Final Summary Report

**Annual PM Modeling and
Analysis of Episode Aggregation Uncertainties for
Annual Average PM$_{2.5}$ and Visibility Modeling**

CRC Project A-44a/b

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EXECUTIVE SUMMARY

The Coordinating Research Council (CRC) sponsored Project A-44 to perform the following activities:

- Implement CMU sectional PM modules of PMCAMx into the latest version of CAMx (Version 4.03) so that the "full-science" sectional and abbreviated "one-atmosphere" 2-section (course/fine) PM modules are available in the same modeling platform (CAMx Version 4.10).
- Test the new multiple PM versions of CAMx (Version 4.10) using the October 1995 database for Southern California, update the CAMx user's guide and post on CAMx website (www.camx.com) to be publicly available.
- Perform annual modeling of 1996 and the western US using the new version of CAMx in its 2-section and multi-section configuration and compare model performance with the REMSAD and CMAQ models.
- Review, evaluate and recommend statistical methods for episode aggregation to longer-term average PM issues including the use of models for annual $PM_{2.5}$ and visibility projections.

DEVELOPMENT OF CAMx VERSION 4.10

At the start of CRC Project A-44, there were two versions of CAMx available for use, PMCAMx Version 3.01, which included the "full-science" multi-section PM modules developed by Carnegie Mellon University (CMU) and CAMx Version 4.03 that included an abbreviated "one-atmosphere" 2-section treatment of PM processes. The first work element under CRC Project A-44 was the implementation of the "full-science" CMU multi-section PM modules from PMCAMx into the latest CAMx Version 4 code so that the "full-science" and "one-atmosphere" PM treatments are available in the same platform. The resultant integrated code, CAMx Version 4.10, was then tested using the October 1995 episode for Southern California (Koo, Morris and Yarwood, 2003) and the CAMx Version 4.10 model and updated User's Guide (ENVIRON, 2004) were made available on the CAMx website (www.camx.com). The initial testing of CAMx V4.10 included testing of the sectional PM representation (2, 4 and 10 sections); use of the equilibrium, dynamic and hybrid PM size representation, and testing of the bulk 1-section (RADM) and multi-section (VRSM) aqueous-phase chemistry modules.

1996 ANNUAL MODELING

The new integrated CAMx code was then applied using the 1996 annual database for the western United States (US) developed by the Western Regional Air Partnership (WRAP). CAMx was applied using two configurations: (1) the Mechanism 4 (M4) "one-atmosphere" 2-section configuration where all secondary PM is assumed to be in the fine fraction; and (2) use of the "full-science" sectional approach using 4 sections (4Sec) that allows secondary PM to grow into the coarse mode. The CAMx annual simulation was evaluated against PM observations from the IMPROVE and CASTNet networks and wet deposition from the NADP networks and the model performance was compared against that of EPA's Models-3 Community Multi-scale Air Quality (CMAQ, Version 4.3) and the Regional Modeling System for Aerosols and Deposition

(REMSAD Version 7). The four models exhibited similar model performance for PM concentrations in the western US and 1996:

- Annual average sulfate (SO4) bias for all four models were within " 20%; however, the low annual bias was partly due to compensating errors with a winter overestimation bias compensated for by a summer underestimation bias; CMAQ and REMSAD exhibited the lowest annual SO4 bias whereas CAMx M4 and 4Sec exhibited the lowest monthly SO4 bias.
- Annual Nitrate (NO3) bias was also fairly low, but exhibited a large (~50%) overestimation bias in the winter and large (~ -100%) underestimation bias in the summer; all four models performed poorly for NO3.
- Organic Carbon (OC) was underestimated by the four models across the year, with the underestimation bias ranging from –30% to –70%.
- Elemental Carbon (EC) was also underestimated by the four models, with REMSAD exhibiting the lowest underestimation bias (-5% to –15%) and CMAQ exhibiting the highest EC underestimation bias (-35% to –50%), with CAMx in between.

In summary, the four models exhibited similar model performance. The use of the CAMx sectional approach indicated that approximately 10% of the Coarse Matter was due to secondary SO4 and NO3, suggesting that the CAMx_M4, CMAQ and REMSAD assumption that all secondary PM is fine, introduces small errors in the modeling results.

50% emission reduction scenarios were conducted for anthropogenic NOx, VOC, NH3 and SO2 emissions using the CAMx M4 and 4Sec configurations. The two model configurations generally agree in the change in PM concentrations in response to the emissions controls. For example, both models agree that the 50% NOx control effect on NO3 has a sub-linear response in January (~40% reduction) but a super-linear response in July (~65% reduction) because in the summer the NOx controls not only reduce NO3 precursors but also reduce photochemistry and the NO3 formation rate. The exception to the agreement between M4 and 4Sec response to emission reductions was the effect of SO2 control on NO3, where the NO3 increase (i.e., nitrate replacement) in M4 is greater than seen in 4Sec; this was due to higher SO4 concentrations in M4 presumably because of lower dry deposition rates since SO4 was not allowed to grow into the coarse mode in M4 as it can in 4Sec.

EPISODE AGGREGATION TESTING AND EVALUATION

The uncertainties associated with using episode aggregation to estimate annual average PM$_{2.5}$ concentrations and visibility were analyzed. The uncertainties associated with using less than a year's worth of modeling results to simulate annual average PM$_{2.5}$ concentrations were analyzed using 1999 CAMx modeling results for the eastern US and results for a 1999 Base Case and a 2010 "Clear Skies" control scenario generated by the Lake Michigan Air Directors Consortium (LADCO). The effects of modeling n days in each quarter of the year (n=7, 14 and 28 days) to estimate annual average PM2.5 concentrations were assessed and we found that:

- Relative uncertainties for the 1999 base case annual averages showed wide spatial variability corresponding roughly to spatial variations in annual mean concentrations. Median relative uncertainties over the modeling domain were found to be greater than

±20%, even at n = 28 days. Uncertainties are largest for NO3 and lowest for EC and PI, with mean uncertainties for total $PM_{2.5}$ closer to EC and PI than NO3.

- Restricting attention to just locations with average $PM_{2.5} \geq 15$ µg/m^3 (i.e., exceeding the annual standard) generally lowered the average uncertainties by 40 to 60% (except for BORG and HNO3).

- As expected, uncertainties were found to decrease as n increases. The rate of decline in median uncertainties is somewhat less than the value expected if *individual* days were to have been selected at random from each quarter rather than selecting a single n-day block from each quarter. This difference is most likely due to autocorrelations in the time series of daily PM concentrations.

EPA's draft PM modeling guidance (EPA, 2001) calls for control strategy attainment demonstrations to be based on the use of modeled relative reduction factors (RRFs) which are then applied to monitoring data to obtain the future year annual average. We therefore repeated the Monte Carlo simulation described above but in place of computing the predicted n-day average base case concentration in each quarter, we used the predicted relative reduction factor (RRF) times the "true" base case average in each calendar quarter. Uncertainties in estimated annual averages under the 2010 Clear Skies scenario based on RRFs demonstrated that the <u>uncertainties based on use of RRFs are generally just half or less of the uncertainties for estimating the 1999 base case absolute concentrations</u>. This illustrates the significant advantage of using RRFs in attainment demonstrations rather than absolute model predictions.

A limited evaluation of episode aggregation uncertainty for projecting visibility (Bext) changes was also examined using the July 1996 western US CAMx emission reduction sensitivity tests discussed previously. For some species at some locations, the variation of RRF with Bext is small compared to day-to-day variations suggesting that bias is not a concern and it is better to include all available days in the RRF calculation (so as to get a more precise estimate) instead of restricting attention to just a few modeled 20% best/worst days. In other cases, RRFs appear to be more closely correlated with Bext (at least with the predicted Bext; correlation with observed Bext will in general be weaker). In such cases, the RRF calculation should incorporate the trend with Bext so as to avoid a biased estimate, even though this will result in some loss of precision. Based on our preliminary evaluation with a limited set of model results, it appears that modelers should consider 10 days to be a minimum for estimating RRFs (except for NO3 that exhibits more uncertainty).

RECOMMENDATIONS

The 1996 annual particulate matter (PM) and ozone modeling of the western United States represented the first time the CMAQ, REMSAD and CAMx models were applied, evaluated and intercompared for an annual period using consistent inputs. However, the 1996 database is now dated and further annual modeling analysis should be performed using the more recent 2001 and 2002 databases that are based on improved MM5 simulations and have a larger ambient air quality database for model evaluation.

The episode aggregation results for annual $PM_{2.5}$ support the recommendation that modeling of annual average PM2.5 under future year control scenarios involving changes in SO₂ and VOC emissions should be based on model results obtained for at least 14 days in each calendar quarter. The use of results from just 14 days during each quarter is more reliable when the

model results are just used to compute RRFs as called for in EPA's draft PM modeling guidance. Absolute predictions of base-case annual averages obtained from such a limited set of modeling days can only be considered rough approximations. We recommend that further investigation be performed of NO3 prediction uncertainties and factors driving the extreme variability in NO3 RRFs. We also recommend that the results presented in Section 4 be amended to include analyses of uncertainties under additional control scenarios, especially scenarios involving changes in ammonia emissions.

Only a limited analysis of episode aggregation uncertainties in regional haze modeling could be done with the two months of simplified control strategy runs (single pollutant across-the-board reductions) available to this study. We recommend that additional investigations be performed using a full annual modeling database consisting of a base case and a "realistic" future year multi-pollutant control strategy simulation.

We further recommend that the analysis described above be performed separately for western and eastern US modeling databases so as to properly account for the different conditions associated with haze events in the East as compared to the West.

1.0 INTRODUCTION

The Coordinating Research Council (CRC) sponsored Project A-30 to implement state-of-science particulate matter (PM) modules in Version 3.01 of the Comprehensive Air-quality Model with extensions (CAMx), a photochemical grid model. The resultant model, PMCAMx, was then tested and evaluated using an October 1995 episode for Southern California (ENVIRON, 2003; Yarwood et al., 2003; Morris et al., 2004a). The CRC sponsored two additional projects that used the CAMx model, Project A-44a for "Annual PM Modeling Using a Modified CAMx" and Project A-44b, "Statistical Estimation of Annual PM Using Episodic Predictions". This report summarizes the results of the CRC A-44a/b study that performed annual PM modeling using CAMx and other PM models (i.e., CMAQ and REMSAD) and investigated episode aggregation techniques to address annual $PM_{2.5}$ and visibility modeling. CRC Project A-44 performed the following activities:

- Review, evaluate and recommend statistical methods for episode aggregation to annual averages.

- Implement combine the "full-science" sectional PM modules of PMCAMx abbreviated with 2-section (course/fine) PM modules to develop a PM modeling platform that includes a more computationally efficient long-term version of the model along with the full-science sectional algorithms.

- Perform annual modeling of 1996 and the western US using the new computational efficient version of CAMx and compare model performance with the REMSAD and CMAQ models.

Overview of Approach

At the start of CRC Project A-44, there were two current versions of CAMx available for use, PMCAMx Version 3.01, which included the "full-science" multi-section PM modules developed at Carnegie Mellon University (CMU) and CAMx Version 4 that included an abbreviated "one-atmosphere" 2-section treatment of PM processes. The first work element under CRC Project A-44 was the implementation of the "full-science" CMU PM modules from PMCAMx into the latest CAMx Version 4 code so that the "full-science" and "one-atmosphere" PM treatments are available in the same platform. The resultant integrated code, CAMx Version 4.10, was then tested using the October 1995 episode for Southern California (Koo, Morris and Yarwood, 2003) and the CAMx Version 4.10 model and updated User's Guide (ENVIRON, 2004) was made available on the CAMx website (www.camx.com). These activities are summarized in Section 2 of this report with more details provided in Koo, Morris and Yarwood (2003).

The new integrated CAMx code was then applied using its one-atmosphere 2-section and full-science multi-section modes to the western United States and the 1996 annual period using databases developed by the Western Regional Air Partnership (WRAP) (Tonnesen et. al., 2003). The CAMx annual simulation was evaluated against observations and the model performance compared with that from EPA's Models-3 Community Multi-scale Air Quality (CMAQ, Version 4.3) modeling system and the Regional Modeling System for Aerosols and Deposition (REMSAD Version 7). The results of the 1996 CAMx, CMAQ and REMSAD model evaluation

and intercomparison are documented in Koo and Morris (2004) as well as in an independent evaluation report prepared by Seigneur and co-workers (2004). The effects of emission reductions on the CAMx modeling results using the 2-section and multi-section PM modules were also examined (Koo and Morris, 2004). The 1996 annual application and model performance application are summarized in Section 3.

Also under CRC Project A-44, the uncertainties associated with using episode aggregation to estimate annual average $PM_{2.5}$ concentrations and visibility were analyzed. The uncertainties associated with using less than a year's worth of modeling results to simulate annual average $PM_{2.5}$ concentrations were analyzed using 1999 CAMx modeling results for the eastern US, whereas the effects of episode aggregation on visibility was examined using the 1996 western US CAMx modeling results discussed previously. These results are summarized in Section 4 of this report with details provided in Stoeckenius (2004).

Section 5 of this report summarizes the recommendations of the CRC Project A-44 study.

2.0 INTEGRATION OF CAMx WITH PMCAMx

The base model used in this work was Version 4.03 of the Comprehensive Air-quality Model with extensions (CAMx), which has a "one-atmosphere" treatment for ozone and particulate matter (PM). The PM treatment implemented in the CAMx Version 4.03, called Mechanism 4 (M4), consists of the following three modules:

- RADM – bulk aqueous-phase chemistry module similar to one used in CMAQ
- ISORROPIA – inorganic aerosol thermodynamic model
- SOAP – secondary organic aerosol module

M4 uses 2-section PM approach whereby primary species are modeled as fine and/or coarse particles and all secondary PM species are modeled as fine particles.

Under CRC Project A-44, "full-science" aerosol modules were implemented in CAMx Version 4.03 using the aerosol modules implemented in PMCAMx that were developed by Carnegie Mellon University (CMU). These "full-science" aerosol modules include the following:

- VSRM – variable size resolution aqueous-phase chemistry module where either bulk (i.e., 1-section) or 2-section size-resolved approach is executed based on input conditions
- EQUI – size-resolved equilibrium aerosol module where the bulk amount of each species transferred between gas and aerosol phases is determined by ISORROPIA and is distributed over the aerosol size sections by using a weighting scheme
- MADM – multi-component aerosol dynamic module where the mass transfer rates of aerosol species for each size section are determined by a fundamental flux equation; Dr. Chock's Trajectory-Grid (T-G) method has been implemented to replace LSODE
- HYBR – hybrid aerosol module where EQUI is used for the smaller sections and MADM for the larger sections
- SOA module – EQUI uses the same SOAP as M4, but with an improved weighting scheme to distribute SOA onto the aerosol size sections; MADM uses an integrated dynamic SOA module

The new version of CAMx (Version 4.10), available at www.CAMx.com, includes both the "one-atmosphere" 2-section M4 and "full-science" multi-section aerosol modules and has several options. The following CAMx Version 4.1 model configurations were tested using the October 1995 Southern California database in this study:

- M4 (Mechanism 4 2-section treatment with RADM aqueous-phase chemistry)
- RADM with EQUI (called RADM/EQ)
- VSRM with either of EQUI, HYBR, or MADM (called VSRM/EQ, VSRM/HY and VSRM/MA)

CAMx VERSION 4.10 TESTING AND EVALUATION

The aerosol modules in the new CAMx Version 4.10 were tested for an October 17-18, 1995 episode in the Los Angeles area, which has been used for the initial PMCAMx test (ENVIRON 2003; Yarwood et al., 2003). The modeling domain is defined in UTM coordinates with 65 by 40, 5-km grid cells. The vertical grid consists of 10 layers with layer tops at 20, 50, 100, 250, 500, 750, 1000, 1500, 2000, 2300 meters.

Emissions and initial/boundary conditions were originally developed for PMCAMx (ENVIRON, 2003) and thus aerosol species are size-resolved. A pre-processing tool was developed to convert the size-resolved emissions, IC, and BC inputs to those compatible with CAMx M4 2-section treat of PM size distribution.

Results

CAMx was operated for the October 1995 episode with the five configurations of aerosol modules discussed above (i.e., M4, RADM/EQ VSRM/EQ, VSRM/HY and VSRM/MA) to generate hourly three-dimensional fields of ozone, NOx, VOC and PM species concentrations.

Ozone

Hourly average ozone peak concentrations predicted by the five CAMx configurations agree quite well (Figure 2-1). The discrepancy between M4 and EQUI is mostly attributed to the difference between the RADM and VSRM aqueous-phase chemistry modules. The differences between predictions by EQUI, HYBR, and MADM are negligible when using the same aqueous-phase chemistry module, which is not surprising as the treatment of aerosol size distribution has little feedback into ozone chemistry.

Particulate Matter

Daily average total $PM_{2.5}$ predictions using the different CAMx configurations exhibit significant differences in some regions (Figure 2-2). Figures 2-3 and 2-4 display the predicted 24-hour averaged size distributions at cells where maximum positive and negative, respectively, differences between the M4 and VSRM/EQ CAMx configurations occur. Figures 2-5 and 2-6 display similar differences between the VSRM/EQ and VSRM/MA CAMx configurations. The results in these figures can be summarized as follows:

- M4 predicts more fine PM than the size-resolved PM modules due to lower deposition rates and because secondary PM species are assumed to always be fine in M4.
- In general, VSRM predicts more fine sulfate than RADM.
- Nitrate (and/or ammonium) predicted by models with VSRM and RADM can be significantly different, which causes the positive or negative PM differences between the corresponding models.
- In general, the equilibrium model predicts more fine ammonium nitrate than the dynamic model.
- Differences in size distributions predicted by HYBR and MADM over the area around cell (50, 28) are unexpected. Further investigation is needed.

Size Resolution

The simulation was repeated with RADM/EQ using 4 sections rather than 10 sections. Table 2-1 shows the sectional boundary diameters used with 4 and 10 sections. Size distributions predicted by RADM/EQ with 10 and 4 sections give relatively good agreement (Figure 2-7). While peak locations of nitrate and ammonium are slightly different, their $PM_{2.5}$ and PM_{10} predictions agree well.

Table 2-1. Aerosol size section boundaries.

Section	RADM/EQ (10 sections)	RADM/EQ4 (4 sections)
1	0.039063 – 0.078125	0.039063 – 0.15625
2	0.078125 – 0.15625	0.15625 – 0.625
3	0.15625 – 0.3125	0.625 – 2.5
4	0.3125 – 0.625	2.5 – 10
5	0.625 – 1.25	
6	1.25 – 2.5	
7	2.5 – 5	
8	5 – 10	
9	10 – 20	
10	20 – 40	

Efficiency

Figure 2-8 displays CPU hours per simulation day for the models. Times are for one Athlon 1600 CPU. As expected, M4 is the most computationally efficient while the CPU time required by RADM/EQ with 4 sections is slightly slower (~20%) to that of M4. The use of the VSRM aqueous-phase chemistry module results in significant increases on the model run times (~ factor of 5) over using the RADM module.

