

Development and Application of the CAMx Regional One-Atmospheric Model to Treat Ozone, Particulate Matter, Visibility, Air Toxics and Mercury

Paper # 549

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ABSTRACT

The Comprehensive Air quality Model with extensions (CAMx) is a three-dimensional photochemical grid dispersion model that allows for an integrated “one-atmosphere” assessment of gaseous and particulate matter (PM) air pollution over many scales ranging from urban to super-regional. Recently, CAMx Version 4 was released including a “one-atmosphere” particulate matter (PM) module that treats PM in two sections, as fine (particles smaller than 2.5 microns in diameter) and coarse (particles larger than 2.5 microns in diameter) particles. The CAMx4 PM module includes complete inorganic aerosol thermodynamics (ISORROPIA), semi-volatile secondary organic aerosol equilibrium (SOAP), a bulk-phase aqueous chemistry module (RADM) and a new wet-deposition scheme. A more detailed “full-science” PM treatment has also been implemented in CAMx (PMCAMx) where the particle size distribution is represented as a number of fixed size sections (e.g., ten sections) and also includes a variable size resolution aqueous-phase chemistry module to better describe cloud droplets of different sizes as well as options for fully dynamic, equilibrium (ISORROPIA) and hybrid aerosol thermodynamics.

This paper describes the combining of the “full-science” PMCAMx modules within the CAMx4 framework so that the “one-atmosphere” and “full-science” PM treatments are implemented within the same CAMx platform. The new CAMx model is evaluated for a Los Angeles PM episode and the western US for the 1996 year. Results from the “one-atmosphere” and “full-science” PM treatments in CAMx are evaluated including:

- Effects of a two-section versus multi-section treatment of PM size distribution.
- Effects of a single section bulk versus multi-section aqueous-phase chemistry module.
- Effects of equilibrium versus dynamic versus hybrid treatment of aerosol thermodynamics.

INTRODUCTION

The Comprehensive Air-quality Model with Extensions (CAMx) is a three-dimensional multi-scale photochemical grid model¹ that is publicly available (www.camx.com) without fees or restrictions on model application. CAMx was developed with all new code during the late 1990s using modern and modular coding practices. This has made the model an ideal platform for extension to treat a variety of air quality issues including ozone, particulate matter (PM), visibility, acid deposition, air toxics, and mercury. The flexible CAMx framework has also made it a convenient and robust host model for the implementation of “probing tool” techniques such as Process Analysis, the Decoupled Direct Method (DDM) and the Ozone Source Apportionment Technology (OSAT).^{2,3,4} The Coordinating Research Council (CRC) have sponsored ENVIRON, together with Sonoma Technology, Inc. (STI) and Carnegie Mellon University

(CMU), to add state-of-the-science algorithms for aerosol modeling to CAMx to create a new particulate matter model called PMCAMx (version 3.01).⁵

Development of PMCAMx

The development of PMCAMx was based on version 3.01 of CAMx as distributed from <http://www.camx.com>. The extension to treat PM involved the addition of science modules to represent important physical processes for aerosols:

- Size distribution is represented using the Multi-component Aerosol Dynamics Model (MADM), which uses a sectional approach to represent the aerosol particle size distribution.⁶ MADM treats the effects of condensation/evaporation, coagulation and nucleation upon the particle size distribution.
- Inorganic aerosol thermodynamics are represented using ISORROPIA^{7,8} within MADM.
- Secondary organic aerosol thermodynamics are represented using the semi-volatile scheme of Strader and co-workers.⁹
- Aqueous-phase chemical reactions are modeled using the Variable Size-Resolution Model (VRSM) of Fahey and Pandis,¹⁰ which automatically determine whether water droplets can be represented by a single ‘bulk’ droplet-size mode or whether it is necessary to use fine and coarse droplet-size modes to account for the different pH effects on sulfate formation.
- The CAMx deposition algorithms were improved for particle deposition. Dry deposition is represented for the size-resolved particle distribution¹.

PMCAMx incorporates the latest full-science sectional aerosol modules.⁵ The development and testing of PMCAMx took over a year.

Extensions of CAMx to Treat One-Atmosphere PM/ozone Processes

Although PMCAMx represents the latest full-science representation of aerosol processes, the computational requirements of the model may be too great to efficiently model annual periods. Thus, in parallel to the PMCAMx development, CAMx was extended to treat one-atmosphere PM/ozone processes. In addition, further refinements of the CAMx algorithms were implemented, including the inclusions of a one-atmosphere wet deposition algorithm a feature not included in PMCAMx. The result was the public release of the one-atmosphere CAMx Version 4¹ in early 2003 that included a PM treatment, called Mechanism 4 (M4) that includes the following three aerosol modules:

- RADM – bulk aqueous-phase chemistry module similar to one used in CMAQ¹¹
- ISORROPIA – inorganic aerosol thermodynamic model^{7,8}
- SOAP – secondary organic aerosol module⁹

M4 uses 2-section PM approach where primary species are modeled as fine and/or coarse particles while secondary species are modeled as fine particles. CAMx Version 4 (CAMx4) with

the Mechanism 4 (M4) 2-Section PM treatment was publicly released in April 2003 (www.camx.com).

Although CAMx4 was publicly released via the CAMx website (www.camx.com) to all, because of the need for further testing and evaluation, PMCAMx was only released when requested. During Fall 2003, the CRC sponsored a study to combine the PMCAMx full-science sectional aerosol algorithms within the one-atmosphere CAMx4 platform so that the one-atmosphere and full-science PM approaches are available in the same model that is currently called CAMx4+.

