5.4 FINE PARTICULATE MATTER MODELING IN CENTRAL CALIFORNIA, PART II: APPLICATION OF THE COMMUNITY MULTISCALE AIR QUALITY MODEL

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1. INTRODUCTION

Fine particulate matter ($PM_{2.5}$, particles with aerodynamic diameters of 2.5 µm or less) is the most recent criteria pollutant to be regulated by the US EPA. Much of central California has been designated into nonattainment of the $PM_{2.5}$ 24-h National Ambient Air Quality Standard (NAAQS) of 35 $\mu g/m^3$. Exceedances of the $PM_{2.5}$ 24-h NAAQS occurred almost exclusively during the winter months. Air quality planning requires photochemical modeling of winter season $PM_{2.5}$ episodes.

$PM_{2.5}$ modeling was conducted using the US EPA Community Multiscale Air Quality (CMAQ) model. Historically, meteorological inputs to CMAQ and other air quality models applied in central California were prepared using the Fifth-Generation Pennsylvania State University/National Center for Atmospheric Research (NCAR) Mesoscale Model version 5 (MM5). MM5 was successfully applied for modeling both summer ozone and winter $PM_{2.5}$ episodes. MM5 is, however, a discontinued model that is no longer supported. Therefore, the Weather Research and Forecasting (WRF) model is being explored as a potential replacement for MM5. The goal of this study is to test and evaluate the WRF-CMAQ modeling system in preparation of air quality model applications over central California.

This paper describes the second part of a two-part study focusing on developing WRF simulations to drive air quality models over central California (Rogers et al., 2011). Part II of the study applies and evaluates the performance of the CMAQ model following the US EPA guidelines (US EPA, 2007) as closely as feasible. A seasonal modeling approach allowed evaluation of WRF-CMAQ performance across the full range of typical winter-season weather patterns. Sensitivity runs of the model were conducted to estimate $PM_{2.5}$ sensitivities to changes in emissions of directly emitted $PM_{2.5}$, NO_x, VOC, sulfur species, and ammonia.

2. STUDY DOMAIN

Central California (Figure 1) comprises three contiguous regions, most or all of which are non-attainment areas for the 24-hr $PM_{2.5}$ NAAQS: the coastal San Francisco Bay Area (SFBA), the Sacramento Valley (SV), and the San Joaquin Valley (SJV). The inland SV and SJV together form the Central Valley (CV). These three regions connect at the Delta. Extensive analysis of nearly a decade of measurements allowed development of a conceptual model for $PM_{2.5}$ episodes.

Winter $PM_{2.5}$ episodes usually developed under: stable atmospheric conditions inhibiting vertical dispersion; clear and sunny skies favoring enhanced secondary $PM_{2.5}$ formation; pronounced overnight drainage (downslope) flows off the CV rims, causing convergence of the low-level winds toward the CV floor; and an emptying of the accumulated CV air mass through the Delta and into the SFBA along its eastern boundary (see Figure 1). Aloft weather systems strongly influenced the surface winds that determined $PM_{2.5}$ levels. Surface conditions stagnated when an aloft high pressure system moved over central California. Persisting high pressure conditions allowed $PM_{2.5}$ to accumulate to the exceedance level in the SFBA typically after 2-4 days.

The composition of $PM_{2.5}$ during episodic conditions varied substantially throughout the study domain. Primary $PM_{2.5}$ levels were elevated around densely populated areas, major roadways, and regions with intense commercial and/or industrial activity. Secondary $PM_{2.5}$, mostly inorganic ammonium nitrate, accumulated regionally, especially in the sheltered inland valleys. Primary $PM_{2.5}$ accounted for approximately two-thirds, one-half, and one-third of the total $PM_{2.5}$ for the SFBA, SV, and SJV, respectively.

3. MODEL SETUP

The air quality modeling domain (Figure 1) included all low-lying areas within central California over which elevated winter $PM_{2.5}$ levels were monitored. It covered the SFBA, the SV, and the SJV. Outlying areas within the modeling domain included over the Pacific Ocean, coastal locations along the Coast Range, and the inland Sierra Nevada. The simulation period was 1 December 2006 through 2 February 2007, spanning 63 consecutive days.

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CMAQ version 4.7.1 was implemented using the SAPRC99 chemical mechanism, the Models-3 AE4 aerosol module, and the RADM aqueous-phase chemistry model. CMAQ horizontal grid resolution was 4 km with 185×185 grid cells. There were 20 vertical layers. The bottom layer was approximately 22 m thick at sea level. Initial and boundary conditions for CMAQ were obtained from specialized measurements obtained during the California Regional Particulate Air Quality Study (CRPAQS) conducted during 1999-2001.

Meteorological fields were prepared using WRF. Details of the meteorological modeling are described in Part I of this study (Rogers et al., 2011).