RECOMMENDATIONS

The VSRM aqueous-phase chemistry module is much slower than RADM and its computational costs are not uniformly distributed. Also VSRM failed to converge at several cells. Further refinement of the model is needed before it is ready to operate for longer periods.

For annual simulations where computational requirements are important, the M4 and/or RADM/EQ CAMx configurations are most computationally efficient. RADM/EQ is as stable and fast (when using 4 sections) as M4 and can give more detailed information for the aerosol size distribution with only a small computationally penalty (~20% slower) based on the October 1995 episode testing for Southern California.
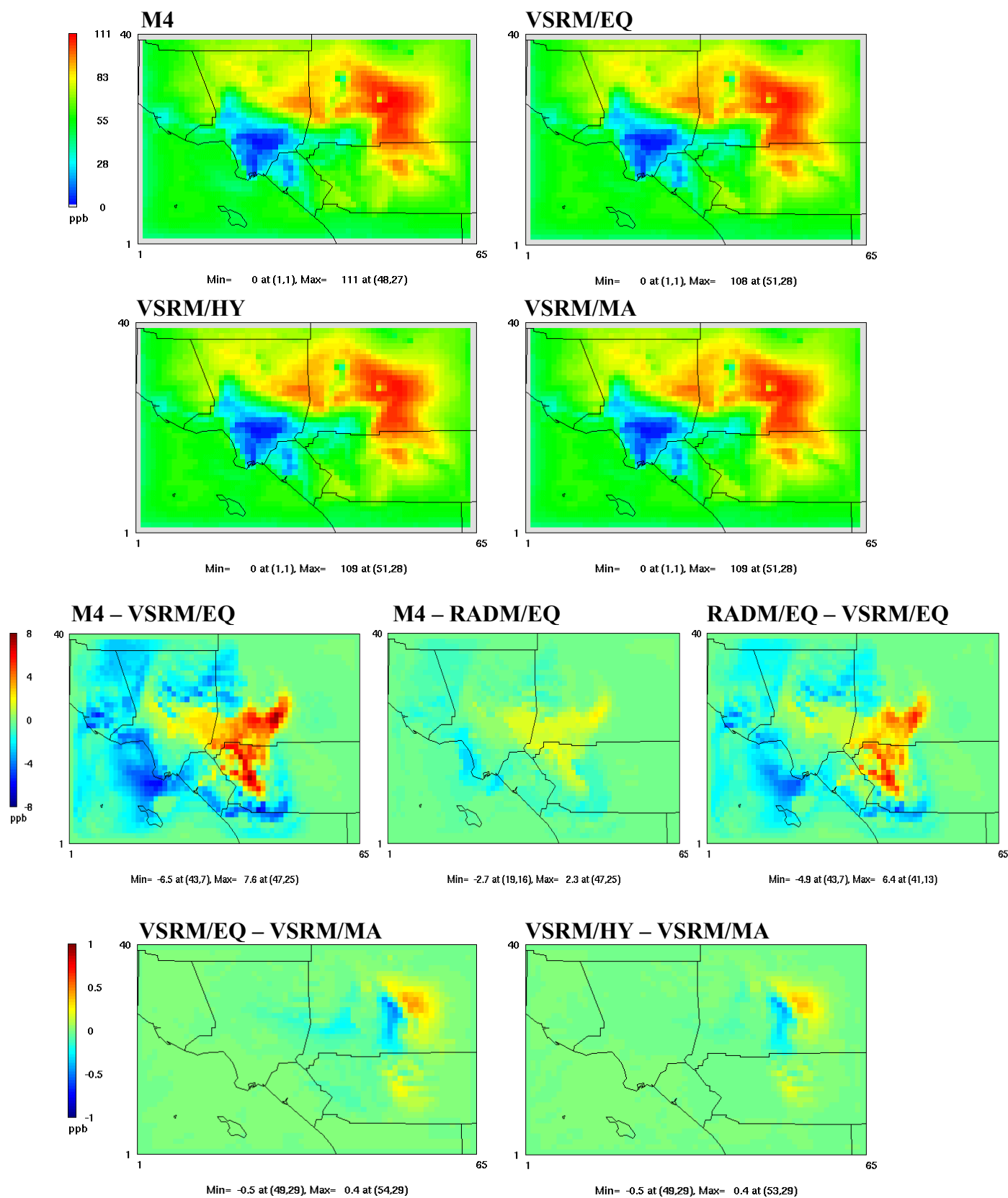
Figure 2-1. Hourly average peak ozone estimates in Southern California on October 18, 1995 for the various CAMx PM configurations.
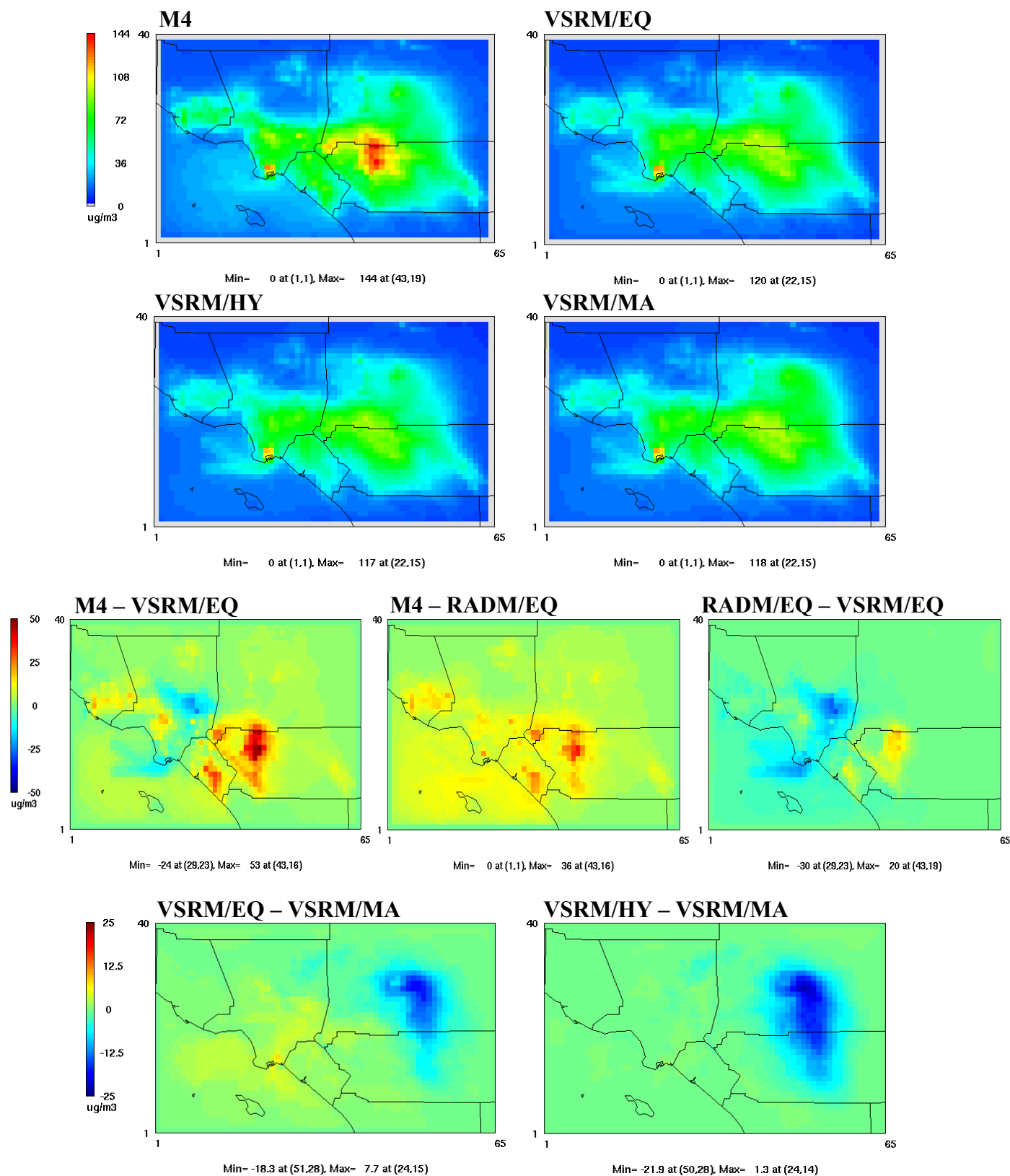
Figure 2-2. Daily average total PM$_{2.5}$ mass estimates in Southern California on October 18, 1995 for the various CAMx PM configurations.
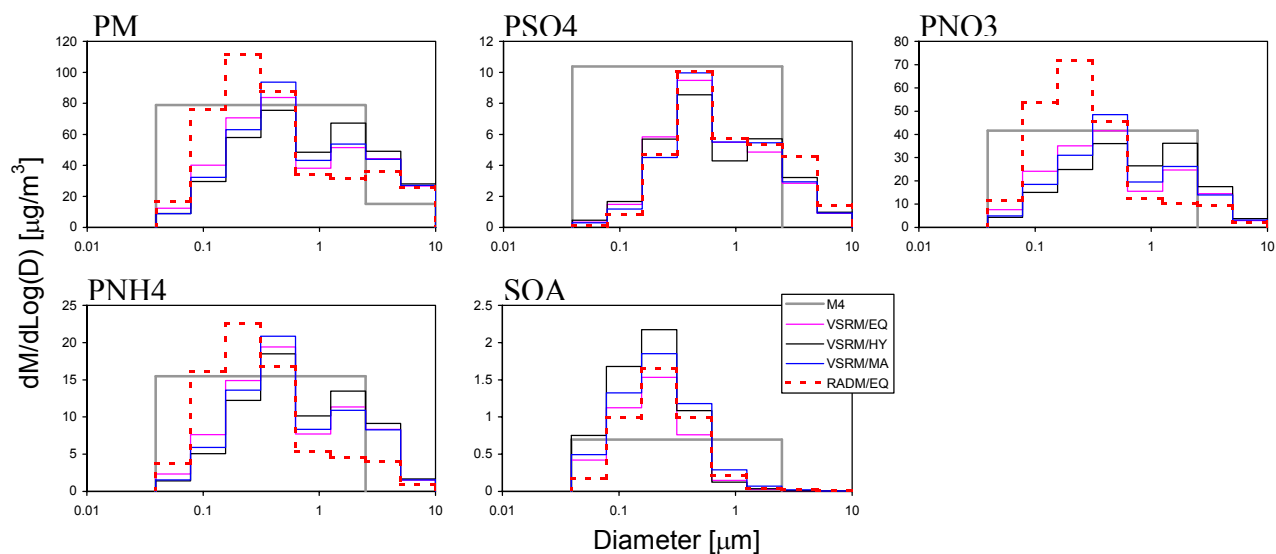
Figure 2-3. Daily average PM size distributions predicted at cell (43, 16) for the various CAMx PM configurations.
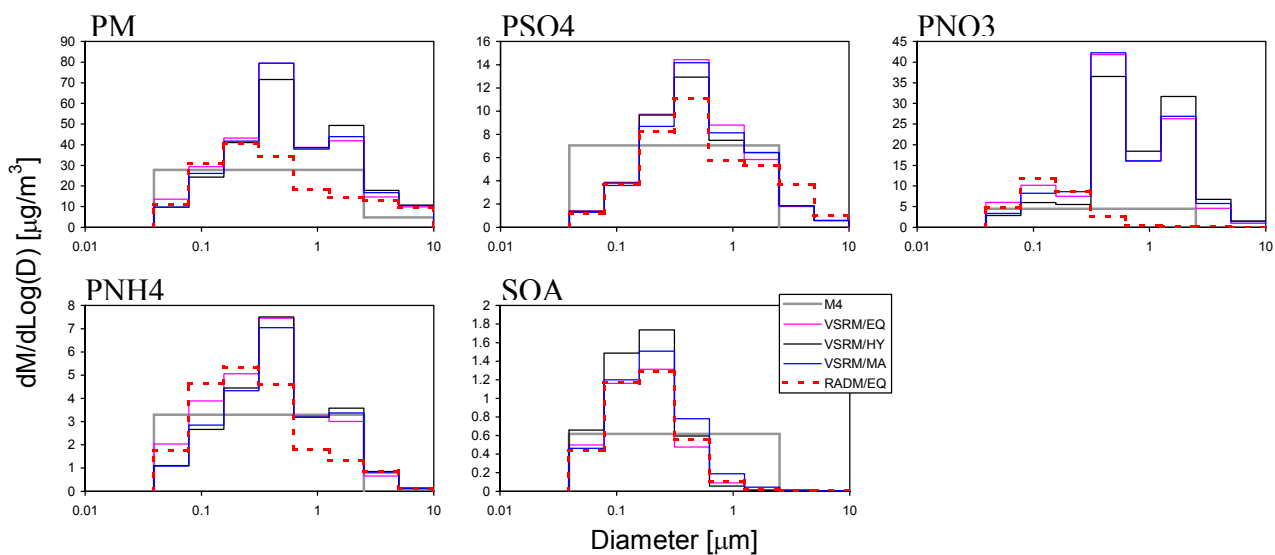
Figure 2-4. Daily average size distributions predicted at cell (29, 23) for the various CAMx PM configurations.
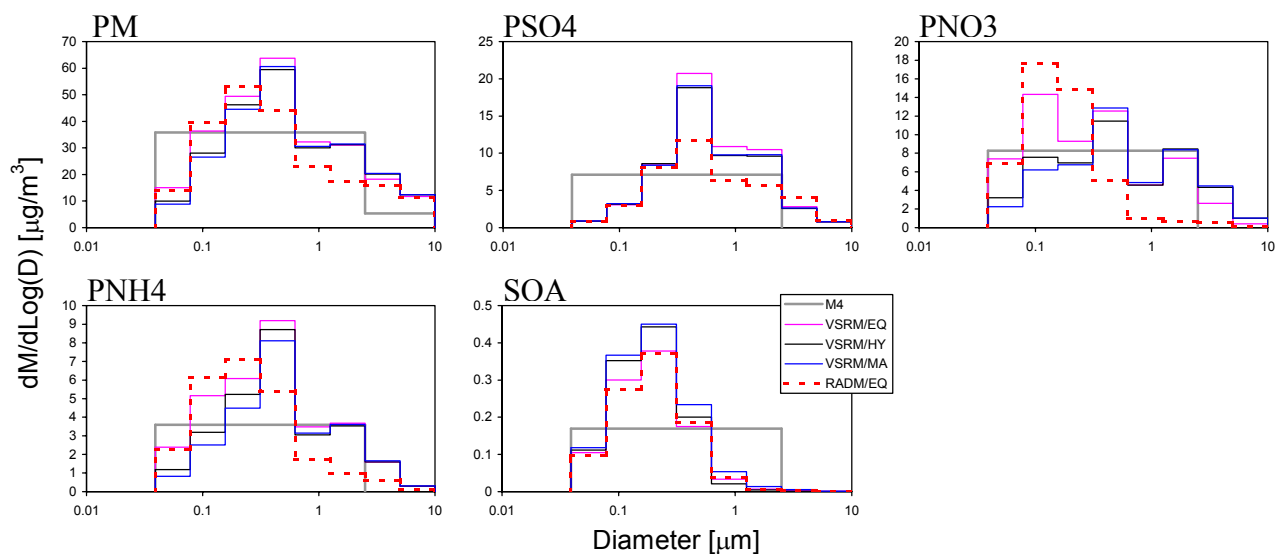
Figure 2-5. Daily average size distributions predicted at cell (24, 15) for the various CAMx PM configurations.
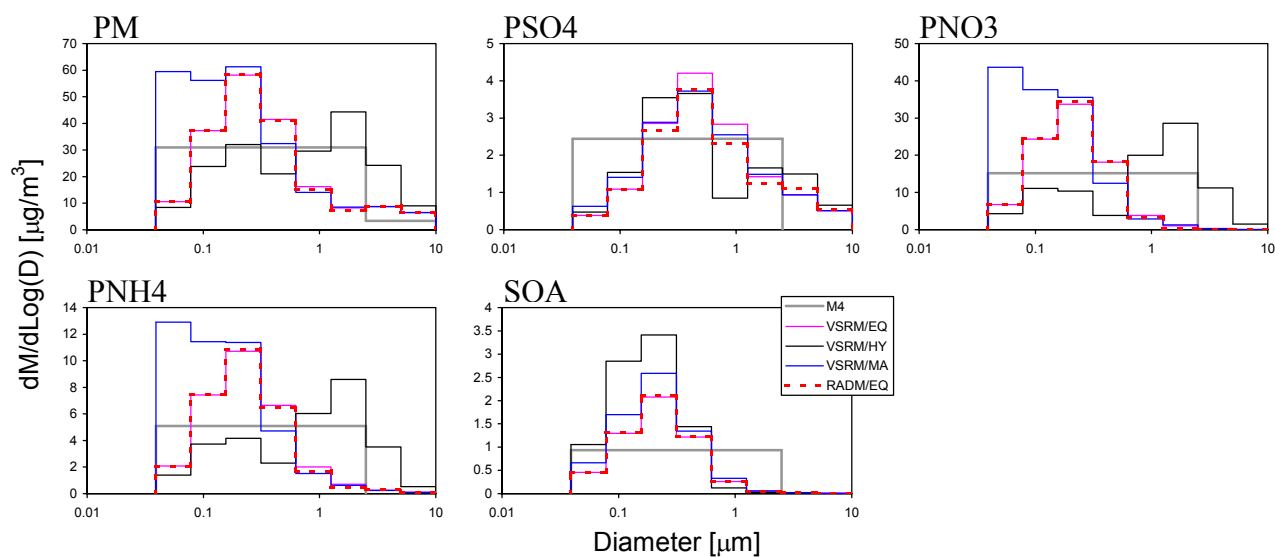
Figure 2-6. Daily average size distributions predicted at cell (50, 28) for the various CAMx PM configurations.
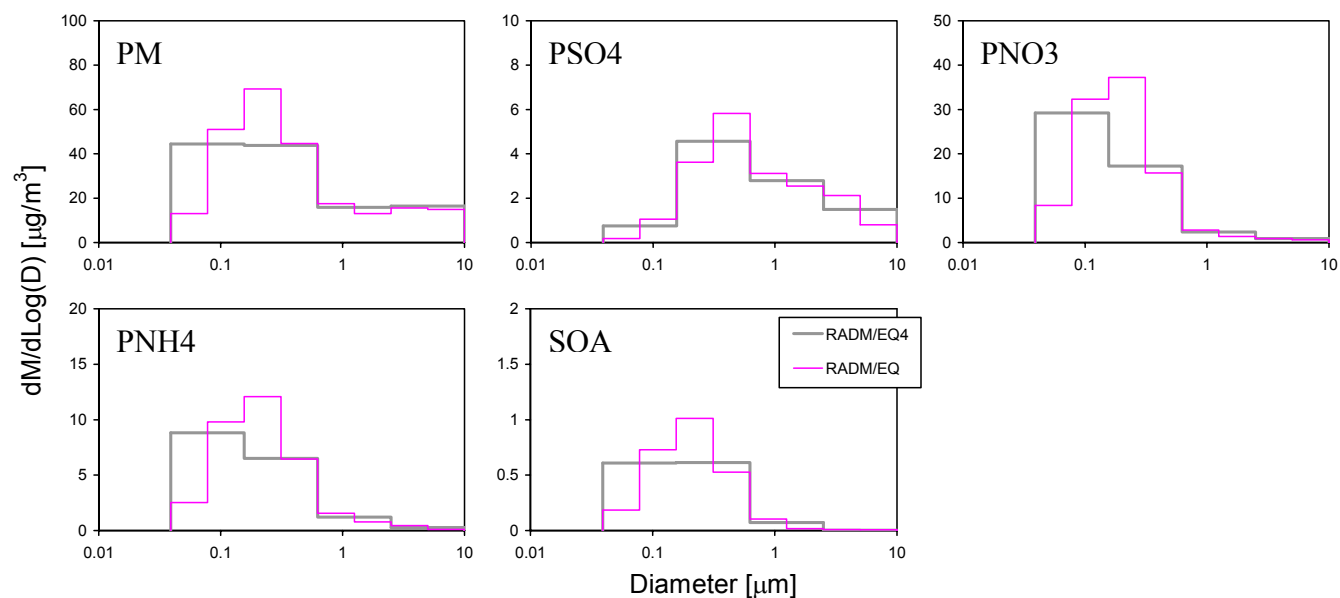
Figure 2-7. PM size distributions predicted by CAMx RADM/EQ with 4 and 10 sections at cell (34, 16).
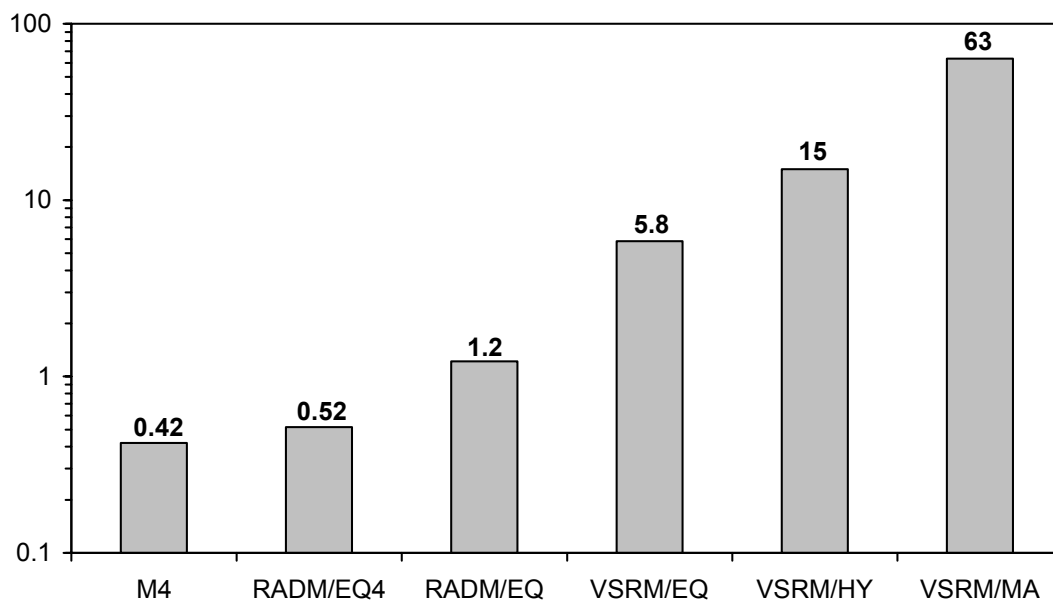
Figure 2-8. CPU hours per simulation day for the various CAMx PM configurations.

