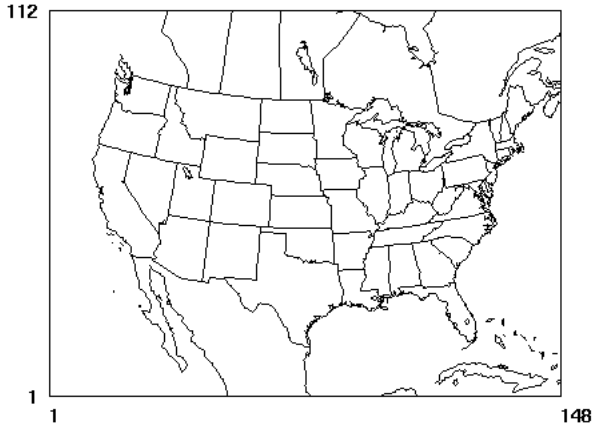


Figure 6. RPO Unified Continental 36-km modeling grid domain

4.2 *Evaluation for Nitrate*

Comparisons of the CMAQ and CAMx nitrate (NO_3) model performance for February and July 2002 are shown in Figure 8. NO_3 model performance for both models is poor exhibiting a summer underestimation and winter overestimation tendency. The summer NO_3 underestimation is more severe in CMAQ than CAMx, whereas the winter NO_3 overestimation is more severe with CAMx than CMAQ. In any event, NO_3 model performance for both models is fairly poor with the winter overestimation tendency more of a concern given that NO_3 can be is a higher fraction of the fine particulate in the winter.

4.3 *Evaluation for Organic Carbon (OC) and Elemental Carbon (EC)*

Figure 9 compares the CMAQ and CAMx OC and EC model performance at IMPROVE sites across the U.S. and February and July 2002. Both models exhibit a similar fractional bias underestimation in July 2002 of -34% and -39% and fractional errors of 83% and 63% for CMAQ and CAMx, respectively. OC performance of the two models in the summer is also very similar only with an overestimation bias with fractional bias values of 30% and 33% and fraction errors of 63% and 65% for CMAQ and CAMx, respectively. EC model performance in July 2002 for the two models is different with CMAQ exhibiting a -16% underestimation and CAMx exhibiting a +16% overestimation fractional bias and the CAMx fractional error (57%) being slightly lower than that for CMAQ (68%). In February 2002, however, CMAQ exhibits a lower EC fractional bias of -4%, whereas the CAMx value is 47% and the CMAQ fractional error (60%) is slightly lower than seen for CAMx (69%).

4.4 *Evaluation for Other $PM_{2.5}$ (SOIL) and Coarse Matter (CM)*

The SOIL and CM model performance across the U.S. IMPROVE monitors for CMAQ and CAMx and February and July 2002 are shown in Figure 10. The SOIL species is overestimated by both CMAQ and CAMx in both July (17% and 62%) and February (102% and 136%). This overestimation is expected as the modeled species mapped to the IMPROVE SOIL measurement contains other unidentified compounds, whereas the IMPROVE SOIL measurement is built up from the elements. In fact, the usual application of the CAMx model is to separately model the fine and coarse crustal PM from the other unidentified PM components so that it can be compared directly against the IMPROVE SOIL measurement. When CAMx treats dust emissions as separate crustal species, much better SOIL performance is seen. However, since the CAMx emissions for this application were generated from the CMAQ emissions using the CMAQ-to-CAMx processor and CMAQ does not separately treat the SOIL species then CAMx could not be configured with the separate fine and coarse crustal species.