Emissions were prepared mostly from an annual inventory for base year 2000 supplied by the California Air Resources Board (ARB). The SFBA portion of this inventory was replaced by the BAAQMD planning inventory that was augmented to reflect regional data for base year 2005. Household wood burning emissions for the SFBA were estimated based on the results of telephone surveys and were spatially allocated in proportion to the density of single family residences. Wood smoke represented around 35% of the direct $PM_{2.5}$ emissions for the SFBA. An ammonia inventory was developed for the SFBA using a bottom-up approach to estimate activity levels from eight source categories. The ammonia inventory estimated using local data had emissions levels around 40% higher than present in the National Emissions Inventory (NEI). The ammonia emissions estimates were the most uncertain of all species in the inventory. Anthropogenic emissions were adjusted for the 2006-07 winter season using ARB Almanac Emission Projection Data. A modeling inventory with day-of-week specific emissions was prepared for the single week 17-23 December 2006. Biogenic emissions were prepared using meteorological data from this week, which exhibited climatologically representative conditions. They were gridded using the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System.

4. RESULTS

Pollutant levels simulated by WRF-CMAQ were compared against observations using paired statistics for: $PM_{2.5}$; its various components elemental carbon (EC), organic carbon (OC), nitrate ion, ammonium ion, and sulfate ion; and gas phase species ozone and NO_x. Monitoring data were paired in space and time against simulated 24-h pollutant levels from the grid cell in which the monitor was located. Here, soccer goal plots were used to visualize statistics for mean fraction bias (MFB) and mean fractional error (MFE), where *model* and *obs* indicate the paired simulated and observed pollutant levels, respectively, for a sample size of *N* exceedance days.

$$\mathrm{MFB} = \frac{2}{N}\sum_{1}^{N}\frac{(model-obs)}{(model+obs)} \times 100\% \quad (1)$$

$$\mathrm{MFE} = \frac{2}{N}\sum_{1}^{N}\frac{|model-obs|}{(model+obs)} \times 100\% \quad (2)$$

Figure 2 plots the statistical performance metrics based on the 17 days for which $PM_{2.5}$ levels exceeded the 24-h $PM_{2.5}$ NAAQS in the Bay Area. One limitation with this diagram is that MFE is constrained to be greater than or equal to the absolute value of MFB. Therefore, half of the area of each soccer goal plot is “out of play” and is shaded grey.

The performance to simulate primary $PM_{2.5}$ was best indicated by the statistics for EC. San Jose had near-zero MFB and MFE around 15%, indicating that better performance was obtained within the SFBA than for the inland regions. For the inland urban sites at Sacramento and Fresno, EC levels were significantly underestimated with MFE ≈ -MFB ≈ 50%. Despite these significant biases, the statistics for EC were generally better than for any other $PM_{2.5}$ component at a given location. The ability to simulate primary $PM_{2.5}$ better than components that include secondary $PM_{2.5}$ suggests that transport and dispersion characteristics may be reasonably simulated.

The performance to simulate secondary $PM_{2.5}$ was best indicated by the statistics for nitrate and ammonium, and to a lesser extent for sulfate. San Jose was the only location for which these secondary $PM_{2.5}$ components were reasonably simulated with near-zero bias. Both nitrate and ammonium were consistently and strongly underestimated for the inland locations. Performance to simulate sulfate was better than for the other inorganic secondary $PM_{2.5}$ components. Consistent underestimation occurred at all sites except San Jose, where the bias was near-zero. The sulfate statistics are not very relevant, however, because sulfate did not contribute strongly to central California $PM_{2.5}$.

Statistics for OC reflect model performance to simulate both primary and secondary $PM_{2.5}$. Consistent significant underestimation for OC levels occurred at all sites. The underestimation of OC levels was moderate for the SFBA and severe for the inland locations. The OC underestimation may have resulted from performance issues to simulate primary OC mostly related to underestimated wood burning emissions and/or secondary OC mostly related to underestimated chemical conversion of biogenic emissions.

Statistics for (total) $PM_{2.5}$ indicated consistently severely underestimated levels with MFE ≈ -MFB ≈ 100% at all locations. Statistics for (total) $PM_{2.5}$ were generally quite poorer than statistics for the $PM_{2.5}$ components. This discrepancy occurred due to different operating schedules for the instruments. $PM_{2.5}$ was measured every day, and sample sizes for the $PM_{2.5}$ statistics were quite larger than for the $PM_{2.5}$

component statistics. $PM_{2.5}$ levels were measured on days with strong downward model biases for which $PM_{2.5}$ component levels were not measured. Similar $PM_{2.5}$ statistics were obtained at over 40 other $PM_{2.5}$ monitoring locations that lacked $PM_{2.5}$ component measurements (not shown).

Statistics for ozone and NO_x reflect model performance to simulate the atmospheric photochemistry upon which daytime secondary ammonium nitrate $PM_{2.5}$ formation depends. For all locations, ozone levels were consistently strongly overestimated with $MFE \approx MFB \approx 50\%$ and NO_x levels were consistently strongly underestimated in the approximate range $-100\% < MFE \approx MFB < -50\%$. Similar ozone and NO_x statistics were obtained at large numbers of other monitoring locations for which ozone levels differed from background levels. Despite significant biases in magnitude, the simulated spatial pattern for ozone was generally consistent with observations. During the winter season, central California ozone levels were observed to be regionally titrated below background levels over the SFBA, the SJV, and the SV.