3.0 1996 ANNUAL MODELING

This section discusses the development of the 1996 annual CAMx inputs for full-science sectional and one-atmosphere Mechanism 4 PM modeling, the comparative model evaluation for CAMx, CMAQ and REMSAD, and the CAMx emission reduction sensitivity tests. A beta version of CAMx Version 4.10 was used in the 1996 annual modeling of the western US. Two updates to the beta version of the model were made prior to the release of the official CAMx Version 4.10 on the CAMx website (**www.camx.com**) in August 2004: (1) update to the MM5CAMx processor to provide a better and more consistent treatment of clouds and precipitation including sub-grid scale cloud processes in MM5; and (2) update to the CAMx wet deposition scheme. The updates primarily affect sulfate. To evaluate whether these updates would have a significant affect on the 1996 annual modeling, the July 1996 period was rerun with the final CAMx Version 4.10 and compared against the CAMx Version 4.10beta results used in the annual modeling.

DEVELOPMENT OF 1996 ANNUAL INPUTS FOR CAMx

The WRAP developed CMAQ 1996 annual 36 km inputs for the western US modeling domain (see Figure 3-1) that were used by WRAP in their Section 309 SIP modeling (WRAP, 2003; Tonnesen et al., 2003). Below we describe how CAMx and REMSAD inputs were developed consistent with the WRAP CMAQ 1996 inputs, more details can be found in Koo and Morris (2004).

CMAQ Emissions and IC/BC Conversion to REMSAD and CAMx

CMAQ-to-CAMx and CMAQ-to-REMSAD interface programs were developed to convert CMAQ emissions and initial concentrations and boundary conditions (IC/BC) input files into either REMSAD (version 7) or CAMx (Mechanism 4 and sectional PM module). These programs consist of two steps: first they convert the Models-3 I/O API files into UAM format (binary Fortran) which is used for REMSAD and CAMx; then it maps the CMAQ species to either the REMSAD or CAMx species.

The emission processors separate the CMAQ 3-D emission inputs into a 2-D low-level emissions source file, that includes emissions from on-road and off-road mobile sources, area sources, biogenic sources and point sources whose plume rise release the emissions in the first layer, and an elevated point source file that includes all emissions that are released above layer 1. All the grid cells between the second layer and the top layer that have non-zero emissions of any species during any time period in the file are assumed to have a point source. The vertical layer index, (k-index) of each hypothetical point source, is stored in the output file as well as X and Y coordinates (i, j) of the source, which is assumed to be located at the mid point of the cell. The k-index is then converted to the actual height of the layer and stored as an effective plume-height (as negative value). Plume-rise calculations in CAMx and REMSAD are disabled when the emissions input has negative plume-height value. All of the CMAQ-to-CAMx, CMAQ-to-REMSAD and modified CAMx and REMSAD codes are considered publicly available.

Note that CAMx includes separate species for fine and coarse crustal material (dust) that has no counterpart in CMAQ. Thus, crustal material was not modeled as a separate species in the CAMx 1996 annual modeling as such information was not available in the CMAQ emission inputs.

1996 Western US Annual Modeling

The western US modeling domain consists of 85 columns and 95 rows with grid cell size of 36 km as depicted in Figure 3-1. The MM5 version 2.12 was used to generate a set of annual meteorological data for 1996. The simulations consisted of multiple nested domains of 108-km and 36-km horizontal resolution on a Lambert Conformal Projection (LCP) coordinate system with 23 vertical sigma layers extending from the surface to the 100mb pressure level. The MM5 outputs were processed by the CMAQ MCIP, MM5CAMx and MM5REMSAD processors to prepare the meteorological inputs for CMAQ, CAMx and REMSAD, respectively. The 23 MM5 vertical layers were collapsed to 18 layers for the air quality modeling. The same horizontal and vertical model configuration was used in all three air quality models for the 1996 annual modeling; the use of identical grid and larger definitions, if possible, is a necessary component when conducting a model comparison study.

Table 3-1 displays the science algorithms selected for the CMAQ, REMSAD and CAMx simulations. CMAQ and CAMx both used the CB4 gas-phase chemistry, RADM aqueous-phase chemistry and ISORROPIA aerosol equilibrium modules, whereas REMSAD used the abbreviated micro-CB4 gas-phase and Martin's aqueous-phase chemistry and simpler MARS-A equilibrium modules. CAMx was configured using both the "one-atmosphere" Mechanism 4 (M4) 2-section (coarse/fine) and the "full-science" multisection (4sec) configurations.

Table 3-1. Science algorithms selected for annual 1996 modeling.

	CMAQ	REMSAD	CAMx_M4	CAMx_4sec
Gas-phase	CB4	Micro-CB4	CB4	CB4
Inorganic	ISORROPIA	MARS-A	ISORROPIA	ISORROPIA
Organic	SORGAM	Odum et al. (1997) Griffin et al. (1999)	SOAP	SOAP
Aqueous	RADM	Martin (1984)	RADM	RADM
Size	3 modes	Fine/Coarse	Fine/Coarse	4 sections
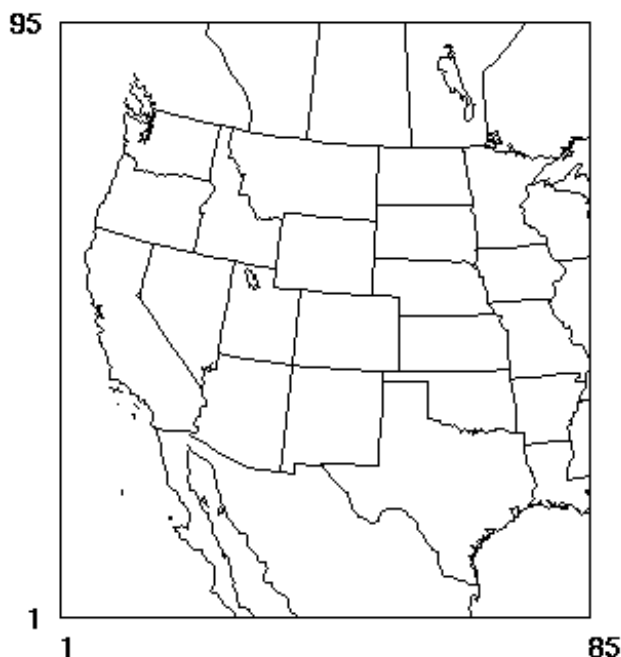
Figure 3-1. Western US 36 km modeling domain used in the 1996 annual modeling

1996 MODELING RESULTS

Major PM components predicted by the four models were evaluated against monitoring data from the IMPROVE and CASTNet networks. Below we describe the PM model performance using the IMPROVE data (Figure 3-2), results using the CASTNet data are similar and can be found in Koo and Morris (2004) and Seigneur et al., (2004). Wet deposition performance was evaluated by comparing modeling results to observations taken from the NADP monitoring network (Figure 3-3). Results are presented for mean fractional bias and mean fractional gross error for the 1996 annual simulation and then separately for January and July 1996. As part of the VISTAS and WRAP modeling, we have found that the fractional bias and gross error provide the most robust and descriptive assessment of summary model performance of the model performance statistic metrics analyzed (Morris et al., 2004a,b,c; Tonnesen et al., 2003) so they are presented here.

Fine Sulfate (SO4)

All the models exhibit an approximately 50-60% fractional gross errors for sulfate over the entire year of 1996 (Figure 3-2). The magnitude of the fractional error is larger in January than in July. The models tend to over-predict fine sulfate in January and under-predict in July making annual fractional biases look better than the monthly values (i.e., the annual performance statistics for sulfate bias suffer from compensatory errors whereby the winter over-prediction is compensated for by the summer under-prediction.)

Based on the annual statistics, under-prediction in fine sulfate by CAMx_4sec is the most significant, which may be due to its sectional representation of PM size distribution. Unlike the other models, CAMx_4sec allows the secondary PM species (e.g., sulfate and nitrate) to grow

into the coarse mode where it deposits out more quickly. The difference in fine sulfate predicted by CAMx_M4 and CAMx_4sec indicates that a significant fraction of total sulfate is in the coarse mode. Figure 3-4 suggests that about 10% of sulfate predicted by CAMx_4sec lie in the 4th (coarse) section. One of the reasons for the coarse sulfate is the aqueous chemistry module. In this work, CAMx_4sec employs the RADM module, which is a bulk (i.e., one size section) aqueous chemistry module. When RADM calculates the total aerosol mass produced by aqueous chemistry, the mass is then distributed into each size section using pre-defined weighting factors developed by CMU. Model sensitivity to this weighting factor is discussed below.

Fine Nitrate (NO3)

Among the major PM components, nitrate shows the largest fractional errors (over 100% in all the models). Annual fractional biases look much better, but individual months show large positive (winter months) and negative (summer months) fractional biases. Overall, CAMx_4sec has smaller nitrate fractional biases than other models, whereas CMAQ exhibits lower errors. However, the performance of all four models is poor and this is an area of more research and refinement of model formulation and inputs to achieve improved model performance.

Fine Ammonium (NH4)

Since IMPROVE doesn't provide direct measurement of particulate ammonium, the evaluation is based on only the CASTNet database (see Koo and Morris, 2004). Annual fractional gross errors for ammonium are less than 65% for all the models. Again the errors are larger in January than in July. From the fractional biases, it appears that CAMx models tend to over-predict fine ammonium for the year and especially in January. The lower annual bias for CMAQ and REMSAD appears to be due to summer under-prediction compensating for winter over-prediction bias.

Organic Carbon (OC)

All the models under-predict organic carbon mass. It should be noted that the models treat primary organics differently in their SOA formations. Primary organics in the CAMx model do not make a solution with primary and secondary organics. In CMAQ and REMSAD, however, they serve as an absorbing organic material, which results in more SOA formation. This can partly explain why CAMx models show lower (larger negative) fractional biases for organics than CMAQ and REMSAD.

Other PM Species (EC, SOIL and COARS)

In all the models, PM species like EC and SOIL (other PM2.5) do not participate in chemistry. Therefore, they are affected only by emissions, depositions, and transport. Yet the model performances for the species are not very promising. There are likely emission inventory issues related to the model performance for these species.

COARS ($PM_{2.5-10}$) defines the particulate mass in the coarse mode (usually PM_{10}-$PM_{2.5}$). All the models show negative fractional biases for COARS. CAMx_4sec is different from other models in that some of the secondary particles can contribute to the coarse mode, which helped improve its performance for COARS.

Wet Deposition

All the models tend to under-estimate wet removal of PM species (Figure 3-3). CMAQ performed better than the others. Under-prediction of wet deposition flux by the CAMx model is more significant in July than in January. It should be noted that recent updates in the final CAMx Version 4.10 that improve cloud-precipitation reconciliation algorithm in MM5CAMx has resulted in a significant increase in the aerosol wet removal rates and improved sulfate model performance (Baker, 2003) over the CAMx Version 4.10beta used in the 1996 annual modeling. After the completion of the 1996 annual modeling, the final CAMx Version 4.10 was tested for July 1996 and compared against the CAMx version 4.10beta results used in the 1996 annual modeling. These results are discussed at the end of this Section.

(a) SO$_4$

(b) NO$_3$

(c) OC

(d) EC
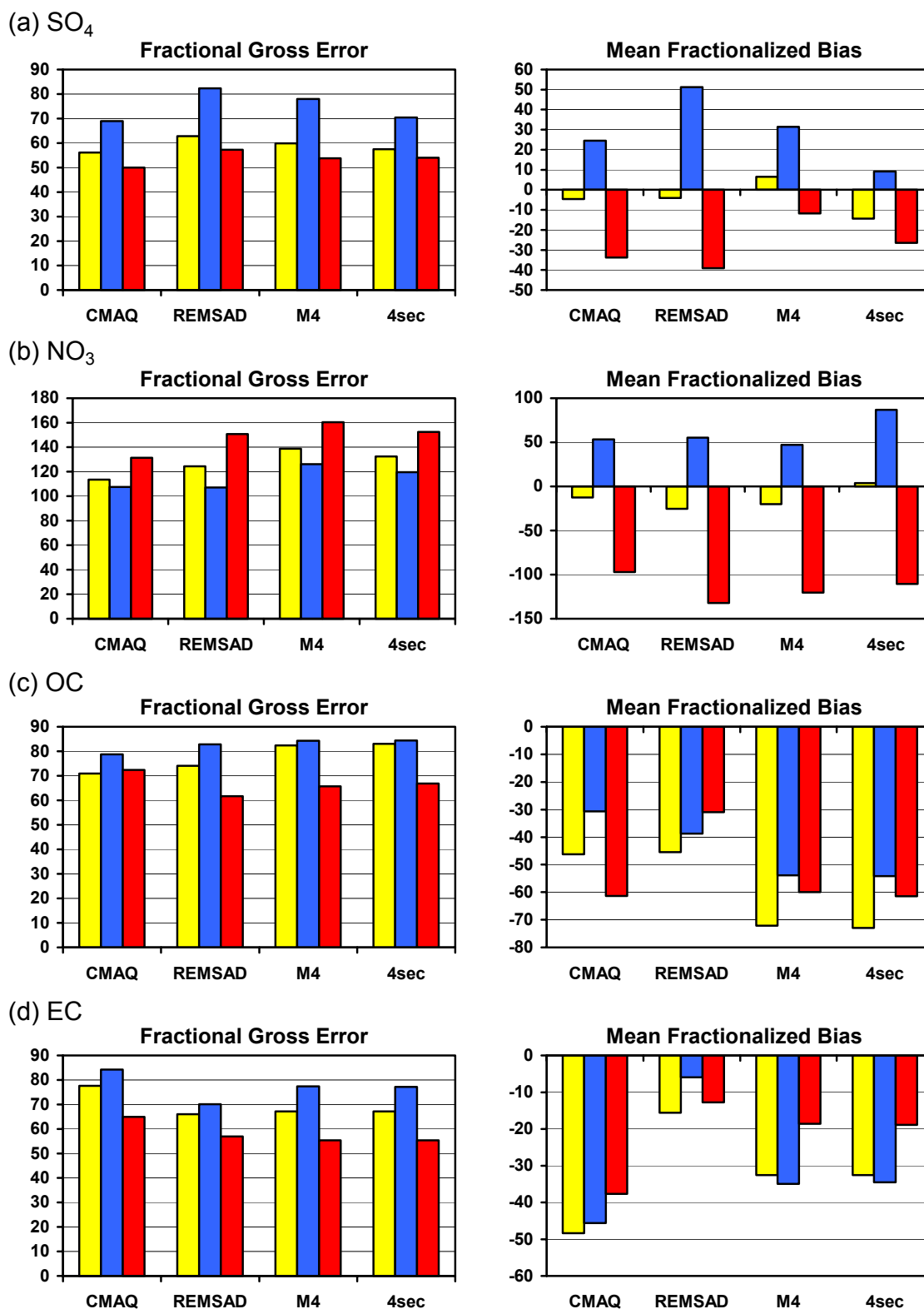
Figure 3-2a. Evaluation of models against IMPROVE observation data for the whole year of 1996 (yellow), January (blue) and July (red).