5. CONCLUSIONS

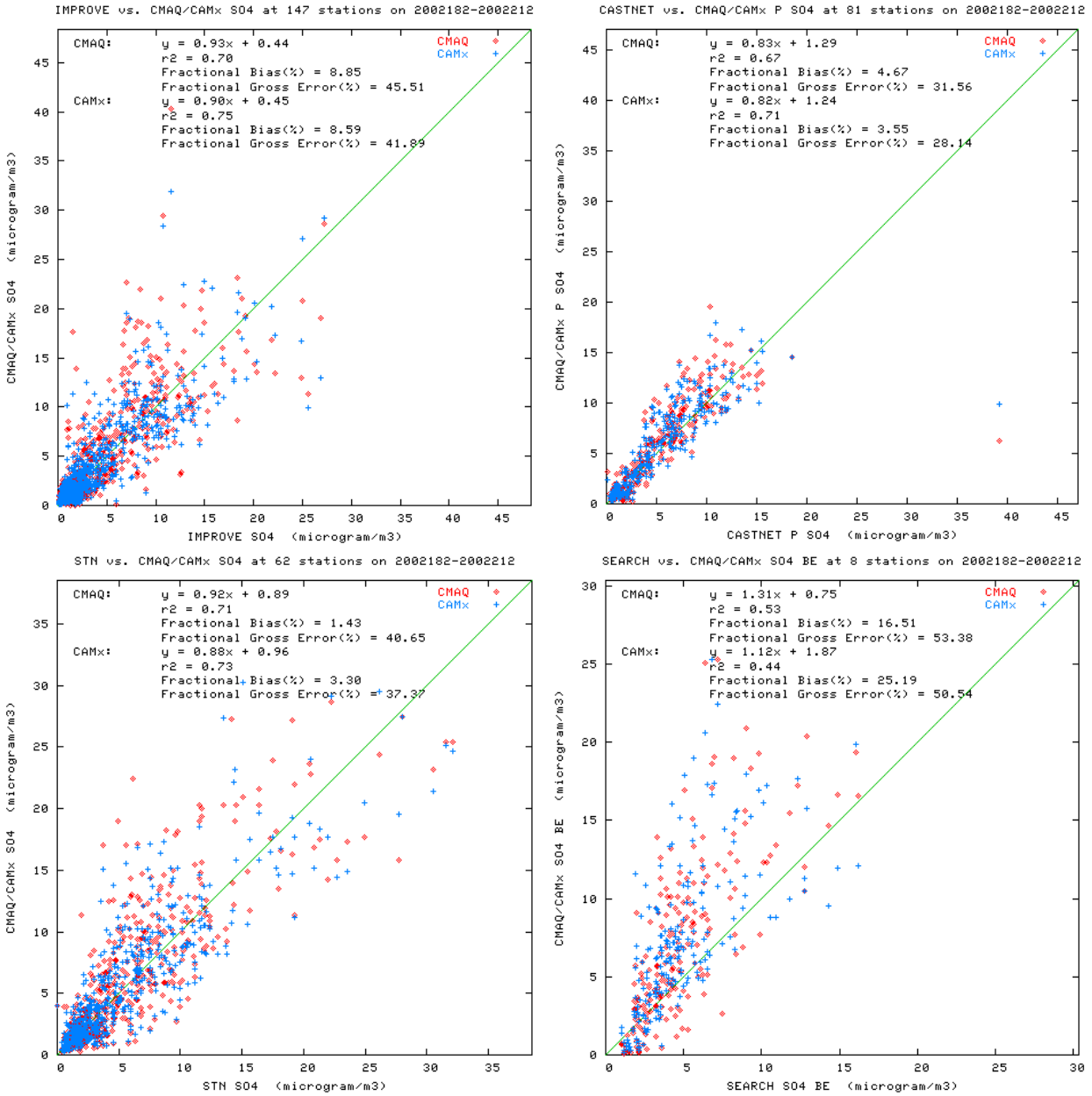


Figure 7a. Comparison of CMAQ (red) and CAMx (blue) sulfate (SO4) model performance at sites across the U.S. and July 2002 using the IMPROVE (top left), CASTNet (top right), STN (bottom left) and SEARCH (bottom right) monitoring networks

CMAQ and CAMx are two widely used photochemical grid models and three of the five RPOs are running the two models side-by-side to address regional PM/ozone/visibility in the western (WRAP), central (CENRAP), and southeastern (VISTAS) portions of the US.

PM Source Apportionment Technology (PSAT) is one of the 'probing tools' provided in CAMx to assist the assessment of source impact on regional air quality. The initial test results show that PSAT provides consistent results with other methods such as zero-out method and SOEM while being much more efficient.