INITIAL TESTING OF THE CAMx4 MODEL USING THE SOUTHERN CALIFORNIA DATABASE

The new CAMx4+ model can be run in several configurations using various combinations of the one-atmosphere CAMx4 and full-science PMCAMx modules. The M4 one-atmosphere option uses the CAMx4 2-Section approach with the RADM aqueous-phase chemistry module and ISORROPIA equilibrium aerosol thermodynamic module. The PMCAMx full-science N-Section approach can be paired with either the RADM 1-Section or CMU Variable Size Resolution Module (VSRM) multi-section aqueous-phase model using either the ISORROPIA equilibrium (EQ), MADM fully dynamic (MA), or Hybrid (HY) aerosol thermodynamic modules.

The aerosol modules in the new CAMx4+ were tested for an October 17-18, 1995 episode in the Los Angeles area, which was also used for the initial PMCAMx testing⁵. 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 and thus aerosol species of the inputs are size-resolved. A pre-processing tool is developed to convert the size-resolved emissions, IC, and BC inputs to those compatible with M4. Initially, five different CAMx4+ model configurations were analyzed.

- M4 -- Mechanism 4 2-Section with Equilibrium (ISORROPIA) aerosol thermodynamics and RADM aqueous-phase chemistry.
- VSRM/EQ -- 10-Section with Equilibrium (ISORROPIA) aerosol thermodynamics and VSRM aqueous-phase chemistry.
- VSRM/MA -- 10-Section with dynamic (MADM) aerosol thermodynamics with VSRM.
- VSRM/HY -- 10-Section with hybrid aerosol thermodynamics with VSRM.
- RADM/EQ -- 10-Section with Equilibrium (ISORROPIA) aerosol thermodynamics and RADM (also run using 4-Sections, 3 fine and 1 coarse).

Ozone

1-hr average ozone peak concentrations predicted by all model configurations agree quite well (within 3% of each other, Figure 1). The discrepancy between M4 and VSRM/EQ is mostly attributed to the difference between RADM and VSRM as ozone is an important component of aqueous-phase chemistry. The differences between predictions by VSRM/EQ, VSRM/HY and VSRM/MA are negligible.

Particulate Matter

24-hr average total PM_{2.5} mass predictions for the various model configurations show significant differences in some regions (Figure 2). Figures 3 and 4 display the predicted 24-hr average size distributions at cells where maximum positive and negative, respectively, differences between M4 and VSRM/EQ occur. Figures 5 and 6 display the differences between VSRM/EQ and VSRM/MA. These figures suggest the following:

- The M4 2-section approach predicts more fine PM than the size-resolved PM modules due the assumption that all secondary PM are in the fine mode that has a lower dry deposition rate than the coarser modes.
- 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 (EQ) predicts more fine ammonium nitrate than the dynamic model (MA).
- Size distributions predicted by HY and MA 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 1 shows the sectional boundary diameters used models with 4 and 10 sections. Size distributions predicted by RADM/EQ with 10 and 4 sections give relatively good agreement (Figure 7). While peak locations of nitrate and ammonium are slightly different, their PM_{2.5} and PM₁₀ predictions agree well.

Table 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 8 shows the computational requirements, in terms of CPU hours per simulation day, for the various model configurations. CPU 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 comparable to that of M4 (~20% slower). Use of 10 sections with RADM/EQ results in CPU

times that are 2.3 times slower than the 4-section version of RADM/EQ or almost 3 times slower than M4. Couched in terms of annual simulations using one Athlon 1600 CPU, M4, RADM/EQ with 4-sections and RADM/EQ with 10-sections would require approximately 6, 8 and 18 CPU days to complete, respectively for the Los Angeles 65 x 40 x 10 grid. Of course significant speed up in run times can be obtained by using multiprocessors and faster CPUs.

Conclusions from Initial Testing in Southern California

The aqueous-phase chemistry Variable Size Resolution Model (VSRM) is much slower than the RADM module and requires significantly more computational time. Also, there are conditions under which VSRM failed to converge in several cells. Thus at this time its use in annual production runs is not recommended and further refinement of the module is needed. However, its technical formulation is superior to RADM and the sulfate estimates are significantly different so that it should still be considered in sensitivity analysis.

For annual simulations where computational requirements are important, based on this work, M4 and/or RADM/EQ are recommended. RADM/EQ is as stable and fast (when using 4 sections) as M4 and can give more detailed information for the aerosol size distribution.

FURTHER EVALUATION USING A WESTERN US 1996 DATABASE

CAMx4+ was also applied to a western US modeling domain that was used by the Western Regional Air Partnership (WRAP) (www.wrpaair.org). This modeling domain covers the western US with a 95 by 85 36 km resolution grid and 18 vertical layers.¹² Several models have been run on the WRAP western US modeling domain for January and July 1996:

- CMAQ
- REMSAD
- CAMx4+

CAMx4+ was exercised using both the 2-section Mechanism 4 (M4) approach as well as using the RADM/EQ configuration with 4-sections (3 fine and 1 coarse). The IMPROVE monitoring network only speciates the fine fraction of PM and the IMPROVE Coarse Matter (CM) species is the difference in total mass between the PM₁₀ and PM_{2.5} measurements. Thus, any coarse sulfate or nitrates would be reported in the IMPROVE CM species. CMAQ, REMSAD and CAMx_M4 all assume that all sulfate and nitrate is in the fine mode. CAMx_4Sec, however, has a separate representation of the coarse component of sulfate. Thus, the CAMx_4Sec sulfate estimates are compared against the IMPROVE SO₄ measurements two ways:

- CAMx_4Sec (F) uses only the first three sections of sulfate (i.e., PM_{2.5}) for comparison with the IMPROVE SO₄ measurement; and
- CAMx_4Sec (C) uses all four sections (i.e., PM₁₀) for comparison with the IMPROVE SO₄ species (i.e., assume all sulfate is fine).

Figure 1. 1-hr average ozone peak predictions on October 18, 1995.