(e) SOIL
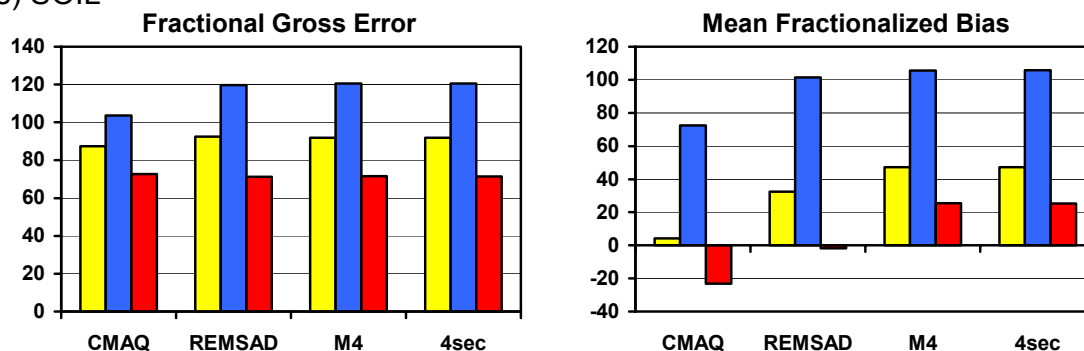
(f) COARS
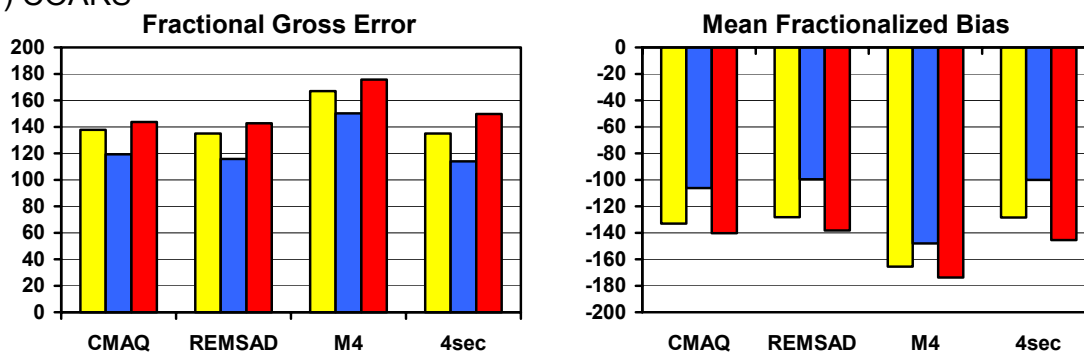
(g) PM$_{2.5}$
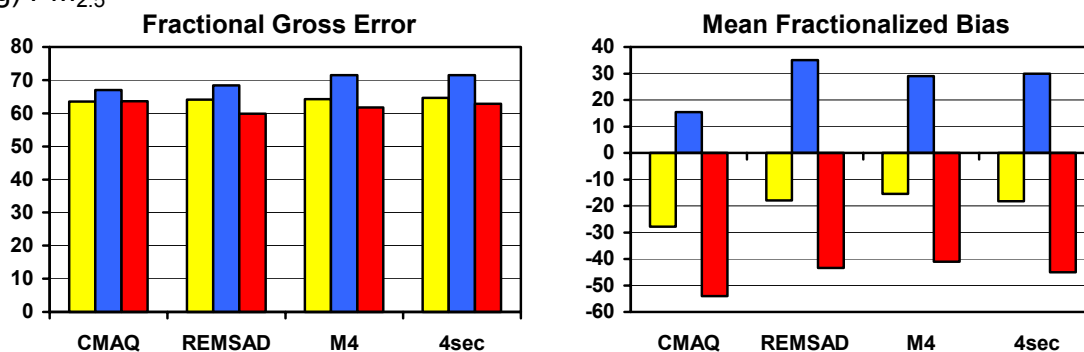
(h) PM$_{10}$
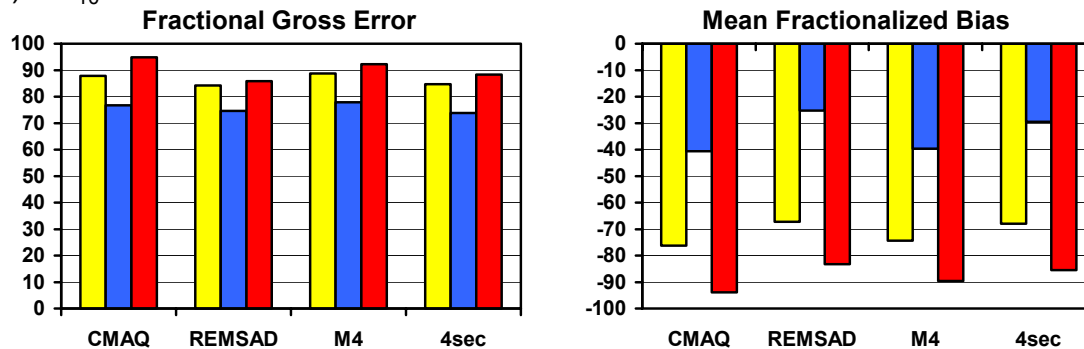
Figure 3-2b. Evaluation of models against IMPROVE observation data for the whole year of 1996 (yellow), January (blue) and July (red).

(a) SO$_4$ wetdep
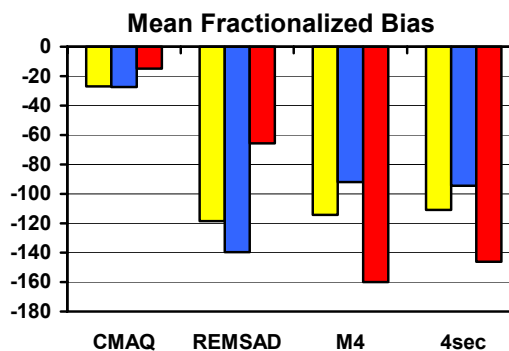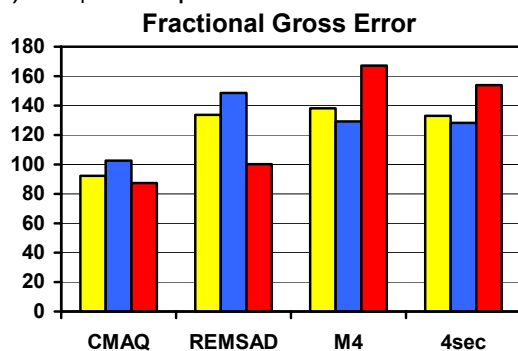
(b) NO$_3$ wetdep
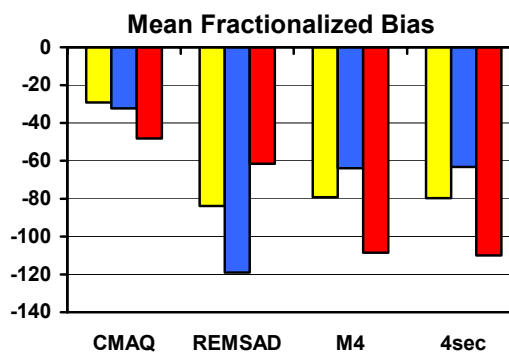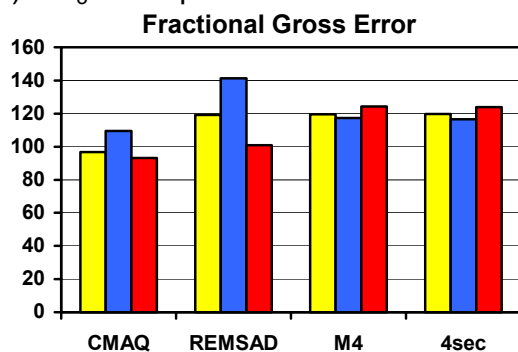
(c) NH$_4$ wetdep
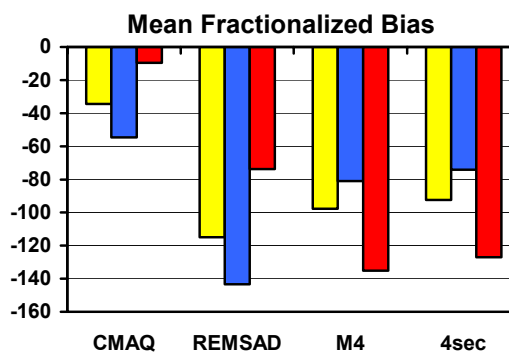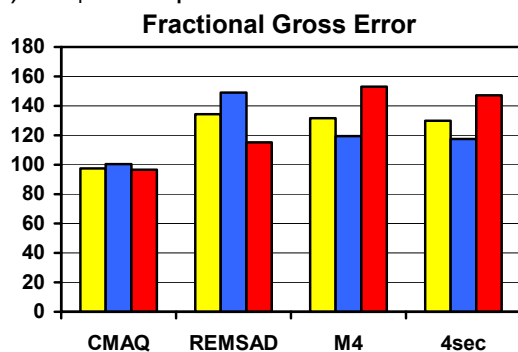
Figure 3-3. Evaluation of models against NADP observation data for the whole year of 1996 (yellow), January (blue) and July (red).
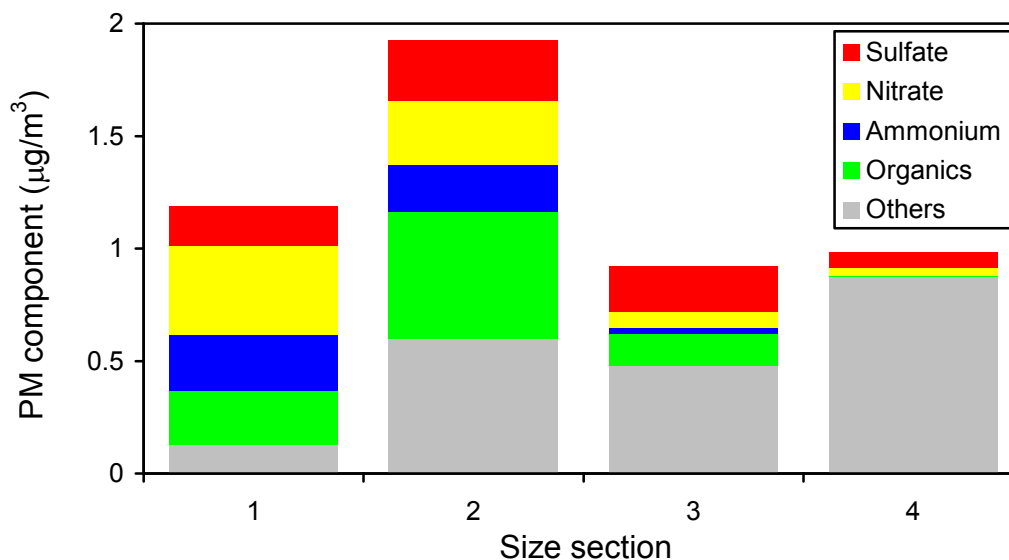
Figure 3-4. Annual average PM size distribution predicted by CAMx_4sec over IMPROVE monitoring sites.

SENSITIVITY TO EMISSION CHANGES

A set of 4 emission sensitivity tests for the January and July 1996 periods was performed using CAMx_M4 and CAMx_4sec configurations to assess whether the level of science modules affects how the model responds to changes in emissions. The 4 emissions reductions sensitivity tests are:

    (1) 50% NO_x control;
    (2) 50% VOC control;
    (3) 50% NH_3 control; and
    (4) 50% SO_2 control.

Table 3-2 displays the summary of model responses to the emissions reduction sensitivity scenarios averaged across monitors in the IMPROVE network. The predicted responses by CAMx_M4 and CAMx_4sec are in good agreement with each other, except for the SO_2 emissions control case. With 50% reduction in domain-wide SO_2 emissions, both models predicted fine sulfate would decrease by 15% in January and 26% in July. In CAMx_M4, this frees up ammonium and allows for increased levels of particulate ammonium nitrate formation of 28% in January and 14% in July 1996 (i.e., nitrate replacement). In CAMx_4sec, however, this nitrate ammonium formation is more limited with only a 7% increase in particulate nitrate in January and no change in July. The primary reason for these differences in the nitrate replacement between CAMx_M4 and CAMx_4sec is due to the higher SO4 concentrations in CAMx_M4, which is presumably due to more SO4 dry deposition loss in CAMx_4sec due to allowing the SO4 to grow into the coarse mode. Less SO4 in CAMx_4sec results in higher nitrate concentrations. Therefore, further nitrate replacement due to the SO2 control is not favorable thermodynamically in CAMx_4sec.

CAMx_M4 and CAMx_4sec nitrate exhibits similar sensitivity to NOx control, with a 50% NOx control resulting in a 41% (January) and ~65% (July) reduction in particulate nitrate. It is interesting that nitrate response to NOx control is sublinear in the winter and superlinear in the summer; in the summer, the 50% NOx controls not only reduces the available nitrate precursor by 50% but also reduces photochemical activity and therefore the nitrate formation rate resulting in the superlinear (~65%) response at the rural IMPROVE monitors. Note that a different response signal may occur in the urban atmosphere. The CAMx_M4 and CAMx_4sec nitrate estimates exhibit slightly different responses to ammonia controls, with CAMx_M4 being slightly more sensitive than CAMx_4sec.

Another interesting finding in Table 3-2 is that the VOC controls results in small increases in sulfate and nitrate. This is due to the competition between VOCs and sulfate/nitrate for the hydroxyl (OH) radical at these rural sites. Reducing VOC emissions reduces OH destruction by the VOC+OH reaction thereby leaving more OH available to react with SO2 and NO2 to form sulfate and nitrate, respectively.

Table 3-2a. Model responses (% change) to the selected emissions reductions.

	50% NO$_x$ control				50% VOC control			
	January		July		January		July	
	M4	4sec	M4	4sec	M4	4sec	M4	4sec
Peak ozone	2.4	2.4	-13	-13	-1.0	-1.0	-2.8	-2.8
Fine sulfate	2.6	1.3	-5.7	-7.0	0.20	0.37	3.5	4.1
Fine nitrate	-41	-41	-64	-65	1.9	2.5	11	12
Fine ammonium	-19	-22	-7.4	-11	0.90	1.3	3.1	4.2
Fine SOA	1.5	1.5	-5.9	-5.9	-39	-39	-9.9	-9.9
Total PM$_{2.5}$	-11	-13	-5.7	-6.6	-0.97	-0.69	0.34	0.64

Table 3-2b. Model responses (% change) to the selected emissions reductions.

	50% NH$_3$ control				50% SO$_2$ control			
	January		July		January		July	
	M4	4sec	M4	4sec	M4	4sec	M4	4sec
Peak ozone	0.004	0.006	0.048	0.060	-0.007	-0.005	-0.050	-0.049
Fine sulfate	-1.6	-1.3	-1.8	-1.6	-15	-14	-26	-26
Fine nitrate	-58	-42	-76	-68	28	7.1	14	0.27
Fine ammonium	-37	-33	-23	-11	3.5	-3.1	-16	-23
Fine SOA	-0.003	-0.15	0.017	0.003	0.11	0.13	0.011	0.002
Total PM$_{2.5}$	-16	-16	-5.5	-4.3	-0.65	-1.2	-10	-10

SIZE DISTRIBUTION OF AEROSOLS FROM AQUEOUS CHEMISTRY

As mentioned earlier, a pre-defined factor determines the size distribution of aerosols from the RADM bulk (i.e., one size section) aqueous chemistry in CAMx_4sec. Unlike CAMx_M4, where all the secondary aerosols are in fine mode, CAMx_4sec assigns half of the total aerosol mass from the aqueous chemistry into the coarse mode based on analysis by CMU (Figure 3-5). We modified the distribution factors so that no aerosol mass from the aqueous chemistry should be assigned to size bins larger than 2.5μm and repeated the base case simulation for January 1996. Figure 3-6 shows that the difference in fine sulfate predicted by CAMx_M4 and CAMx_4sec was mainly due to the aqueous chemistry. With the modified distribution factors,

the level of fine sulfate predicted by CAMx_4sec becomes similar to that by CAMx_M4. The changes in fine nitrate and ammonium due to the modified distribution factors are not significant.
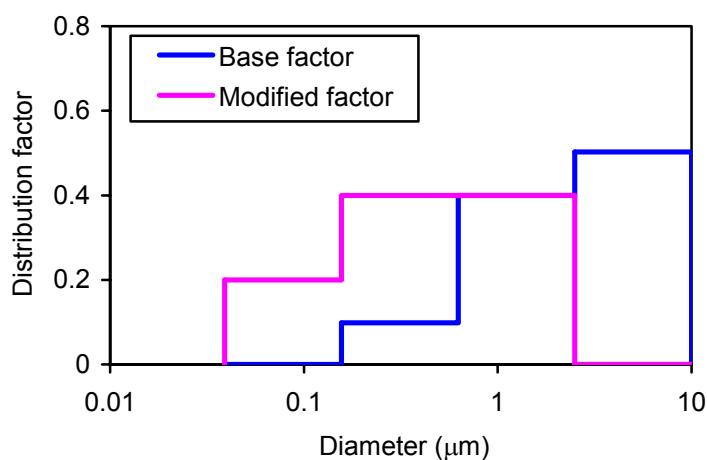
Figure 3-5. Size distribution factors for aerosols from aqueous chemistry.
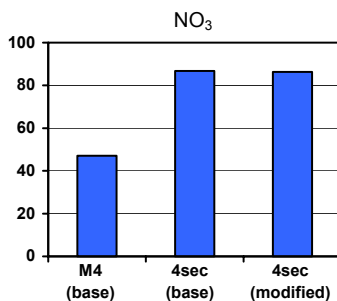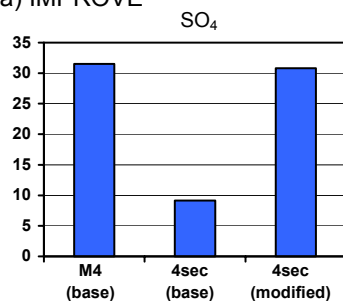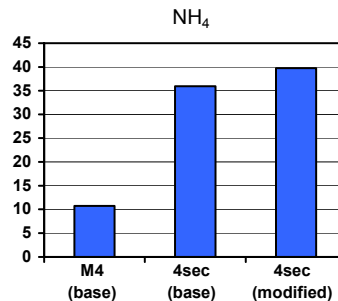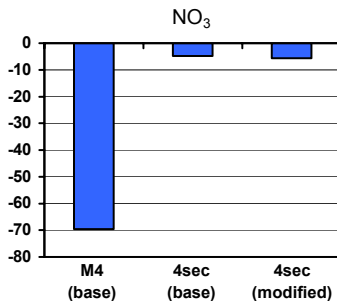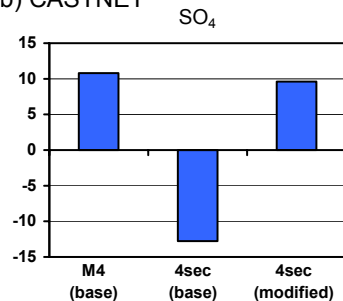
Figure 3-6. Fractional biases of the model predictions against IMPROVE and CASTNET observation data during January 1996.