The model performances by CMAQ and CAMx were compared using consistent emissions, meteorological and air quality databases. Both models yield comparable performance for most species. Sulfate performance is quite good while nitrate performance exhibits a summer underprediction and winter overprediction tendency. SOIL is overpredicted by both models due to inconsistency between modeled and measured SOIL species. Underestimation of CM is partially because all the secondary PM species in the

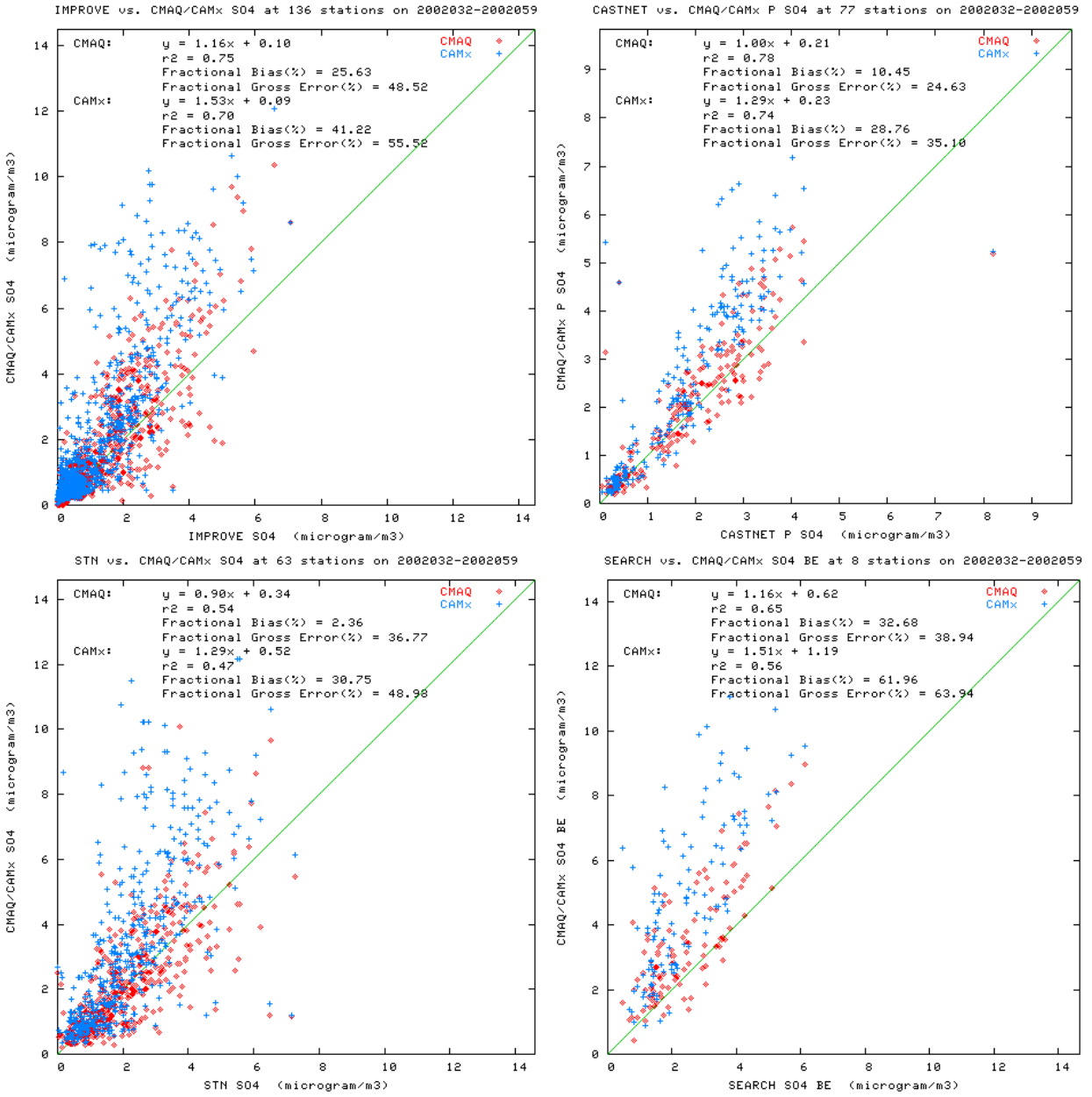


Figure 7b. Comparison of CMAQ (red) and CAMx (blue) sulfate (SO4) model performance at sites across the U.S. and February 2002 using the IMPROVE (top left), CASTNet (top right), STN (bottom left) and SEARCH (bottom right) monitoring networks

models are assumed to be fine.