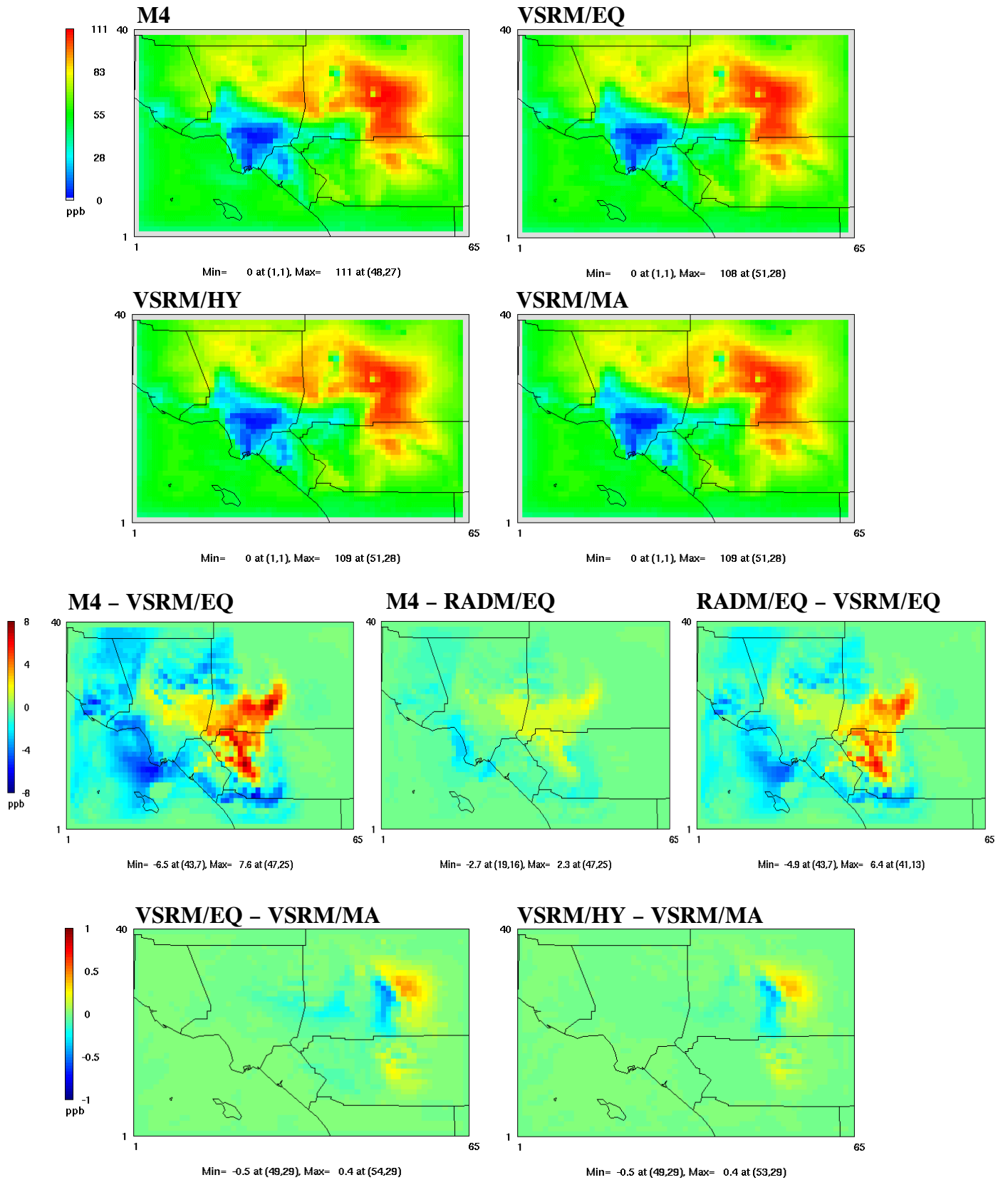


Figure 2. 24-hr average total PM_{2.5} predictions on October 18, 1995.