Final Updates to CAMx Version 4.10

As discussed previously, after the completion of the western US annual 1996 modeling, updates were made to the MM5CAMx processor and CAMx wet deposition scheme to more consistently treat clouds and wet deposition including the treatment of subgrid-scale clouds. A comparison of CAMx Version 4.10beta used in the 1996 annual modeling and the final CAMx Version 4.10 available on the CAMx website (www.camx.com) for the July 1996 period and IMPROVE SO4 concentrations and NADP SO4 and NH4 wet deposition are presented in Figure 3-7. Although the cloud/precipitation/wet deposition update results in a slight degradation in sulfate model performance, with the IMPROVE and CASTNet under-prediction bias increasing from –12% to –15% and –15% to –22%, respectively (Figure 3-7), there are more significant improvements in wet deposition performance. As shown in Figure 3-7 bottom, the SO4 wet deposition under-prediction bias improves substantially from a –160% under-prediction bias to a –105% under-prediction bias using the new meteorology update. Similar significant improvements are also seen in the NH4 wet deposition whose under-prediction bias improves from –135% to –91%.
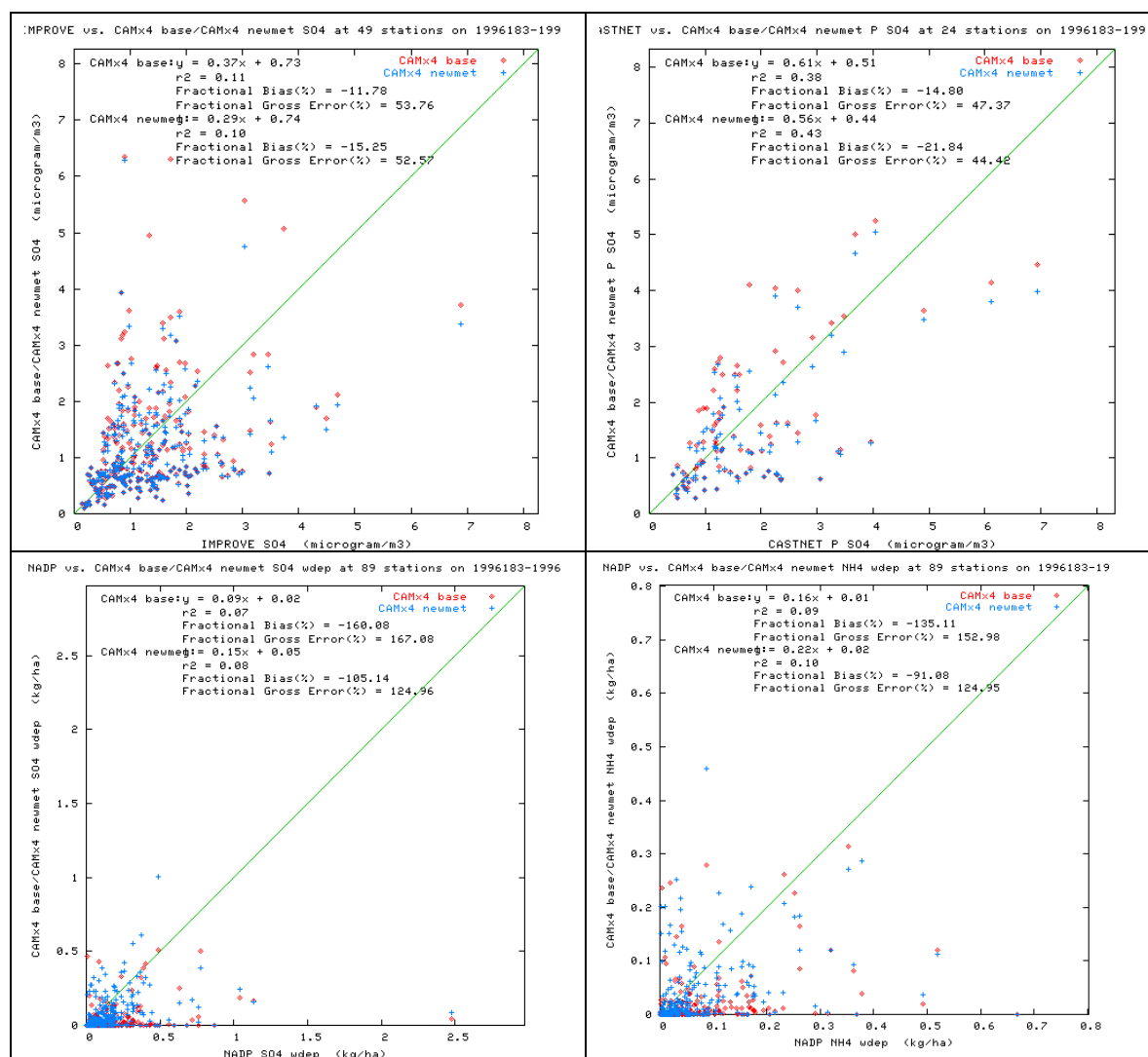
Figure 3-7. Comparison of CAMx Version 4.10beta (red) versus CAMx Version 4.10 final (blue) model performance for July 1996, the western US, and IMPROVE SO4 (top left), CASTNet So4 (top right), NADP wet SO4 (bottom left) and NADP wet NH4 (bottom right).

4.0 EPISODE AGGREGATION UNCERTAINTIES

A key consideration in modeling of atmospheric particulate matter (PM) for regulatory purposes is the need to estimate the impact of emissions scenarios over a full annual cycle of meteorological conditions. A variety of models for analyzing air quality management options related to atmospheric particulate matter have been developed and PM model development efforts are continuing. These models vary from relatively simple models suitable for quickly estimating impacts under a variety of emission scenarios using a full year of hourly meteorological and emission inputs to more complex, detailed models that are very resource intensive to setup and run and are thus more suitable for analyzing selected episodic conditions rather than being run with a full year of hourly inputs. Although there has been some success in performing annual simulations using detailed models with relatively coarse grid resolution (e.g., 36 km), performing annual simulations using more refined grid resolution would require extensive computational resources. Nevertheless, finer grid resolutions may be necessary to properly characterize the effects of emission controls on fine PM ($PM_{2.5}$) concentrations and resulting light extinction. For example, finer grid resolution may be needed to properly simulate the adverse effects of NOx controls on secondary $PM_{2.5}$ which can occur under certain circumstances. It may be necessary under some situations to estimate annual $PM_{2.5}$ concentrations and total light extinction summary statistics by aggregating episodic modeling results. Uncertainties in estimated annual summary statistics resulting from episode aggregation must be addressed to understand the significance of the model results and to guide decisions related to selection of episodes for modeling. Quantitative information about the uncertainties associated with aggregation of episodic model results to estimate annual summary statistics is currently very limited (see results of the literature review described by Stoeckenius, 2004). Of particular interest are uncertainties in estimation of the annual average $PM_{2.5}$ concentration (for applications related to the annual $PM_{2.5}$ National Ambient Air Quality Standard) and in the mean total light extinction on the 20% best and 20% worst visibility days (for applications related to the regional haze regulations).

EPISODE AGGREGATION UNCERTAINTIES IN MODELING ANNUAL AVERAGE $PM_{2.5}$

We evaluated uncertainties in annual mean $PM_{2.5}$ estimated by aggregation of episodic model results for both a base case simulation and a future year control scenario simulation. In the base case simulation, the model is used in an absolute sense to directly compute the annual average concentration. In the future year control scenario simulation, the model is used in a relative sense, that is to compute the ratio of control scenario to base case concentration (i.e., the so-called relative reduction factor). As described below, episode aggregation uncertainties differ substantially between these two simulation scenarios.

Uncertainties in annual average PM and PM component species concentrations estimated by aggregating model results from a limited number of short duration periods (modeling episodes) were evaluated using results from a full year photochemical model simulation of the eastern two-thirds of the U.S. conducted by the Lake Michigan Air Directors Consortium (LADCO) using the CAMx model. The CAMx simulation was driven by meteorological fields from an MM5 mesoscale meteorological model simulation of calendar year 2002. Model results were obtained from LADCO for two emission scenarios:

1. A base case simulation using the 1999 "Base E" emission inventory

2. A "Clear Skies" emissions scenario for the year 2010 that incorporated SO$_2$ and NO$_x$ reductions associated with EPA's proposed Interstate Air Quality Rule.

Under the 2010 Clear Skies scenarios, total SO$_x$ emissions were reduced by approximately 30%, NO$_x$ emissions by approximately 33%, and VOC emissions by approximately 15% from base case levels. Primary PM$_{2.5}$ and ammonia emissions remained nearly unchanged.

A summary of the episode aggregation uncertainty analysis procedures, results and conclusions is presented in the following subsections. Recommendations for further research are provided in Section 5. A more detailed discussion of the episode aggregation uncertainty analysis is presented by Stoeckenius (2004).

Uncertainties in Estimated Base Case Annual Means

Uncertainties were computed for annual averages estimated via aggregation of results from a single n-day episode period during each calendar quarter extracted from the output of the CAMx simulation described above. Uncertainties were compared over a range of values for n (n=7, 14 and 28 days).[1] A Monte Carlo simulation procedure was used to compute uncertainties. Each iteration of the Monte Carlo simulation consisted of the following steps:

1. Select a period of n consecutive days at random from the first calendar quarter (Q1) and compute the n-day average for each PM species at each surface layer grid cell.
2. Repeat Step 1 for Q2, Q3, and Q4.
3. Average the results of steps 1 and 2 over all four quarters. This is the estimated annual mean.

By repeating the above steps a large number of times and compiling the results, an uncertainty distribution for the estimated annual mean was obtained from which confidence intervals were then computed. Given the large volume of results (equal to the number of iterations times number of grid cells times number of species), only the mean and variance of the estimated annual averages for each species in each grid cell were stored for further analysis. Uncertainties were computed separately for total PM$_{2.5}$ and for PM components as defined in Table 4-1.

Table 4-1. Species used in the uncertainty analysis.

Species Abbreviation	Species	CAMx output
SO4	Sulfate Ion (SO4)	PSO4
NO3	Nitrate Ion (NO3)	PNO3
NH4	Ammonium Ion (NH4)	PNH4
HNO3	Nitric Acid (HNO3)	HNO3
AORG	Anthro Org (AORG)	POA + SOA1 + SOA2 + SOA3
BORG	Bio Org (BORG)	SOA4
EC	Elemental C (EC)	PEC
PI	Other Prim Inorg. (PI)	FPRM + FCRS
PM2.5	Total PM$_{2.5}$ (PM25)	PNO3+PSO4+PNH4+POA+PEC+FPRM+SOA1+SOA2+SOA3+SOA4+FCRS+PCL+NA

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[1] Multiples of seven were used for n as this insures that each modeled episode includes a proportionate number of weekdays and weekends.

<u>Absolute</u> uncertainties were expressed as one-half the width of the 95% confidence interval for the estimated mean. <u>Relative</u> uncertainties were defined as the absolute uncertainty divided by the "true" annual mean where the "true" mean is the mean over all 365 predicted daily values. Uncertainties were computed for each of the 95 x 88 = 8,360 36 x 36 km surface grid cells in the modeling domain for $PM_{2.5}$ and selected PM components.

Results of the above calculations are summarized below. Additional details are provided in a companion report (Stoeckenius, 2004).

- Relative uncertainties for the 1999 base case annual averages showed wide spatial variability corresponding roughly to spatial variations in annual mean concentrations. Relative uncertainties over all surface grid cells in the modeling domain are summarized in Figure 4-1. Median relative uncertainties over the modeling domain were found to be greater than ±20%, even at n = 28 days. Uncertainties are largest for NO3 followed by BORG, NH4, SO4. Uncertainties in TotNO3 are nearly as large as for NO3. Uncertainties are lowest for EC and PI. Mean uncertainties for total $PM_{2.5}$ are closer to that of EC and PI than NO3.

- Restricting attention to just locations with average $PM_{2.5} \geq 15 \ \mu g/m^3$ (see Figure 4-2) lowered the average uncertainties by 40 to 60% (except for BORG and HNO3). For these locations, the median $PM_{2.5}$ uncertainties are less than ±20% for n greater than or equal to 14 days as are the uncertainties for AORG, EC, and PI.

- As expected, uncertainties were found to decrease as n increases. The rate of decline in median uncertainties is somewhat less than the value expected if *individual* days were to have been selected at random from each quarter rather than selecting a single n-day block from each quarter. This difference is most likely due to autocorrelations in the time series of daily PM concentrations.

**Relative Uncertainties in Annual Averages
Over All Grid Cells**

Figure 4-1. Median, 25th and 75th percentiles of relative uncertainties in estimated 1999 base case annual mean concentrations over all surface layer grid cells in the modeling domain.

**Relative Uncertainties in Annual Averages
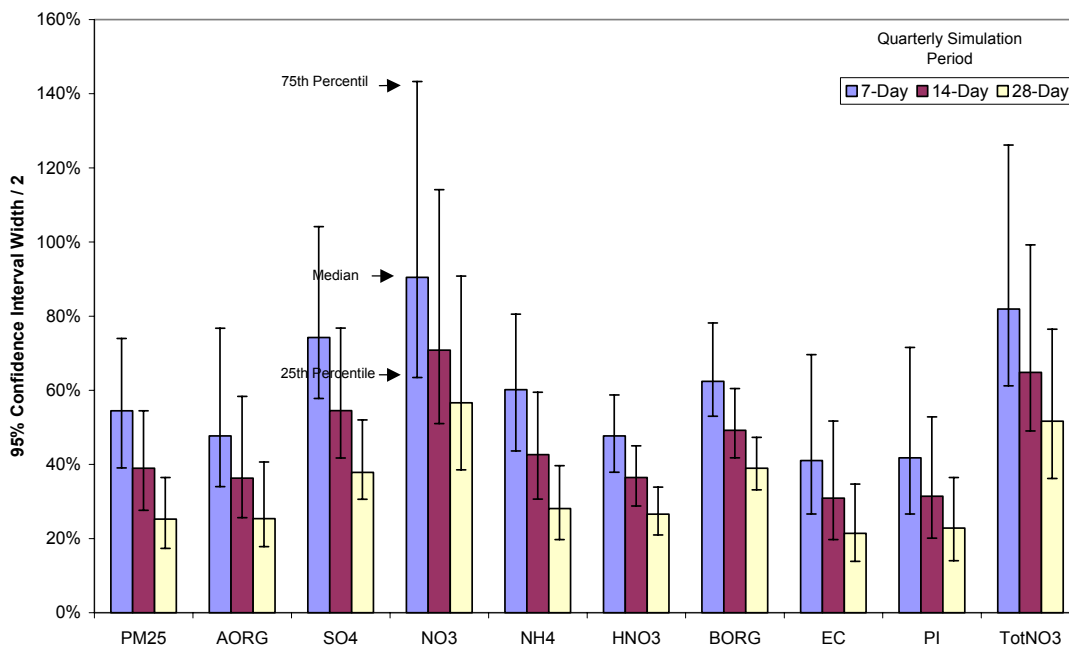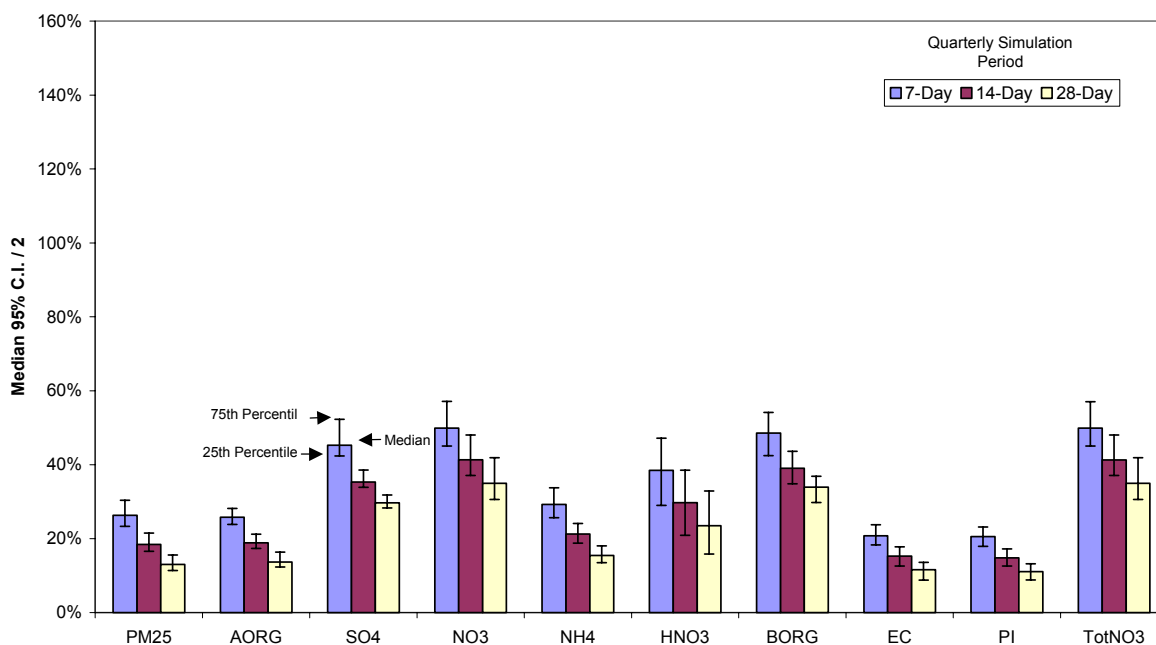Medians Over Grid Cells with Avg PM2.5 > 15 ug/m3**

Figure 4-2. Median, 25th and 75th percentiles of relative uncertainties in estimated 1999 base case annual means for all surface grid cells with annual mean PM$_{2.5}$ greater than 15 µg/m^3 (vertical scale is same as in Figure 4-1).