6. REFERENCES

Baker, K., 2004; Modeling System Performance: PM2.5/Haze. Midwest Regional Planning Organization (MRPO) Workshop. November. (www.ladco.org/tech/photo/present/MPE_PM25.pdf)

Byun, D. W., Ching, J. K. S. (Eds.), 1999; Science algorithms of the EPA Models-3 Community Multi-scale Air Quality (CMAQ) Modeling System. US EPA Report No. EPA/600/R-99/030, Office of Research and Development, Washington, DC.

ENVIRON, 2004; User's Guide: Comprehensive Air Quality Model with Extensions (CAMx) – Version 4.10s. (www.camx.com)

Kleeman, M. J., Cass, G. R., 2001; A 3D Eulerian source-oriented model for an externally mixed aerosol. Environ. Sci. Technol., **35**, 4834-4848.

Yarwood, G., Morris, R. E., Wilson, G. M., 2004;

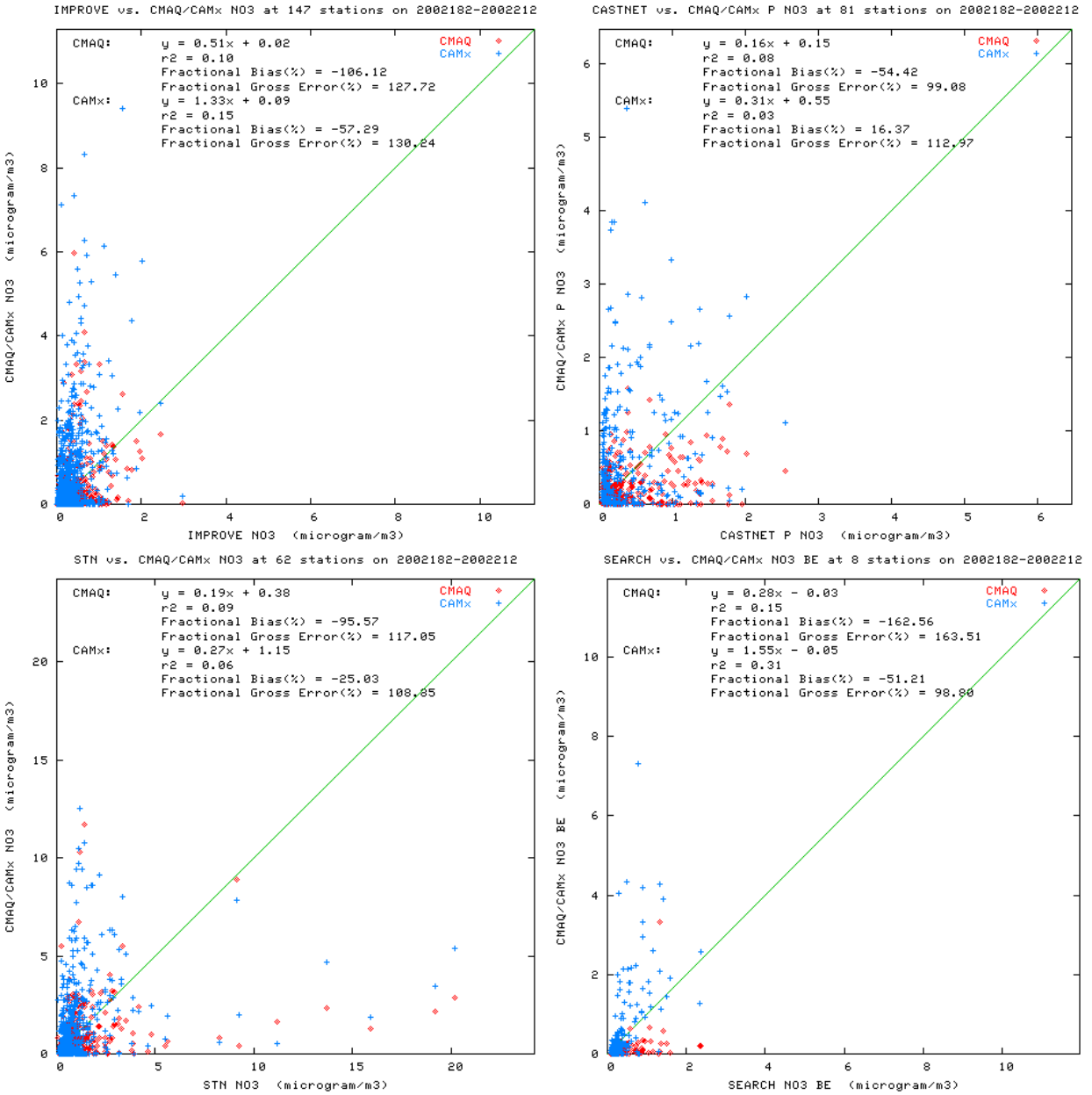
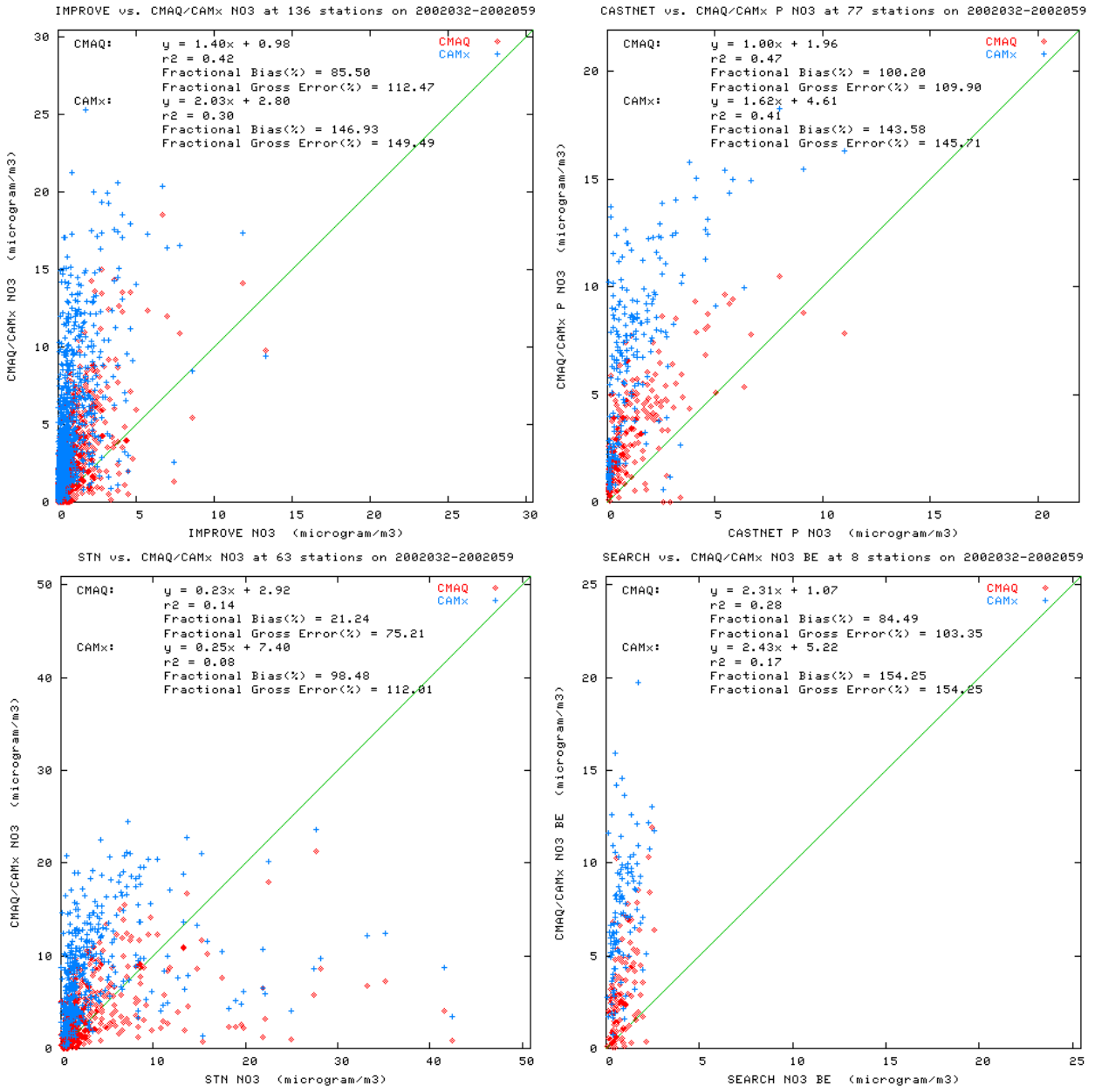


Figure 8a. Comparison of CMAQ (red) and CAMx (blue) nitrate (NO3) model performance at sites across the U.S. and July 2002 using the IMPROVE (top left), CASTNet (top right), STN (bottom left) and SEARCH (bottom right) monitoring networks

Particulate Matter Source Apportionment Technology (PSAT) in the CAMx Photochemical Grid Model. Presented at the International technical Meeting. Banff, Canada. October.