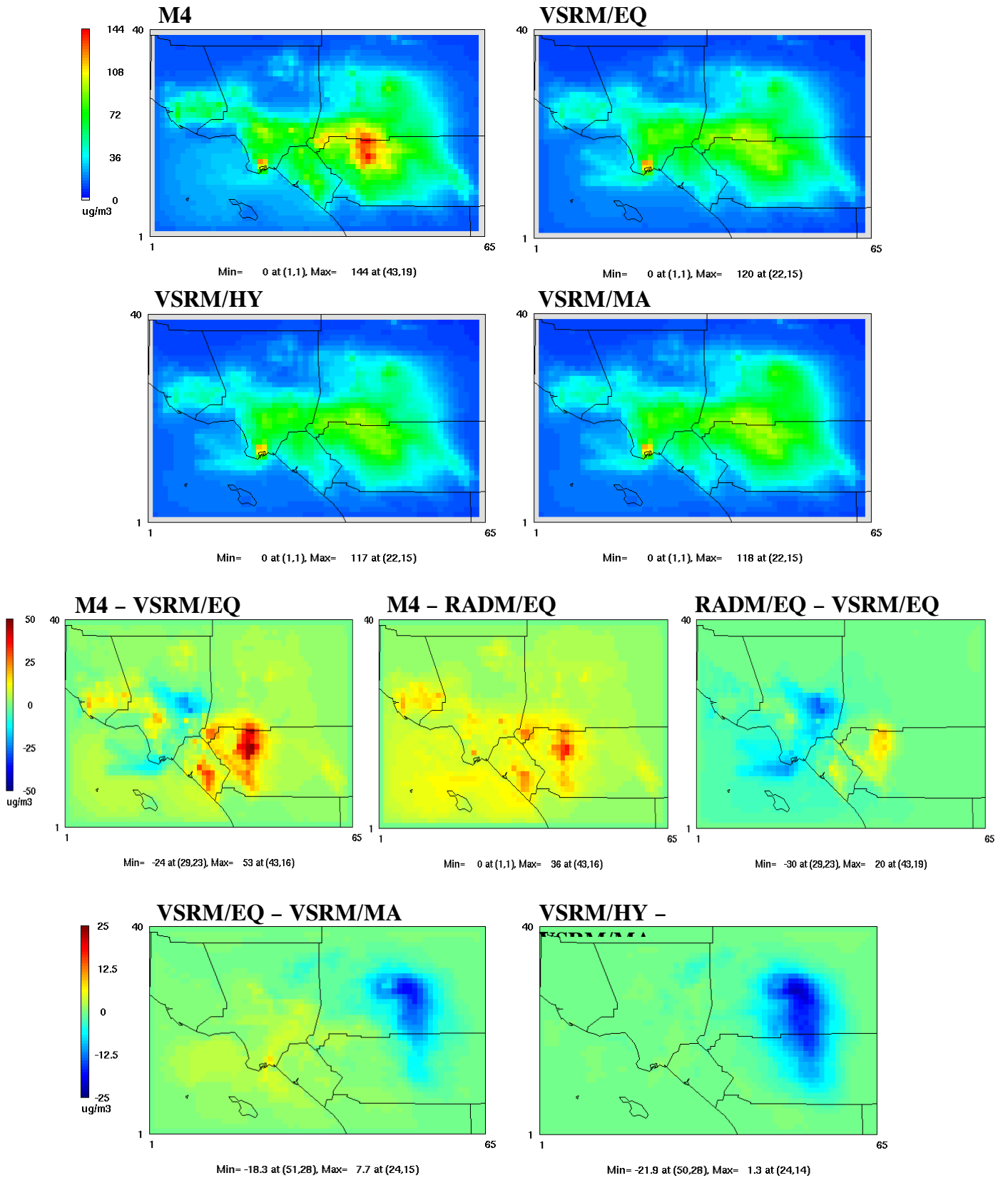


Figure 3. 24-hr average size distributions predicted at cell (43, 16).

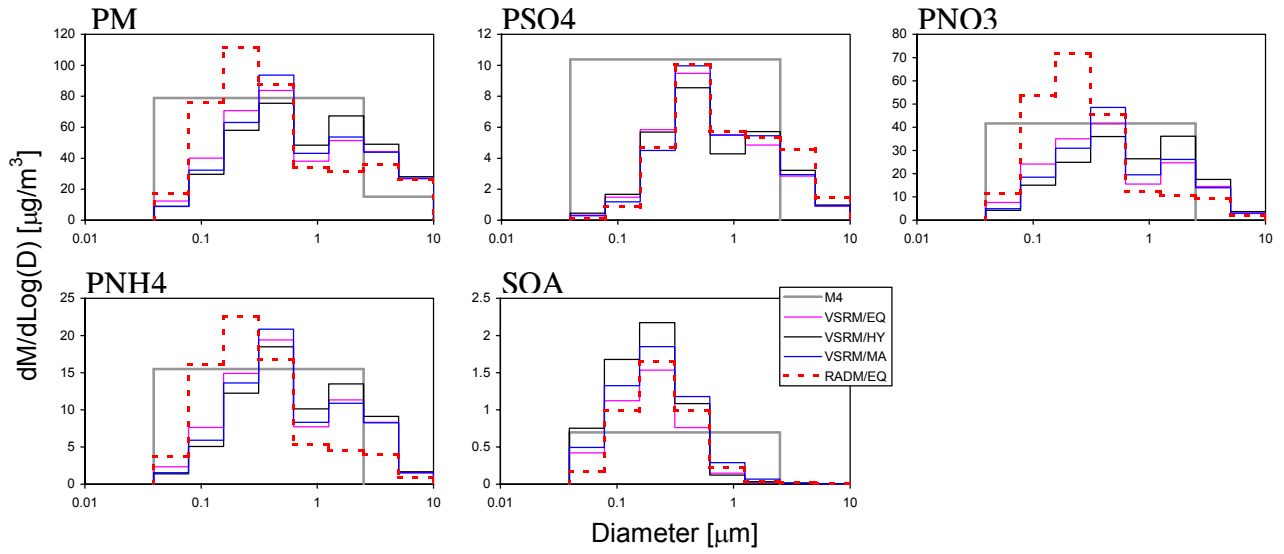


Figure 4. 24-hr average size distributions predicted at cell (29, 23).

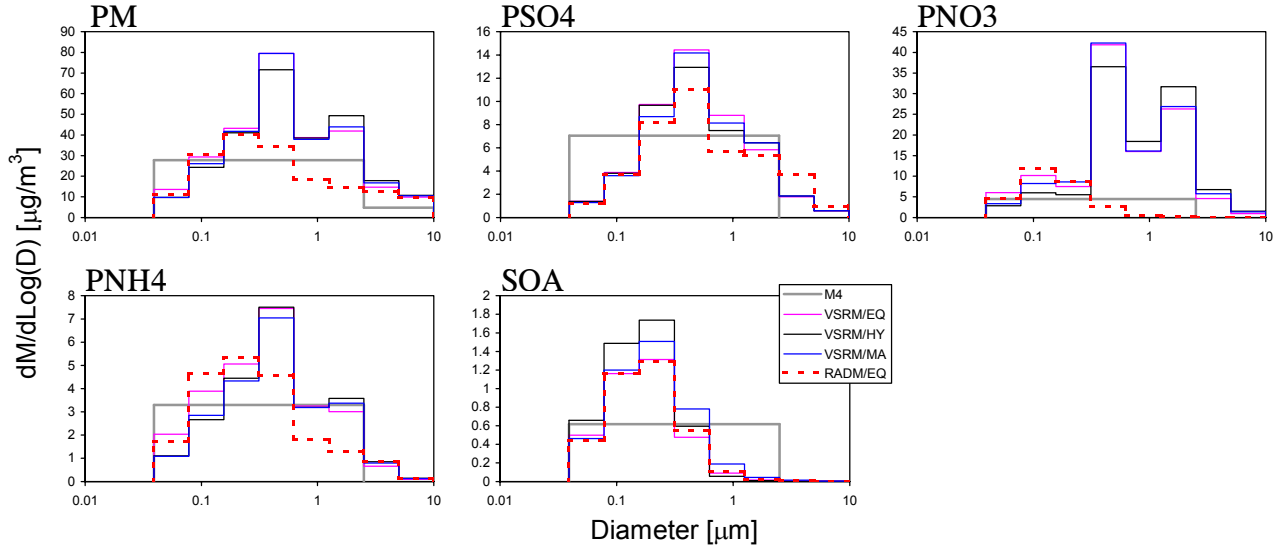


Figure 5. 24-hr average size distributions predicted at cell (24, 15)