Uncertainties in Estimated Annual Means Under an Emissions Control Scenario

As noted above, EPA's draft PM modeling guidance (EPA, 2001) calls for control strategy attainment demonstrations to be based on the use of modeled relative reduction factors (RRFs) which are then applied to monitoring data to obtain the future year annual average. We therefore repeated the Monte Carlo simulation described above but in place of computing the predicted n-day average base case concentration in each quarter in step 1, we used the predicted relative reduction factor (RRF) times the "true" base case average in each calendar quarter:

$$X' = X * RRF$$

Where

X' is the estimated 2010 Clear Skies scenario quarterly average,
X is the "true" 1999 base case average and
RRF = estimated quarterly average 2010 Clear Skies for an n-day period divided by estimated quarterly average 1999 base case for the same n-day period.

RRFs were computed for both total PM$_{2.5}$ and each PM component in Table 4-1.[2]

Uncertainties in estimated annual averages under the 2010 Clear Skies scenario based on RRFs are summarized in Figure 4-3 for all grid cells with predicted annual average PM$_{2.5}$ greater than 15 µg/m^3. Comparison of these results with those in Figure 4-2 shows that the <u>uncertainties based on use of RRFs are generally just half or less of the uncertainties for estimating the 1999 base case absolute concentrations</u>. This illustrates the significant advantage of using RRFs in attainment demonstrations rather than absolute model predictions. Median uncertainties for n = 14 days are below ±10% for PM25 and all component species except SO4, NO3, HNO3 and TotNO3. Furthermore, in contrast to uncertainties for the 1999 base case, analysis of the relative uncertainties for the 2010 RRF scenario showed that they are not a strong function of mean PM$_{2.5}$: median uncertainties were found to be similar whether the medians were taken over all grid cells or just those grid cells with mean PM$_{2.5}$ greater than 15 µg/m^3.

As shown in Figure 4-3, relative uncertainties for estimated annual mean NO3 and TotNO3 under the 2010 scenario are much larger than for the other species. Uncertainties in HNO3 are also relatively high. A more detailed look at NO3 relative uncertainties under the 2010 RRF scenario as a function of annual mean PM$_{2.5}$ reveals uncertainties at some locations (including locations with predicted PM$_{2.5}$ greater than 15 µg/m^3) are extremely large (see Figure 4-4). Examination of the Monte Carlo simulation results revealed that these elevated uncertainty levels were due to the presence of some anomalous NO3 RRFs computed from ratios of n-day average NO3 that occurred primarily at locations with low to medium predicted mean NO3 and PM$_{2.5}$ levels. These anomalous NO3 RRFs result in unrealistic estimates of annual average NO3 under the 2010 scenario. As illustrated in Figure 4-5, even the mean estimates from the Monte Carlo simulation are unrealistically large for locations with base case NO3 as high as 4 µg/m^3. These results indicate that predictions of RRFs for NO3 and TotNO3 based on episodic model results are subject to large uncertainties and should be treated with caution.

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[2] This differs from the draft PM Modeling Guidance procedure in that the guidance calls for generating the reconstructed PM mass from the sum of masses associated with each component (so an RRF would not actually be computed for total PM2.5 or NH4). Also, AORG and BORG were treated separately in our calculations whereas the guidance deals with application to monitoring data in which only the total organic carbon is available. RRFs for BORG are of little interest as biogenic emissions remain uncontrolled in the 2010 scenario.

Relative Uncertainties in 2010 Annual Averages
Medians Over Grid Cells with Avg PM2.5 > 15 ug/m3
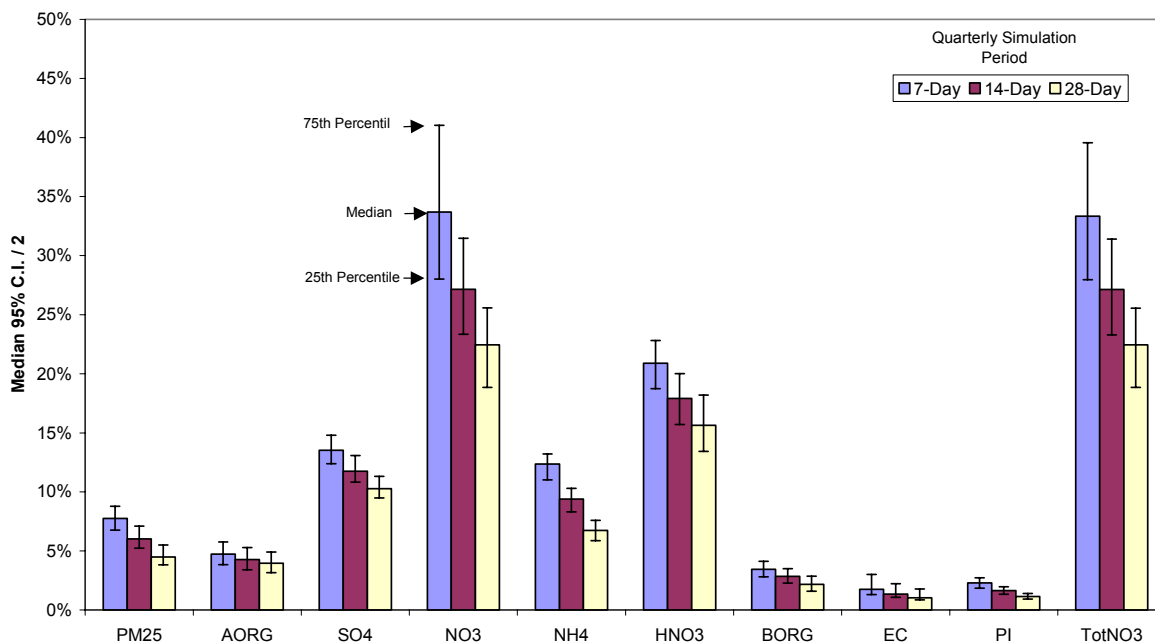
Figure 4-3. Median, 25th and 75th percentiles of relative uncertainties in estimated annual averages under the 2010 Clear Skies scenario based on application of relative reduction factors; values are based on results from surface grid cells with annual mean $PM_{2.5}$ greater than 15 µg/m^3.

AnnAvg.NO3 Uncertainties
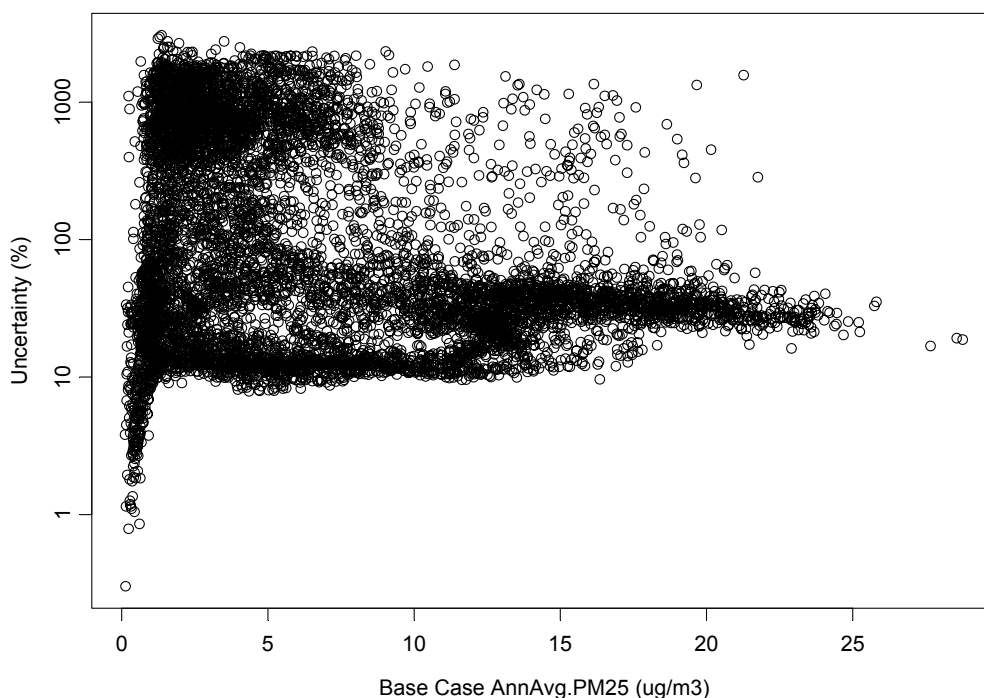
Figure 4-4. Uncertainties in annual average NO3 under the 2010 Clear Skies scenario estimated from relative reduction factors calculated over n = 7 day periods in each quarter; uncertainties are plotted against the corresponding 1999 base case annual average $PM_{2.5}$ concentrations.
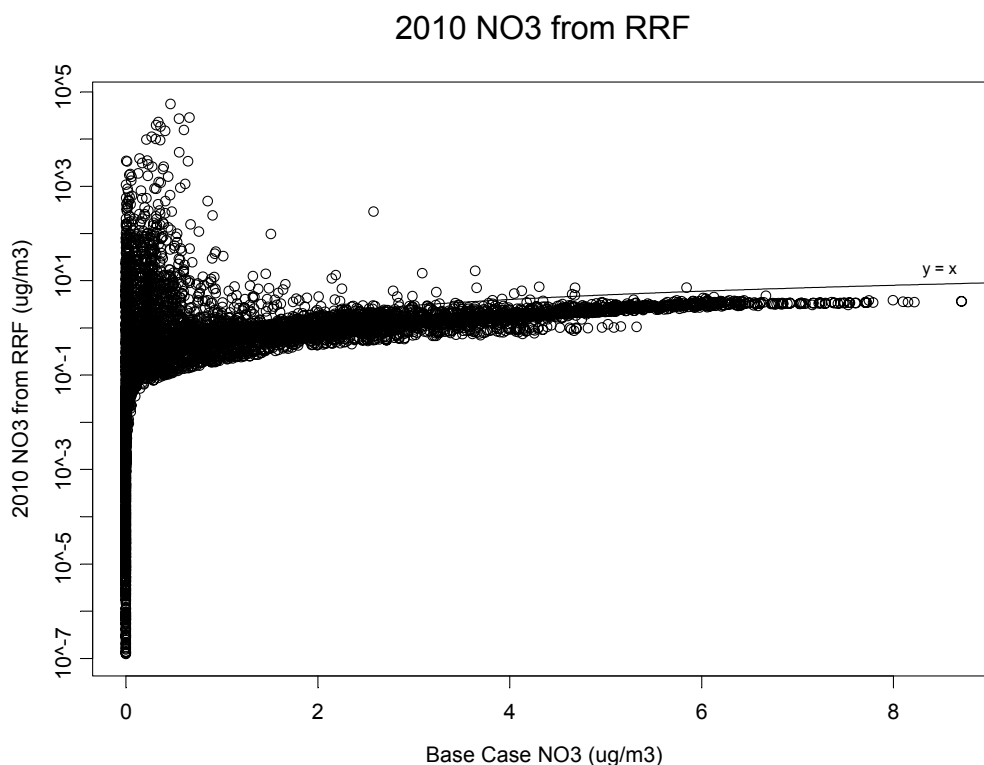
Figure 4-5. Averages of the estimated annual average NO3 concentrations under the 2010 control scenario simulated by the Monte Carlo procedures described in the text using randomly selected 7-day periods.

EPISODE AGGREGATION UNCERTAINTIES IN VISIBILITY MODELING

EPA's draft regional haze (RH) modeling guidance (EPA, 2001) calls for multiplying average predicted RRFs for individual PM components by corresponding observed base year concentrations on the 20% best and worst visibility days, computing the reconstructed light extinction, and comparing the difference between the resulting average estimated "future year" light extinction under the control strategy and the observed base year light extinction to the regional haze goals. EPA's guidance calls for computing average predicted RRFs based on ratios of average model results for groups of days corresponding to the 20% best and worst observed base case visibility days during the modeling period. [3] This is to be done separately at each Class I area. Currently, the EPA guidance calls for modeling a full representative year if possible or, if this is not possible, a sufficiently long period so that at least 10 or more days corresponding to the 20% "best" and "worst" observed visibility days are included among the modeled days. This recommendation is based on results from a study of ozone modeling results by Hogrefe et al. (2000) and the assumption that RRFs are likely to be different between "best" and "worst" visibility days.

To our knowledge, a systematic analysis of episode aggregation uncertainties for PM components in the context of RH regulatory modeling has not been performed. Clearly, RRFs

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[3] The relative reduction factor (RRF) is defined as the predicted concentration under a future year emission control scenario divided by the corresponding predicted concentration under the base year emissions scenario.

based on averages over n modeling days will have less uncertainty as n grows larger. It is therefore not clear how uncertainties are best minimized given the conflicting requirements to base RRFs on as many days as possible while simultaneously computing RRFs separately for a (possibly small) group of "best" and "worst" visibility days. We explored this issue with the western U.S. modeling data sets described in Section 3 above (see Koo and Morris, 2004 for additional information) by:

1. Examining the relationship between daily RRFs and predicted base case light extinction and;
2. Examining the relationship between RRF uncertainties and the number of days used to compute the RRFs.

The western US CAMx modeling databases used in our analysis (see Section 3) included the "one atmosphere" (M4) simulations for 1996 base case and four sensitivity runs consisting of 50% across-the-board reductions in either NO_x, VOC, NH_3, or SO_2 emissions. [4] The sensitivity run results were available for January and July only. Thus, there are no full year sensitivity runs available to compare with the full year base case run.

Relationship of Daily RRFs to Bext

Predicted daily average PM component concentrations for surface layer grid cells corresponding to 132 IMPROVE and IMPROVE protocol monitoring site locations were extracted from the CAMx (M4) model output. Extracted PM components are defined in Table 4-2. Daily RRFs were computed by dividing the predicted daily average concentration under each of the four 50% emission reduction sensitivity ("control scenario") runs described above by the corresponding prediction from the base case run.

Table 4-2. Daily average PM components extracted from CAMx model output.

PM Component		CAMx species
SO4	Sulfate Ion	PSO4
NO3	Nitrate Ion	PNO3
NH4	Ammonium Ion	PNH4
OC	Organic Carbon	POA + SOA1-4
EC	Elemental Carbon	PEC
SOIL	Soil	FPRM + FCRS
CM	Coarse Mass	CPRM + CCRS

RRFs for each control scenario are summarized in Table 4-3. EC, SOIL and CM RRFs are always equal to one since emissions that contribute to these components are not affected by any of the four control scenarios. OC reductions under the VOC control scenario reflect the influence of secondary anthropogenic organic PM. OC is slightly reduced in a few cases under the NO_x reduction scenario due to reduced photochemical oxidation rates. NH4 is reduced most broadly under the NH_3 reduction scenario as one would expect but reductions also occur in some cases under each of the other control scenarios where NH_3 is not the limiting factor in PM formation. There are also isolated cases of NH4 RRFs >> 1 under each of the emission

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[4] The analysis was performed for the M4 model output only; we would expect results based on the four section, full science (4sec) model output to be similar.

reduction scenarios due to detailed photochemical simulation effects that are not relevant to the present study.

Table 4-3. Summary statistics for relative reduction factors for each PM component computed under four emission scenarios (Inf indicates the RRF is equal to x/0 where x is non-zero).

50% SOx Reduction Scenario							
	SO4	NO3	NH4	OC	EC	SOIL	CM
Min.	0.55	0.00	0.43	0.99	0.99	0.99	1.00
1st Qu.	0.70	1.00	0.78	1.00	1.00	1.00	1.00
Median	0.78	1.02	0.97	1.00	1.00	1.00	1.00
Mean	0.79	Inf	0.94	1.00	1.00	1.00	1.00
3rd Qu.	0.90	1.16	0.99	1.00	1.00	1.00	1.00
Max.	1.00	Inf[5]	9.24	1.01	1.01	1.01	1.00
50% NOx Reduction Scenario							
	SO4	NO3	NH4	OC	EC	SOIL	CM
Min.	0.77	0.00	0.26	0.87	0.99	0.99	1.00
1st Qu.	0.93	0.37	0.78	0.95	1.00	1.00	1.00
Median	0.99	0.53	0.90	0.97	1.00	1.00	1.00
Mean	0.99	Inf	0.87	0.98	1.00	1.00	1.00
3rd Qu.	1.02	0.67	0.96	0.99	1.00	1.00	1.00
Max.	2.16	Inf[6]	9.65	1.16	1.02	1.02	1.00
50% NH3 Reduction Scenario							
	SO4	NO3	NH4	OC	EC	SOIL	CM
Min.	0.33	0.00	0.03	0.98	0.99	0.98	1.00
1st Qu.	0.99	0.05	0.53	1.00	1.00	1.00	1.00
Median	1.00	0.30	0.69	1.00	1.00	1.00	1.00
Mean	0.98	Inf	0.69	1.00	1.00	1.00	1.00
3rd Qu.	1.00	0.54	0.88	1.00	1.00	1.00	1.00
Max.	1.02	Inf[7]	1.67	1.02	1.03	1.03	1.00
50% VOC Reduction Scenario							
	SO4	NO3	NH4	OC	EC	SOIL	CM
Min.	0.93	0.00	0.26	0.67	1.00	1.00	1.00
1st Qu.	1.00	1.01	1.00	0.87	1.00	1.00	1.00
Median	1.01	1.02	1.02	0.92	1.00	1.00	1.00
Mean	1.02	Inf	1.02	0.91	1.00	1.00	1.00
3rd Qu.	1.04	1.07	1.03	0.95	1.00	1.00	1.00
Max.	1.17	Inf[3]	1.30	1.05	1.00	1.00	1.00

Summary statistics shown in Table 4-3 are taken over 132 sites and 60 days in the model output (January and July combined) for a total of 7,920 site-days. In the case of the NO3 RRFs under the 50% NO$_x$ reduction scenario, 2,389 of these (30%) are of the form 0/0 which are not included

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[5] Largest value not of the form x/0 = Inf is order 10^4

[6] Largest value not of the form x/0 = Inf is order 10^2

[7] Largest value not of the form x/0 = Inf is order 10^3

in the summary statistics in Table 4-3. Another 0.6% are of the form x/0 where x (the predicted control scenario NO3 concentration) is greater than zero but the base case NO3 concentration is equal to zero and 3% are of the form 0/x = 0 and for these, x ranges from near zero to 1 $\mu g/m^3$. There are also a few very small non-zero NO3 RRFs (some as small as 10^{-3}), all of which are associated with base case NO3 values less than about 1 $\mu g/m^3$.