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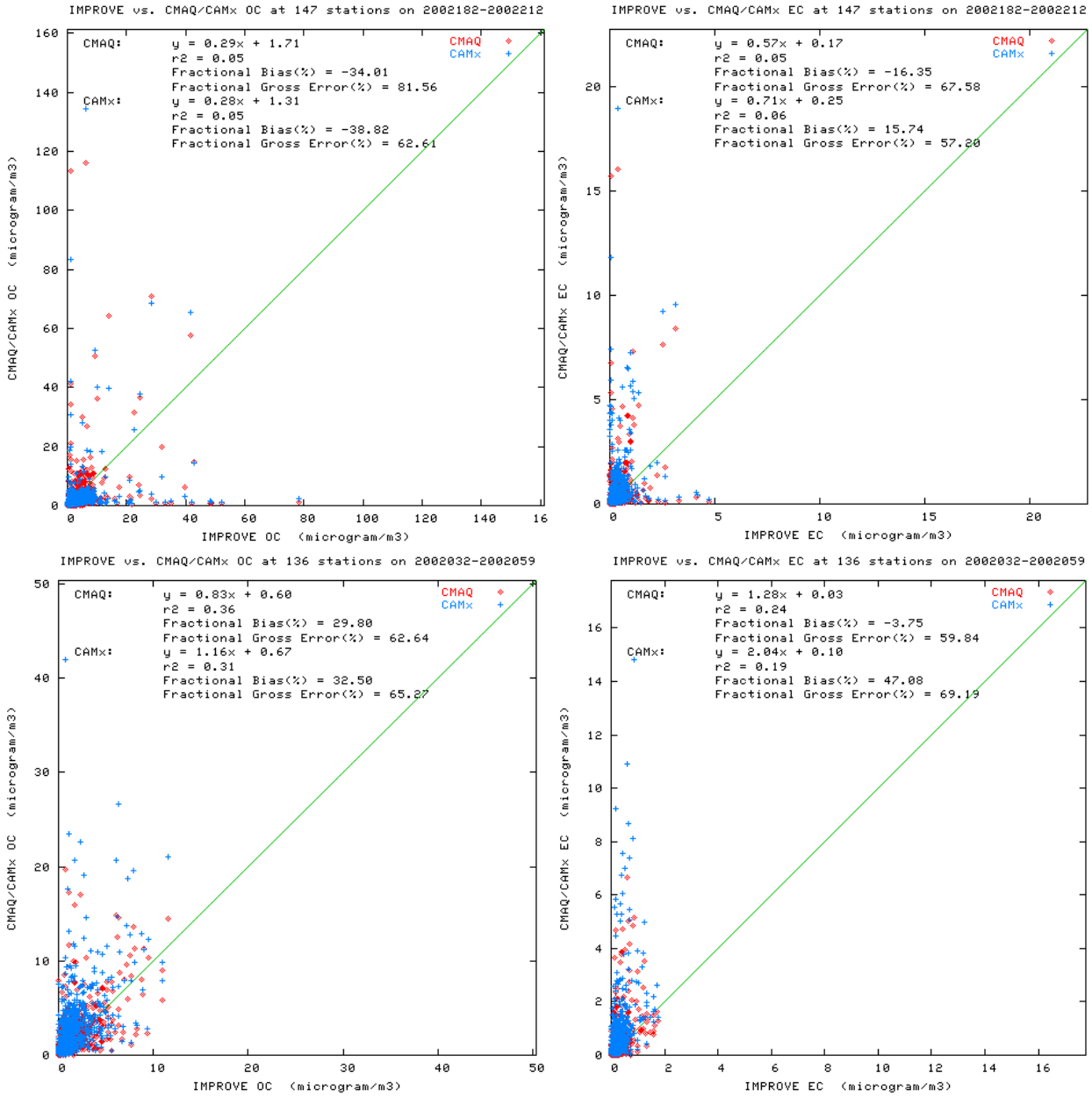