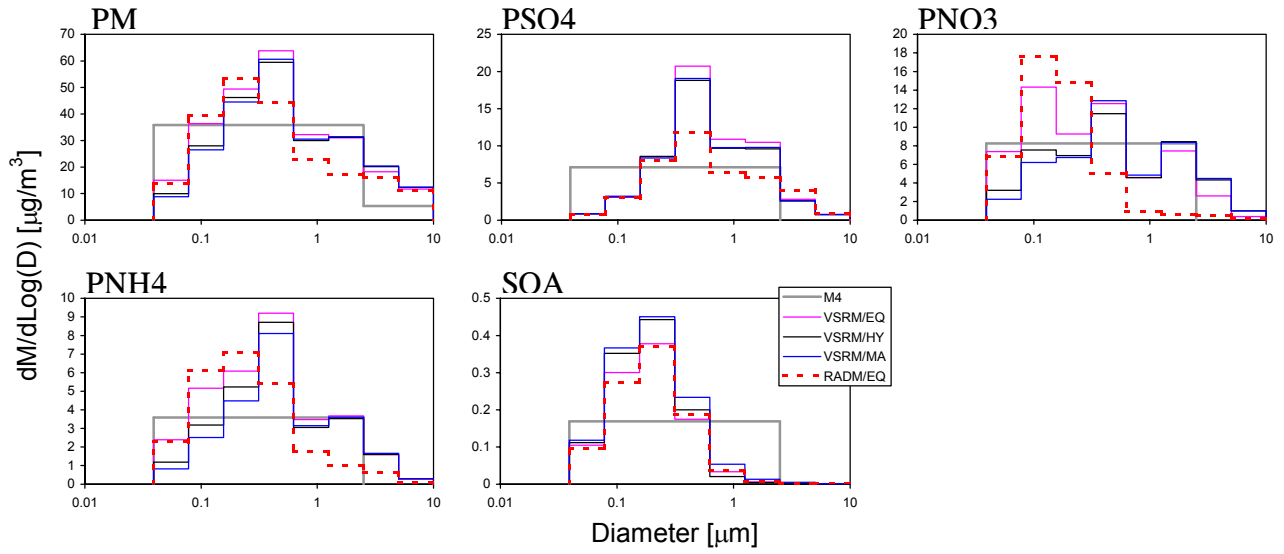


Figure 6. 24-hr average size distributions predicted at cell (50, 28).

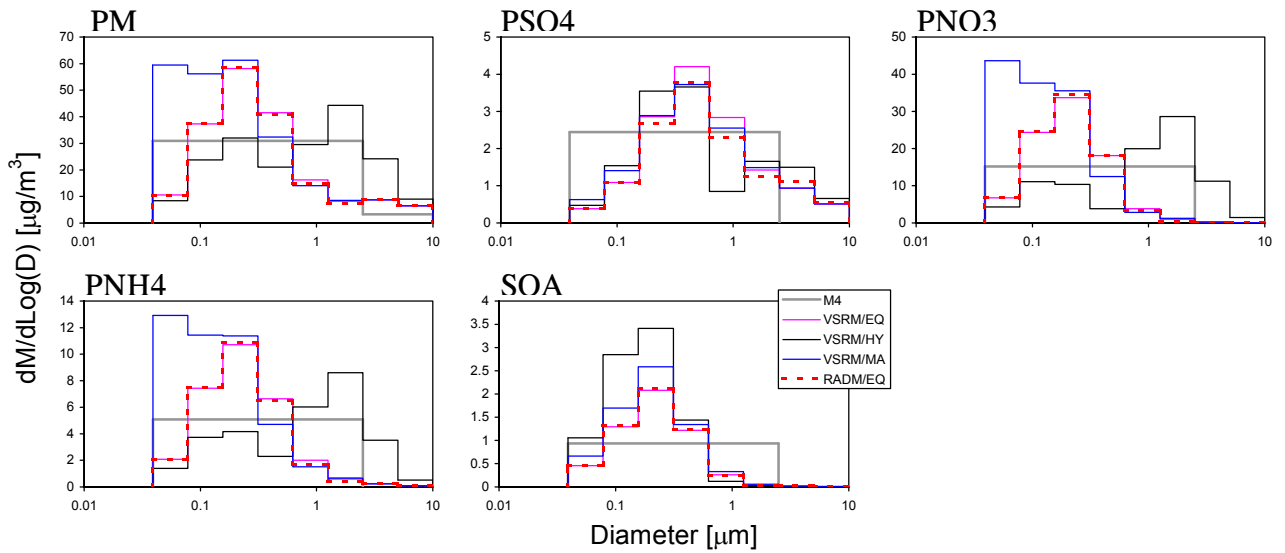


Figure 7. Size distributions predicted by RADM/EQ with 4 and 10 sections at cell (34, 16).