Daily average total reconstructed light extinction (Bext) was computed from the base case PM component predictions using procedures described in EPA's draft regional haze (RH) modeling guidance.[8] Daily RRFs for each PM component under each control scenario were computed at locations corresponding to each IMPROVE monitoring site and regressed against Bext. An example for SO4 RRFs under the 50% SO₂ reduction scenario at two sites (Yellowstone and Mesa Verde) is shown in Figure 4-6; the association of total light extinction with SO4 concentration at these two locations is shown in Figure 4-7. These results illustrate a range of behaviors depending on the degree to which SO4 contributes to total light extinction at a given location. At Yellowstone, SO4 concentrations are low and only weakly associated with Bext while at Mesa Verde predicted SO4 concentrations are relatively high and closely associated with Bext; the relationship is linear with Bext expressed in 1/Mm as one would expect (Figure 4-7). Since RRFs tend to be smaller at higher concentrations, this results in a stronger relationship between the SO4 RRFs and Bext at Mesa Verde (where the regression slope is statistically significant) as compared to Yellowstone (where the slope is near zero).

SO4 RRFs: 50% SO2 Reduction Scenario

y = -0.0057x + 0.8674
R^2 = 0.0424

y = -0.0215x + 0.9244
R^2 = 0.3637

Yellowstone NP (YELL2)
Mesa Verde NP (MEVE1)
Linear (Yellowstone NP (YELL2))
Linear (Mesa Verde NP (MEVE1))
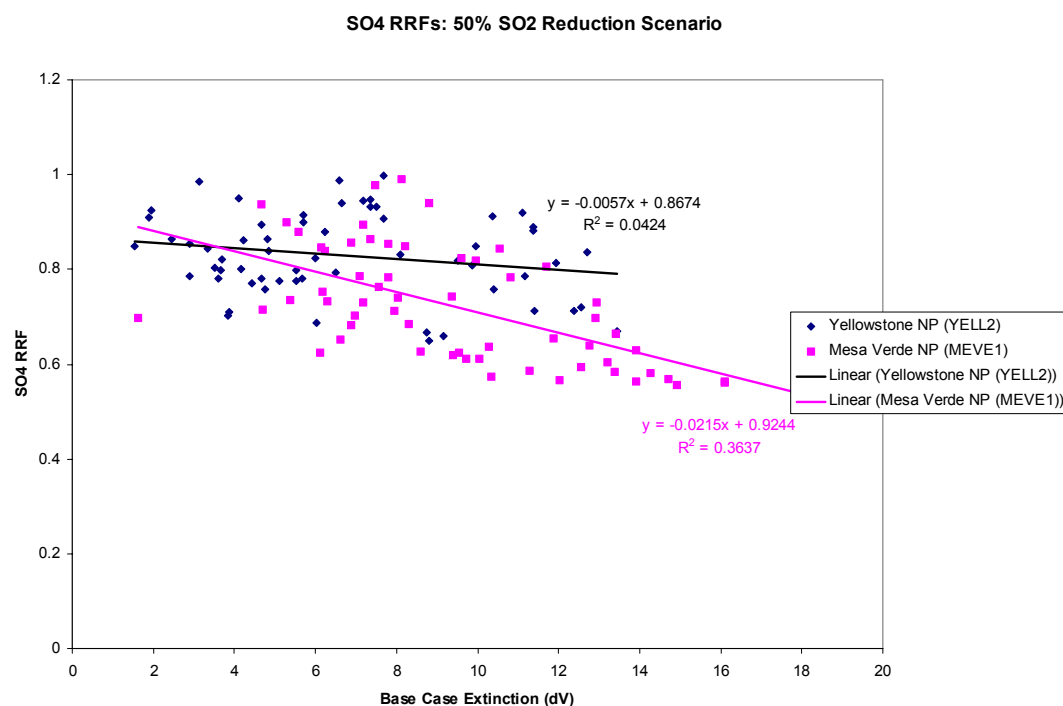
Figure 4-6. Relationship of SO4 relative reduction factor (RRF) to base case total light extinction (deciviews) at Yellowstone and Mesa Verde.

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[8] For the purposes of this limited analysis, we were not able to easily match the climatological average f(RH) values at individual Class I areas compiled by EPA (2003b) with the IMPROVE monitoring sites where model results were extracted as described above. Instead we simply applied f(RH) from a single site (Yellowstone National Park) to calculate Bext from the model results at all of the monitoring sites. This should have little effect on the results described here so long as correlations between average humidity and PM component concentrations are weak.
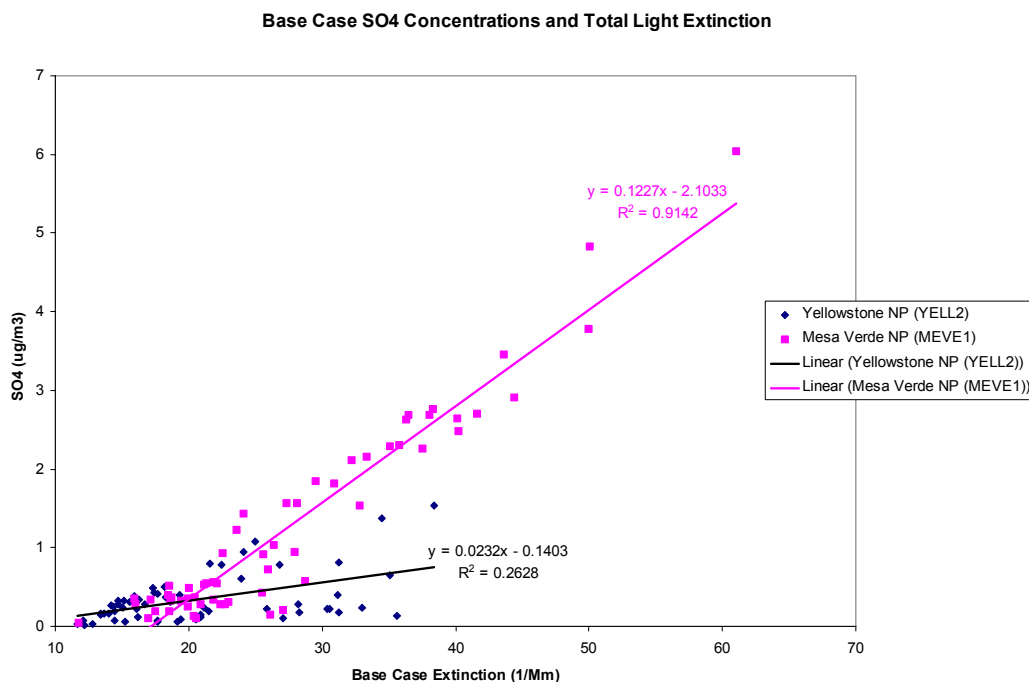
Figure 4-7. Relationship of SO4 concentration and base case total light extinction (deciviews) at Yellowstone and Mesa Verde.

In practice it should be noted that EPA's draft guidance calls for using (to the maximum possible extent consistent with other episode selection criteria) RRFs predicted on days when the observed Bext falls within the 20% best/worst days. Since the correlation between predicted and observed daily average Bext will not be perfect, the relationships between predicted RRFs and observed Bext will be weaker than the correlations between predicted RRFs and predicted Bext shown in Figure 4-6.

From the standpoint of selecting modeling episodes for predicting progress on the 20% best/worst visibility days as required by the Regional Haze regulations, the results described above imply that for some species at some locations (such as SO4 at Yellowstone), it may be appropriate to select days for computation of the RRF regardless of Bext (and thereby include a larger number of days in the RRF calculation and consequently obtain a more precise estimate). However, in other cases (such as SO4 at Mesa Verde) it is necessary to include Bext as a factor in the selection of days used in the RRF calculation (to avoid bias) even though this means that the estimate is less precise.

In actual regulatory modeling applications, the tradeoff between precision and accuracy (bias) will vary from site to site and will depend on the particular combination of emission controls being simulated. This makes it impossible to draw any general conclusions regarding episode aggregation uncertainties in regional haze modeling based on the two months of simplified control strategy model results available for use in this study. Recommendations for a follow-on study to address this issue are presented in Section 5.

Relationship of RRF Uncertainty to Length of Modeling Period

Episode aggregation uncertainties increase as the number of modeled episode days used to estimate the predicted change in light extinction on best and worst visibility days decreases. We prepared a limited quantitative evaluation of the sensitivity of episode aggregation uncertainties to changes in the number of modeled days using the January and July model results described above. Using model results at a given IMPROVE monitoring site for one month (January or July), we computed RRFs for all possible running n-day averages and calculated approximations to the relative 95% confidence interval half-widths of the RRFs from the resulting distribution as one-half the difference between the 0.975 and 0.025 quantiles divided by the mean. These confidence intervals are likely to somewhat underestimate the confidence interval widths for the full winter or summer calendar quarters (especially for n >> 1) since the range of n-day averages calculated over a single month (January or July) of model output is more limited than the range one would expect over a full three month period.   Calculations were carried out for values of n ranging from 1 to 15 days at five representative IMPROVE sites:

Site ID	Location
SAAN1	San Andres, NM
GRCA1	Hopi Point (Grand Canyon), AZ
SAGO1	San Gorgonio Wilderness, CA
BLIS1	D.L. Bliss State Park, CA
SAWT1	Sawtooth National Forest, ID

Results illustrated in Figures 4-8 (January) and 4-9 (July) reveal several key points:

- o Modeling a period at least 10 days in length in each month is estimated to be sufficient to reduce the SO4 RRF uncertainties under the SO_x reduction scenario to less than about ±10% at all sites in July; as little as 8 days is sufficient in January.

- o Uncertainties in OC RRFs under the VOC reduction scenario are less than ±10% even for just a 1 day modeling period, the single exception being an uncertainty of ±12% for n=1 at GRCA1.

- o Uncertainties in NO3 RRFs are highly variable from site to site and are quite large in some cases. NO3 RRF uncertainties could not be calculated in some cases because the base case predicted NO3 is reported as zero.

- o NO3 RRF uncertainties shown in Figures 4-8 and 4-9 may be poor estimates of actual confidence interval widths given the problem of dealing with extremely low NO3 predictions. Clearly, better modeling procedures are needed to more accurately estimate the impact of future year NO3 changes on visibility levels.

While these results provide initial guidance on uncertainty levels to be expected in "real world" regulatory modeling applications, it must be noted that they are based on very simple, uniform across-the-board emission reduction scenarios; more spatially and temporally complex emission scenarios can be expected to yield somewhat larger uncertainties.

Relative Uncertainties in January Mean SO4 RRFs
50% SOx Reduction Scenario

Figure 4-8(a). Relative uncertainties in January mean SO4 RRFs under the 50% SO_x reduction scenario.

Relative Uncertainties in January Mean OC RRFs
50% VOC Reduction Scenario

Figure 4-8(b). Relative uncertainties in January mean OC RRFs under the 50% VOC reduction scenario.

Relative Uncertainties in January Mean NO3 RRFs
50% NOx Reduction Scenario

Figure 4-8(c). Relative uncertainties in January mean NO_x RRFs under the 50% NO_x reduction scenario.

Relative Uncertainties in July Mean SO4 RRFs
50% SOx Reduction Scenario

Figure 4-9(a). Relative uncertainties in July mean SO$_4$ RRFs under the 50% SO$_x$ reduction scenario.

Relative Uncertainties in July Mean OC RRFs
50% VOC Reduction Scenario

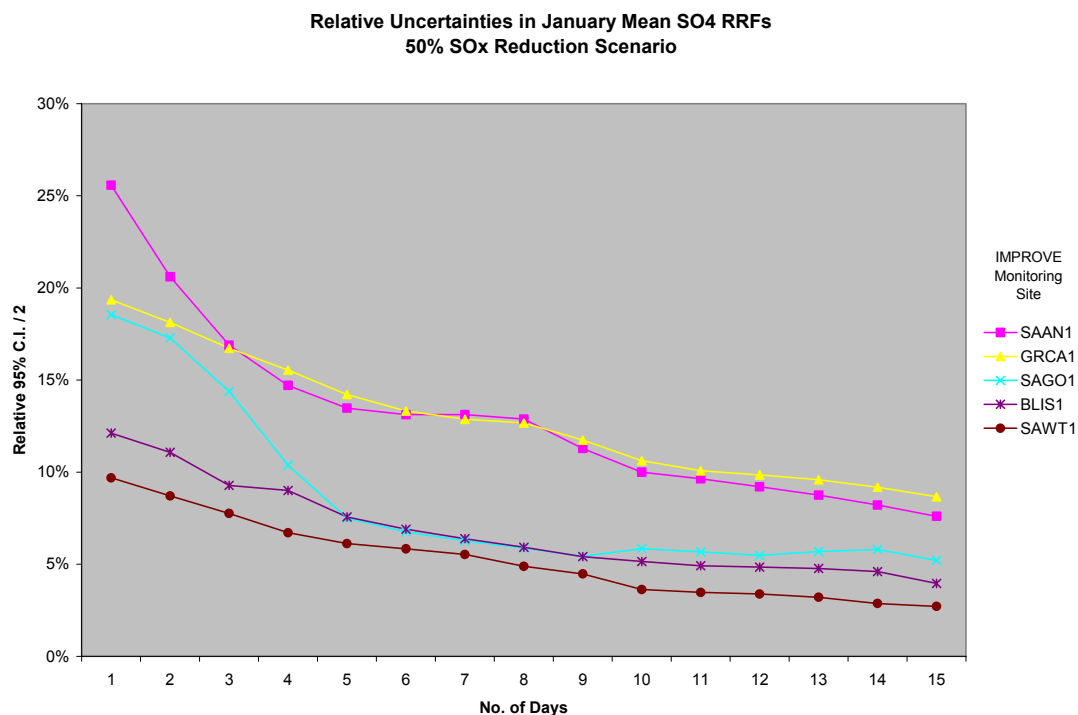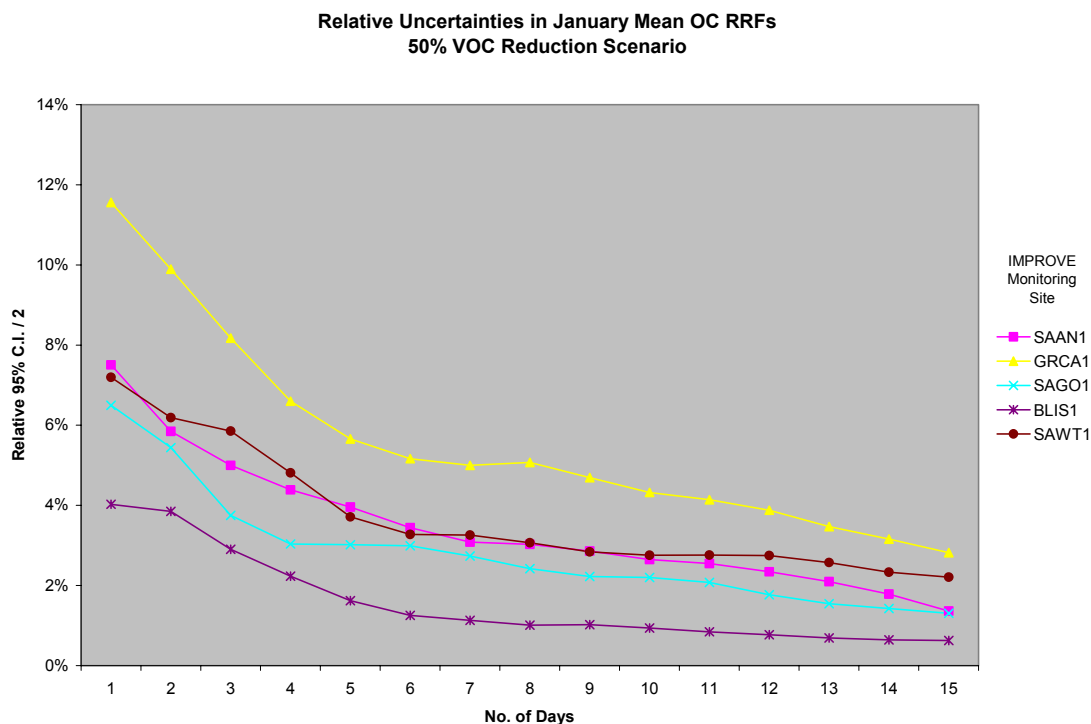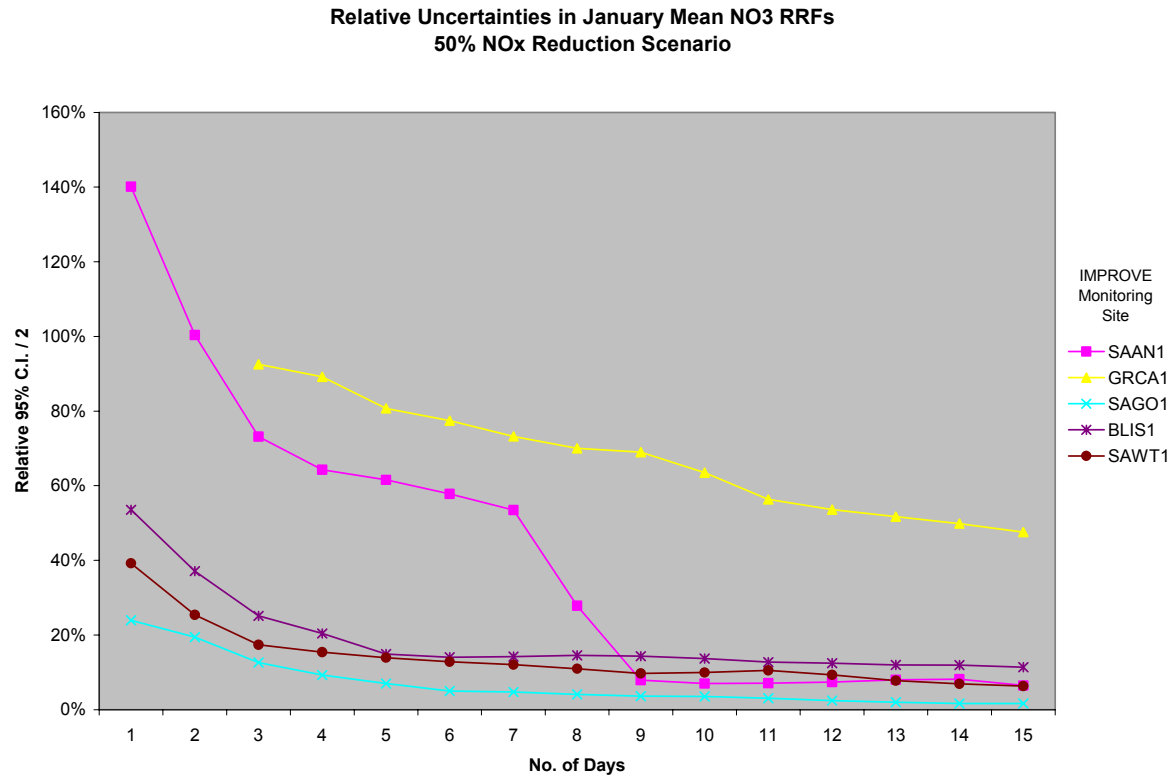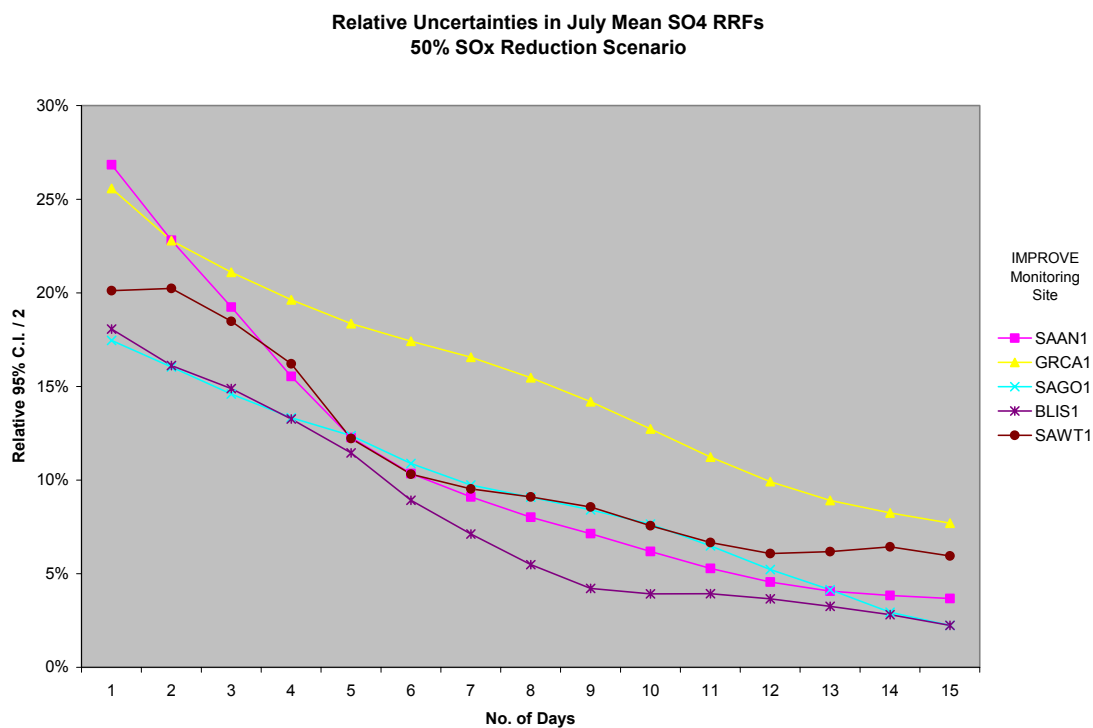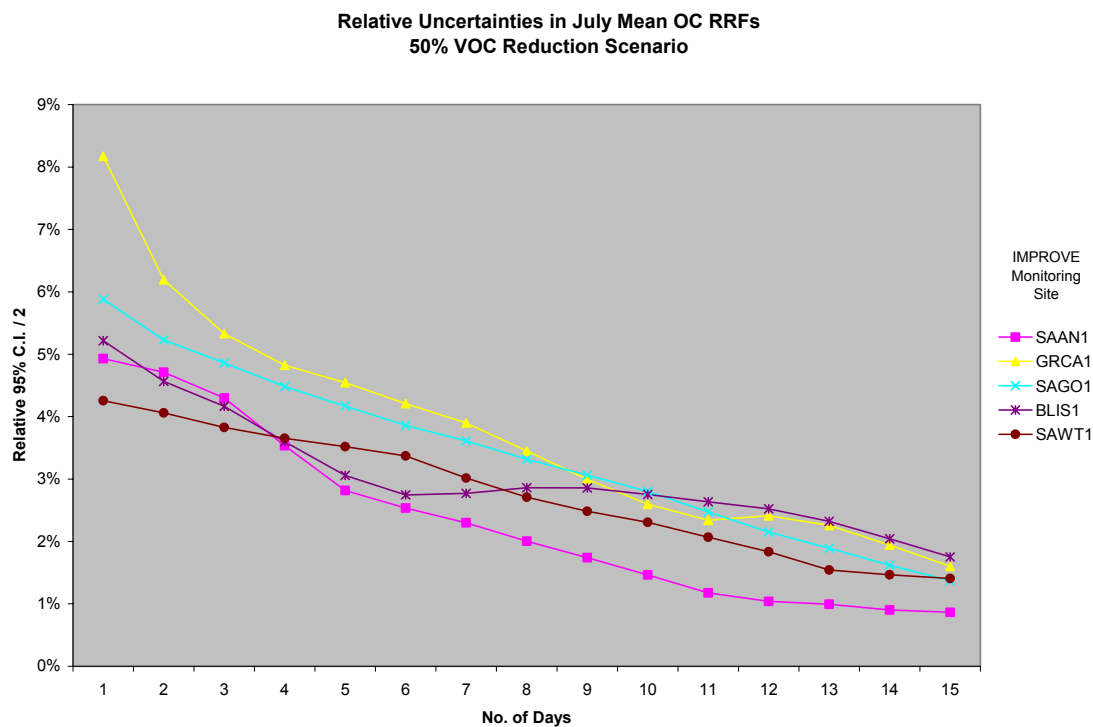**Figure 4-9(b)**. Relative uncertainties in July mean OC RRFs under the 50% VOC reduction scenario.