Figure 9. Comparison of CMAQ (red) and CAMx (blue) OC (left) and EC (right) model performance at sites across the U.S. for July 2002 (top) and February 2002 (bottom) using the IMPROVE monitoring network

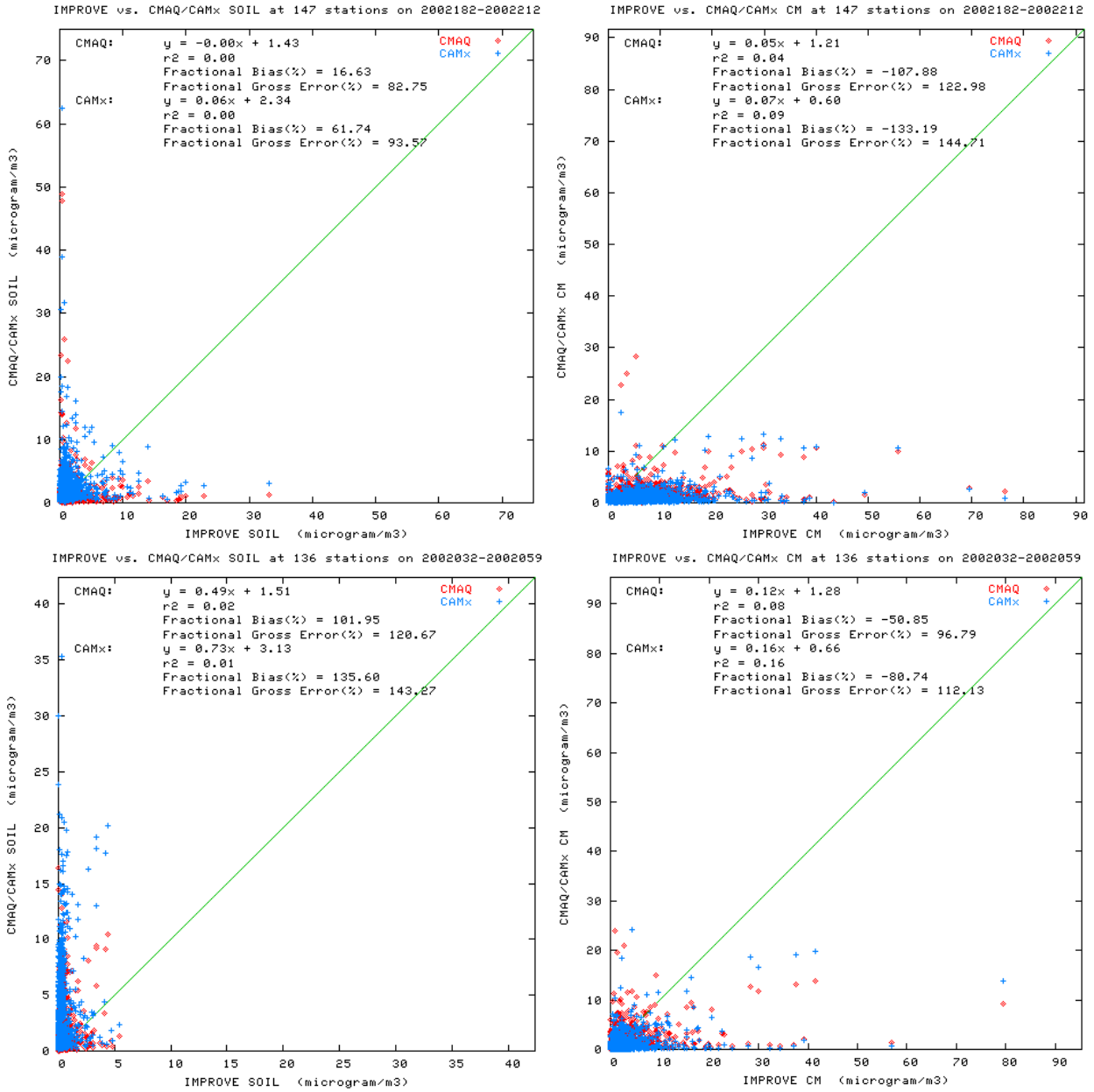

Figure 10. Comparison of CMAQ (red) and CAMx (blue) SOIL (left) and CM (right) model performance at sites across the U.S. for July 2002 (top) and February 2002 (bottom) using the IMPROVE monitoring network