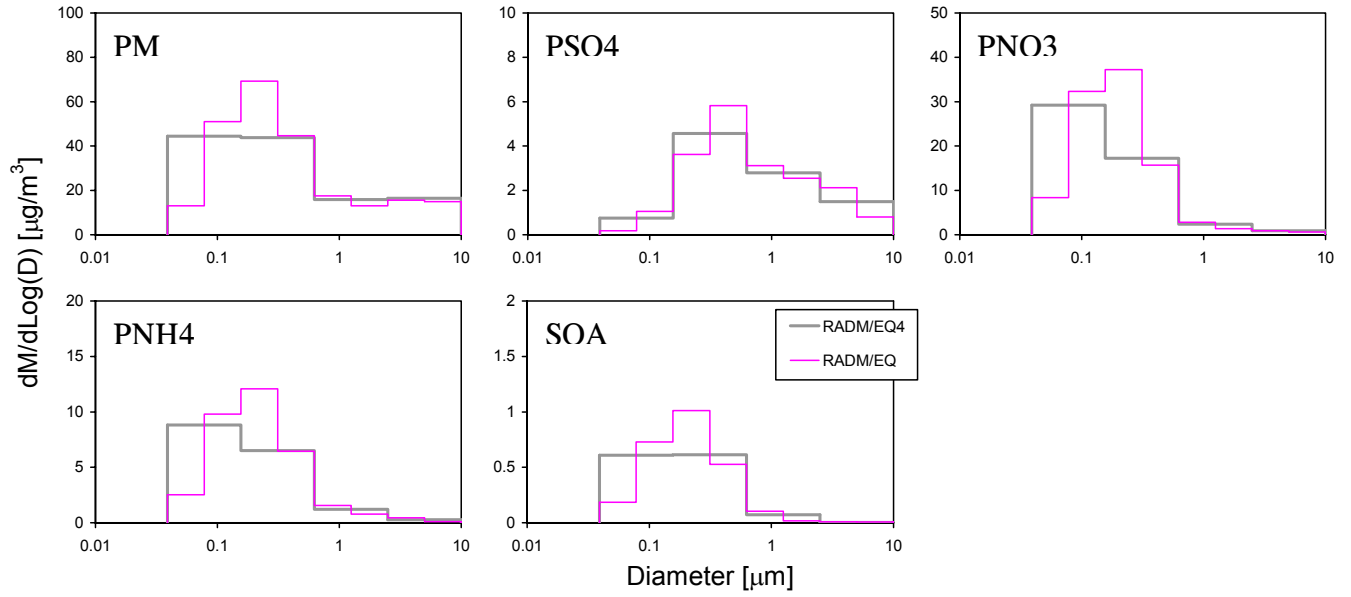
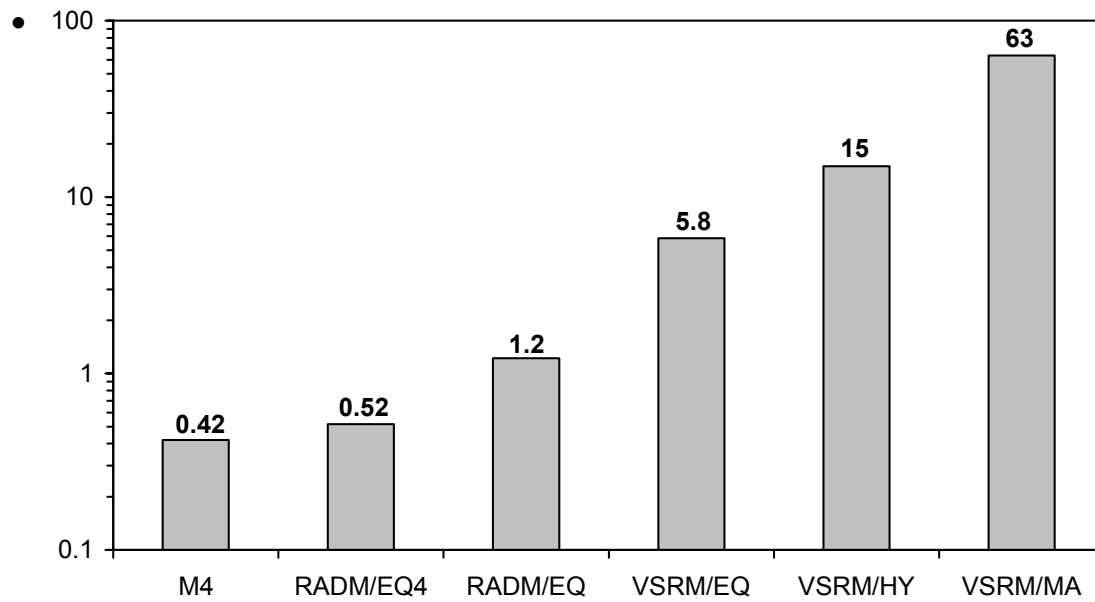


Figure 8. CPU hours per simulation day using a single Athlon 1600 CPU on the 65 x 40 5 km SCOS grid with 10 vertical layers.



Figures 9 and 10 compares the sulfate model performance for the four model configurations and, respectively, January and July 1996. Summary statistics are provided in Table 2. The models exhibit similar sulfate model performance with a slight overestimation bias in January and an underestimation bias in July. With the exception of the REMSAD fractional bias in January, all of the SO₄ bias measures are within approximately ± 30%, which is encouraging. Table 3 summarizes model performance for particulate nitrate (NO₃). The nitrate performance is not as good as seen for sulfate with all the models exhibiting an overestimation bias in January and underestimation bias in July whose magnitudes ranges from 30% to 200%. The comparisons of the CAMx_4Sec (F) and CAMx_4Sec (C) results suggest that approximately 10% of the average SO₄ and NO₃ in the western US is estimated to be coarse. However, we would expect a lot of spatial variation in the coarse fraction due to the higher sodium nitrate near the coast that is typically in the coarse mode.

CAMx_4Sec required approximately twice the computer time as CAMx_M4. Why CAMx_4Sec (RADM/EQ) takes twice the computing time with the western US application but only 20% more for the Southern California application is under study.

Table 2. Fractional bias and normalized mean bias statistics performance measures for sulfate (IMPROVE SO₄) in the western US and the five model configurations.