Relative Uncertainties in July Mean NOx RRFs
50% NOx Reduction Scenario
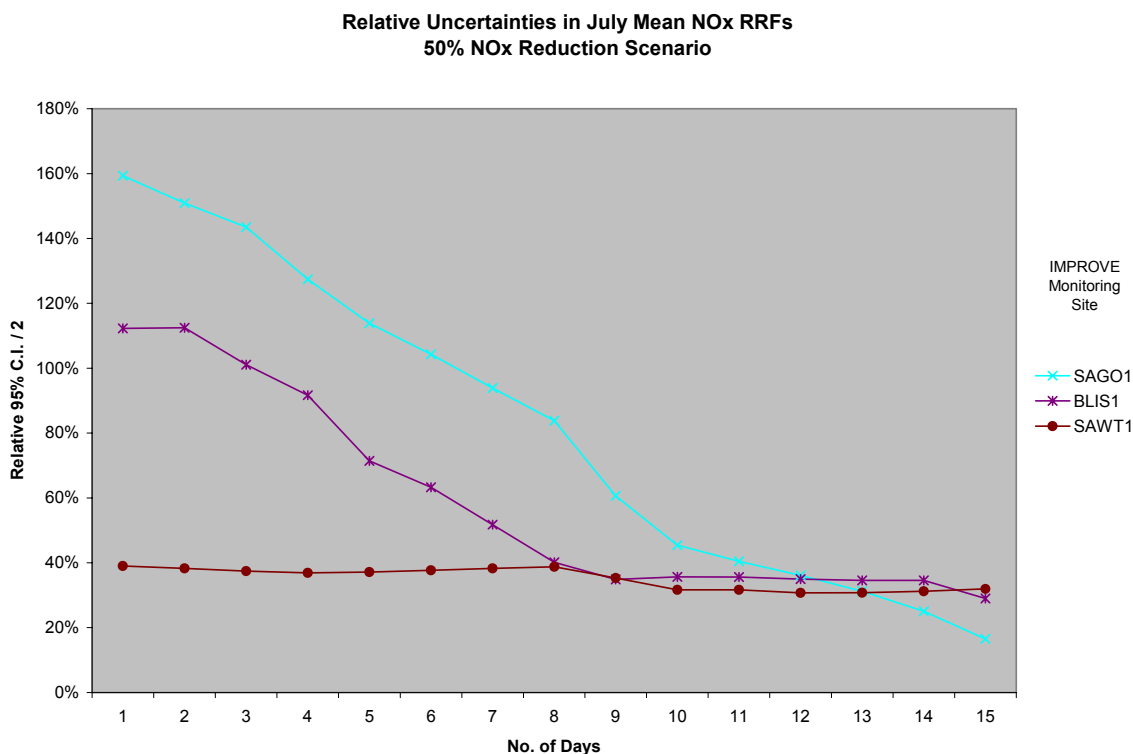
Figure 4-9(c). Relative uncertainties in July mean NO_3 RRFs under the 50% NO_x reduction scenario.

CONCLUSIONS

Results of the analyses summarized above provide a basis for improving upon the episode selection guidance contained in EPA's draft PM modeling guidance document for modeling annual average $PM_{2.5}$ and regional haze.

Annual Average $PM_{2.5}$

Aggregation of episodic particulate matter model results can introduce significant uncertainties in direct estimates of annual mean concentrations of PM and PM components even if periods as long as four weeks are simulated in each calendar quarter. These uncertainties will partially or wholly offset any precision gained by the use of more detailed episodic models as compared to use of a simplified model to simulate an entire year. Uncertainties are particularly large for sulfates and nitrates which are the focus of much of the simulation improvements incorporated into the more detailed, "state-of-the-science" models.

Significant reductions in episode aggregation uncertainty of 50% or more can be achieved by restricting attention to the prediction of *relative changes* in PM concentrations under alternative emission scenarios (i.e., using predicted relative reduction factors). While this conclusion is based on analysis of a single alternative emissions scenario (the 2010 Clear Skies scenario), one would expect similar uncertainty reductions under other similarly broad scale SO_x, NO_x and

VOC control scenarios. Primary PM and ammonia control strategies were not considered in this study. Uncertainties in primary PM RRFs will be low since this material is largely inert; including ammonia control strategies could lead to somewhat different results.

Episode aggregation uncertainties in PM components under a future year emissions scenario predicted on the basis of relative reduction factors for one 14-day period per calendar quarter are all below ±16% at locations with annual average $PM_{2.5}$ greater than 15 $\mu g/m^3$ except for nitrates. Uncertainties in nitrates at these locations remain significant, averaging ±27% and ranging as high as ±66%. Thus, even when models are used only in a relative sense, episode aggregation uncertainties are significant in applications where nitrates are a species of concern. This finding, coupled with the poor model performance for nitrates described for the CAMx runs used in this study by Baker (2003) and also currently being experienced in other model applications by several of the Regional Planning Organizations (MRPO, WRAP, VISTAS), suggest that current modeling techniques are not adequate for addressing nitrate issues.

Results of this study are based on a *random* selection of episode periods. Many analysts have speculated that episode aggregation uncertainties can be reduced by grouping together days with similar meteorological conditions and selecting modeling episodes from among the most representative days within each of the resulting groups. It should be noted that such an approach is really only feasible for regional or smaller scale model applications as there is likely to be little commonality in daily meteorological conditions associated with peak PM events over a large, multi-regional domain such as the one used in this study. Even for regional scale applications, however, the small uncertainty estimates for model results based on relative reduction factors presented above (except for nitrates) suggest that there may be little to gain by going through a more complex meteorological stratification process for episode selection.

Visibility Metrics

In contrast to episode aggregation uncertainty for annual average $PM_{2.5}$ where bias is not an issue so long as episode selection is not dependent on $PM_{2.5}$ concentration, uncertainties in estimated changes in total light extinction involve both bias and imprecision since episodes used in the RRF calculations may be selected on the basis of total light extinction (Bext).

For some species at some locations, the variation of RRF with Bext is small compared to day-to-day variations suggesting that bias is not a concern and it is better to include all available days in the RRF calculation (so as to get a more precise estimate) instead of restricting attention to just a few modeled 20% best/worst days. In other cases, RRFs appear to be more closely correlated with Bext (at least with the predicted Bext; correlation with observed Bext will in general be weaker). In such cases, the RRF calculation should incorporate the trend with Bext so as to avoid a biased estimate, even though this will result in some loss of precision.

Based on our preliminary evaluation with a limited set of model results, it appears that modelers should consider 10 days to be a minimum for estimating RRFs (except for NO3). As pointed out in Section 3, model performance is poor for NO3 and NO3 RRFs are highly variable; this makes it difficult to obtain a reliable prediction of Bext under future year scenarios where NO3 is a significant contributor to total Bext.

Recommendations for extending the above analysis using full year modeling of a "realistic" regional control scenario are described in Section 5.

5.0 RECOMMENDATIONS

RECOMMENDATIONS REGARDING PM MODEL DEVELOPMENT AND PERFORMANCE EVALUATION

The 1996 annual particulate matter (PM) and ozone modeling of the western United States represented the first time the CMAQ, REMSAD and CAMx models were applied, evaluated and intercompared for an annual period using consistent inputs. The three models exhibited similar performance attributes, with model performance for sulfate being better than the other species and performance for nitrate being poor. More recent CMAQ and CAMx applications, such as the VISTAS applications for the southeastern US and the July 1999 and July 2001 episodes (Morris et al., 2004b,c) have exhibited improved CMAQ and CAMx model performance over that seen in the 1996 western US applications. There are several reasons for the improved model performance in the more recent applications:

- Better meteorological fields than the historical 1996 MM5 simulation;
- Improved emission inventories over the WRAP Section 309 inventories used in the 1996 modeling; and
- Air quality modeling of the western US is more difficult than the eastern US:
    - More complex terrain in the west;
    - More isolated sources and receptors; and
    - Lower PM concentrations that are not dominated by sulfate.

Given the limitation in the 1996 database and the emerging 2001 and 2002 PM modeling databases, we do not recommend any additional analysis using the 1996 database. However, even with these limitations, the 1996 annual modeling and episode aggregation analysis did identify several areas where the state of regional PM and ozone modeling can be improved.

Recommended Updates to CAMx

The CAMx model continues to be updated and enhanced. CAMx Version 4.10 was released in August 2004 with the "one-atmosphere" M4 2-section and "full-science" multi-section aerosol treatment. The next release is expected by the end of 2004 and would include two more enhancements:

- Inclusion of a treatment for mercury; and
- Implementation of the PM Source Apportionment Technology (PSAT).

In addition, further CAMx research and development into the following areas is underway:

- Implementation of a full chemistry Plume-in-Grid (PiG) module;
- Addition of reactive tracers in the PiG and inclusion of subgrid-scale plume sampling;
- Implementation of the Decoupled Direct Method (DDM) sensitivity and Process Analysis (PA) in the PM modules (CRC Project A-51).

Based on the 1996 modeling, we have the following recommended improvements to the CAMx model:

Implementation of MPI Multi-Processing Capability: The current version of CAMx PM modules can only be exercised on one CPU. This severely limits the application of the model for annual simulations and limits the use of the CAMx probing tools. Thus, MPI multi-processing capability must be implemented in CAMx in order for it to be a viable tool for long-term modeling.

Improvements in the CAMx SOA Modules: Current CAMx SOA module does not account for exiting organic particles, which may explain some of the differences seen with CMAQ and REMSAD. The SOA module should be updated, tested and implemented in CAMx.

Updated Aqueous-Phase Module: The CMU aqueous-phase chemistry Variable Size Resolution Model (VSRM) is too computationally demanding and sometimes unstable making it not usable for annual modeling. Consequently, the old (circa 1980s) bulk RADM aqueous-phase chemistry module is used. A more recent and computationally efficient multi-section aqueous-phase module should be tested and implemented in CAMx.

In addition, several of the RPOs (e.g., WRAP and VISTAS) are performing 2002 annual modeling using Models-3 CMAQ. CAMx should be set up for the 2002 period taking advantage of the MM5 and CMAQ modeling performed by the RPOs. In addition to using improved meteorological and emission inputs over what was used in the 1996 application, a 2002 CAMx application would have the following additional enhancements over 1996:

- Cover both the western and eastern US
- Include more ambient measurements in the evaluation that were not available in 1996:
  - Enhanced IMPROVE network;
  - EPA's Speciated Trends Network (STN);
  - Southwest US SEARCH Sites; and
  - PM Supersites.

RECOMMENDATIONS REGARDING EPISODE SELECTION AND ADDITIONAL EPISODE AGGREGATION UNCERTAINTY ANALYSES

The following are our recommendations from the episode aggregation uncertainty analysis.

Episode Aggregation Uncertainties in Annual Average PM$_{2.5}$ Modeling

Results presented in Section 4 support the recommendation that modeling of annual average PM$_{2.5}$ under future year control scenarios involving changes in SO$_2$ and VOC emissions should be based on model results obtained for at least 14 days in each calendar quarter. The use of results from just 14 days during each quarter is acceptable so long as the model results are just used to compute relative reduction factors (RRFs) as called for in EPA's draft PM modeling guidance. Absolute predictions of base-case annual averages obtained from such a limited set of modeling days can only be considered rough approximations. Even when just the relative reduction factors are used, however, model results at locations where nitrates contribute

significantly to annual PM$_{2.5}$ will be subject to considerable uncertainty and must be treated with caution. We recommend that further investigation be performed of NO3 prediction uncertainties and factors driving the extreme variability in in NO3 RRFs. We also recommend that the results presented in Section 4 be amended to include analyses of uncertainties under additional control scenarios, especially scenarios involving changes in ammonia emissions.

Episode Aggregation Uncertainties in Regional Haze Modeling

Only a limited analysis of episode aggregation uncertainties in regional haze modeling could be done with the two months of simplified control strategy runs (single pollutant across-the-board reductions) available to this study. We recommend that additional investigation be performed using a full annual modeling database consisting of a base case and a "realistic" future year multi-pollutant control strategy simulation. A Monte Carlo simulation procedure similar to that employed in the analysis of episode aggregation uncertainties in annual average PM$_{2.5}$ modeling could then be used to examine uncertainties in predicted changes in total light extinction (Bext). One way to perform such an analysis would be to first use the full year of model results to compute the "true" Bext changes at each visibility monitoring site of interest predicted for the 20% observed best/worst visibility days as per EPA's draft modeling guidance. We would then select subsets of days at random, repeat the Bext progress calculation for each subset and compile statistics on bias and precision of the estimates as a function of the number of days selected, the selection criteria, and the method used to compute the RRFs that go into the Bext calculation.

We further recommend that the analysis described above be performed separately for western and eastern US modeling databases so as to properly account for the different conditions associated with haze events in the East as compared to the West.

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