SO₄	January 1996		July 1996	
Model	Fract. Bias	Mean Bias	Fract. Bias	Mean Bias
CMAQ	24	15	-33	-30
REMSAD	51	33	-39	-33
CAMx_M4	31	16	-12	-7
CAMx_4Sec (F)	9	-12	-26	-23
CAMx_4Sec (C)	18	-3	-22	-18

Table 3. Fractional bias and normalized mean bias statistics performance measures for particulate nitrate (IMPROVE NO₃) in the western US and the five model configurations.

SO₄	January 1996		July 1996	
Model	Fract. Bias	Mean Bias	Fract. Bias	Mean Bias
CMAQ	53	122	-97	-47
REMSAD	55	127	-132	-57
CAMx_M4	47	201	-120	-31
CAMx_4Sec (F)	87	217	-110	-30
CAMx_4Sec (C)	92	231	-22	-17

Figure 9. Comparison of January 1996 sulfate (SO₄) model performance in the western US using IMPROVE measurement data and: (1) CMAQ vs. REMSAD (top, left); (2) CMAQ vs. CAMx_M4 (top, right); (3) CMAQ vs. CAMx_4Sec (F) (bottom, left); and (4) CMAQ vs. CAMx_4Sec (C) (bottom, right).

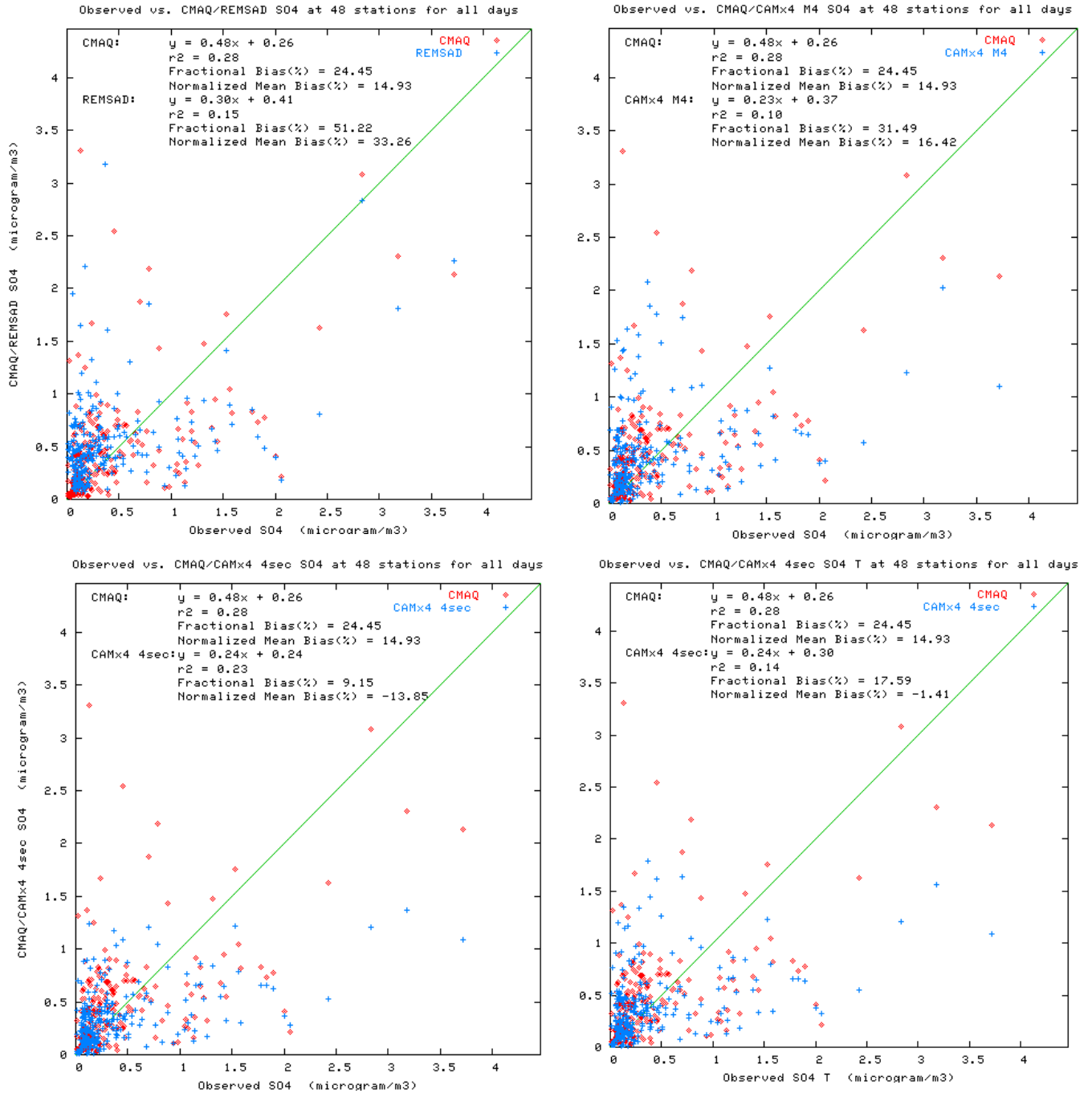
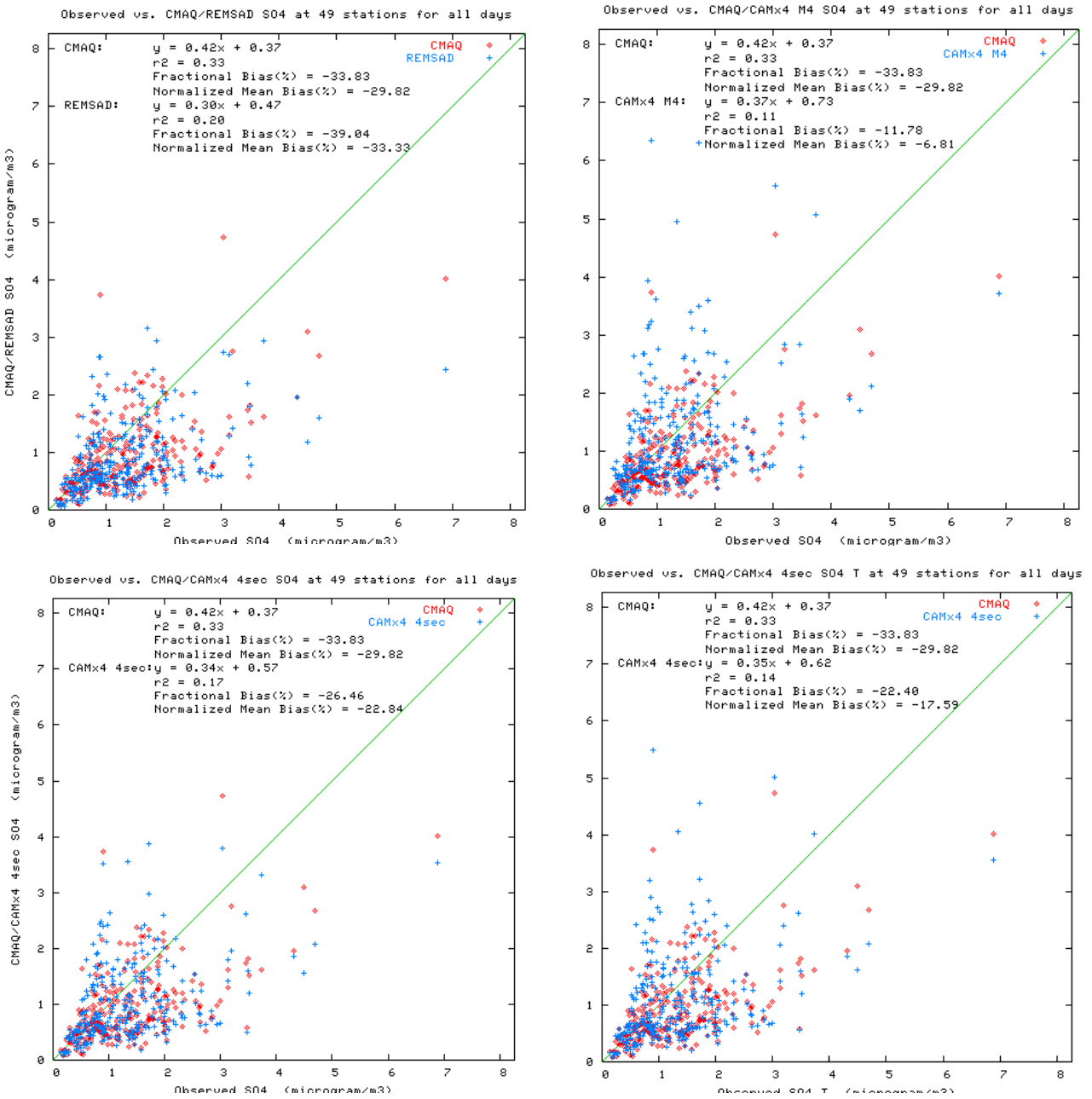


Figure 10. Comparison of July 1996 sulfate (SO₄) model performance in the western US using IMPROVE measurement data and: (1) CMAQ vs. REMSAD (top, left); (2) CMAQ vs. CAMx_M4 (top, right); (3) CMAQ vs. CAMx_4Sec (F) (bottom, left); and (4) CMAQ vs. CAMx_4Sec (C) (bottom, right).



CONCLUSIONS

The one-atmosphere 2-Section Mechanism 4 (M4) and full-science section PM algorithms have been successfully implemented and tested in the CAMx4+ modeling system for Southern California and the western US. The Southern California application tested the one-atmosphere M4 and full-science algorithms and found:

- The RADM and VSRM aqueous-phase chemistry modules produce significantly different sulfate estimates, with the multi-section VSRM module generally estimating higher sulfate. However, the VSRM requires significantly more computer time such that more refinements and optimization are needed before it can be used routinely for annual modeling.
- The use of the MADM dynamic and HYBRID aerosol thermodynamic modules produce results that are comparable to the equilibrium approach (ISORROPIA). Thus, given the increased computer time of the dynamic and hybrid approaches, their use for annual modeling cannot be justified at this time.

The western US application compared the CMAQ, REMSAD, CAMx_M4 and CAMx_4Sec models for a winter and summer month (January and July 1996). In general, the models overestimated sulfate and nitrate in the winter and underestimated them in the summer, with the model performance for sulfate exhibiting more skill than for nitrate. The models exhibited similar trends in model performance. However, it is difficult to say whether one model is exhibiting significantly more skill than another.

Currently there are numerous uncertainties in many of the PM model inputs and science algorithms. Ammonia and fugitive dust emissions are highly uncertain and refinements are under development. Nitrate formation algorithms is an area where more research and development is needed. The computational requirements of some of the full-science algorithms, such as the VSRM aqueous-phase chemistry and the MADM dynamic and hybrid aerosol thermodynamic modules, are such that they are currently not practical for annual modeling. However, it is encouraging that the current generation of one-atmosphere models (e.g., CMAQ, CAMx_M4 and CAMx_4Sec) are capable of efficiently simulating an annual period such that the simplification of the science modules for computational efficiency (e.g., micro-CB4 in REMSAD) is no longer needed.

ACKNOWLEDGEMENTS

The development of PMCAMx and CAMx4+ was funded by the Coordinating Research Council (CRC) Atmospheric Impacts committee under CRC Projects A-30 and A-44. The PM modules implemented in CAMx are the result of over two decades of research involving numerous researchers at the California Institute of Technology, Carnegie Mellon University, and other research institutions. The 1996 comparisons benefited from work performed by the WRAP Regional Modeling Center (RMC) hosted at the University of California at Riverside.

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