Beyond the US EPA guidelines, the simulation results were also evaluated in terms of the distribution of primary and secondary $PM_{2.5}$. Results are shown for the three monitoring locations in Figure 3. The distributions indicate the relative frequencies of occurrence for different combinations of primary and secondary $PM_{2.5}$ in a 3×3 array of grid cells around each monitoring location, pooled for the 17 24-h $PM_{2.5}$ exceedance days for the SFBA during the 2006-07 modeling period. The Sacramento results were contaminated by two grid cells containing a major highway that did not appear to strongly impact the monitoring location. This artifact appears as a small cloud of data labeled as “Local source” on Figure 3. Otherwise, the relative distributions of primary and secondary $PM_{2.5}$ were consistent with observations (described in section 2). San Jose was dominated by primary $PM_{2.5}$ (data below the 1:1 line), Sacramento had roughly equal primary and secondary $PM_{2.5}$ levels (data along the 1:1 line), and Fresno was dominated by secondary $PM_{2.5}$ (data above the 1:1 line).

Preliminary “sensitivity” runs of the model were conducted using modified emissions inventories. Five sensitivity runs were conducted, with 20% across-the-board anthropogenic emissions reductions by pollutant category: NO_x and VOC combined, ammonia, sulfur species, direct $PM_{2.5}$, and all anthropogenic emissions (NO_x, VOC, ammonia, sulfur species, and direct $PM_{2.5}$). The rationale for simulating combined NO_x and VOC reductions was that, historically, these emissions have been reduced by similar levels over previous decades.

Results of the sensitivity runs are shown in Figure 4. Reducing directly emitted $PM_{2.5}$ was by far the most efficient means to reduce $PM_{2.5}$ levels, in terms of percentage change in $PM_{2.5}$ level per percentage change in emissions. Simulated primary $PM_{2.5}$ levels were reduced with a 1:1 ratio in response to direct $PM_{2.5}$ emissions reductions. (A 20% reduction in primary $PM_{2.5}$ level occurred for a 20% reduction in direct $PM_{2.5}$ emissions throughout the modeling domain.) Reductions for combined NO_x and VOC emissions were relatively ineffective or even exhibited a slight disbenefit (yellow shading in Figure 4) for certain locations. These results, however, may conflict with the observed trend for $PM_{2.5}$ nitrate levels, which decreased as emissions were reduced over the last decade. Reductions of ammonia emissions were somewhat effective for reducing regional secondary $PM_{2.5}$ levels. Simulated $PM_{2.5}$ levels were reduced by up to around a 1:5 ratio (i.e. up to 4% decrease in $PM_{2.5}$ level for a 20% reduction in ammonia emissions). Reducing sulfur emissions was relatively ineffective because of the initial low level for sulfur emissions. Reducing all emissions simultaneously produced reductions of $PM_{2.5}$ levels roughly equal to the sum of the other four simulation results, indicating a roughly additive nature for the response of reducing different emissions types.

5. DISCUSSION

Model performance was generally best within the SFBA, the area for which the most recent emissions estimates were available. Still, however, significant and consistent underestimation of the $PM_{2.5}$ levels and its components were common for most locations. Model underestimations of $PM_{2.5}$ levels were generally stronger for days with higher observed $PM_{2.5}$ levels.

Despite the model biases, WRF-CMAQ was able to simulate the proper proportions of primary and secondary $PM_{2.5}$. Moreover, the spatial gradients for the primary-secondary $PM_{2.5}$ split were realistic. This behavior implies that the simulated source-receptor relationships may have been realistic. Therefore, the model results, when used in a relative sense by applying *relative response factors* (RRF), may be accurate.

Model sensitivity runs were preliminary but offered several insights. First, the sensitivity runs highlighted the importance of controlling direct $PM_{2.5}$ emissions as the most efficient means to reduce $PM_{2.5}$ levels. Estimates for the sensitivity of $PM_{2.5}$ to precursors, however, may be uncertain. The relative insensitivity to NO_x and VOC emissions reductions may conflict with historical trends. The model did indicate significant benefits of reducing ammonia emissions. This model response appears qualitatively realistic, because ammonia directly reacts with a variety of atmospheric acids to form secondary ammonium $PM_{2.5}$. Reducing NO_x and VOC emissions, on the other hand, has an indirect and complex impact on $PM_{2.5}$ formation processes because NO_x is not directly converted to nitrate $PM_{2.5}$. The ammonia inventory was, however, the least certain aspect of the emissions inventory. Therefore, additional data gathering efforts should be focused to confirm and refine the ammonia emissions inventory to increase the level of certainty associated with the sensitivity run results.

REFERENCES

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US EPA, 2007. Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for ozone, $PM_{2.5}$, and regional haze. Publication EPA-454/B-07-002. 253 pp.

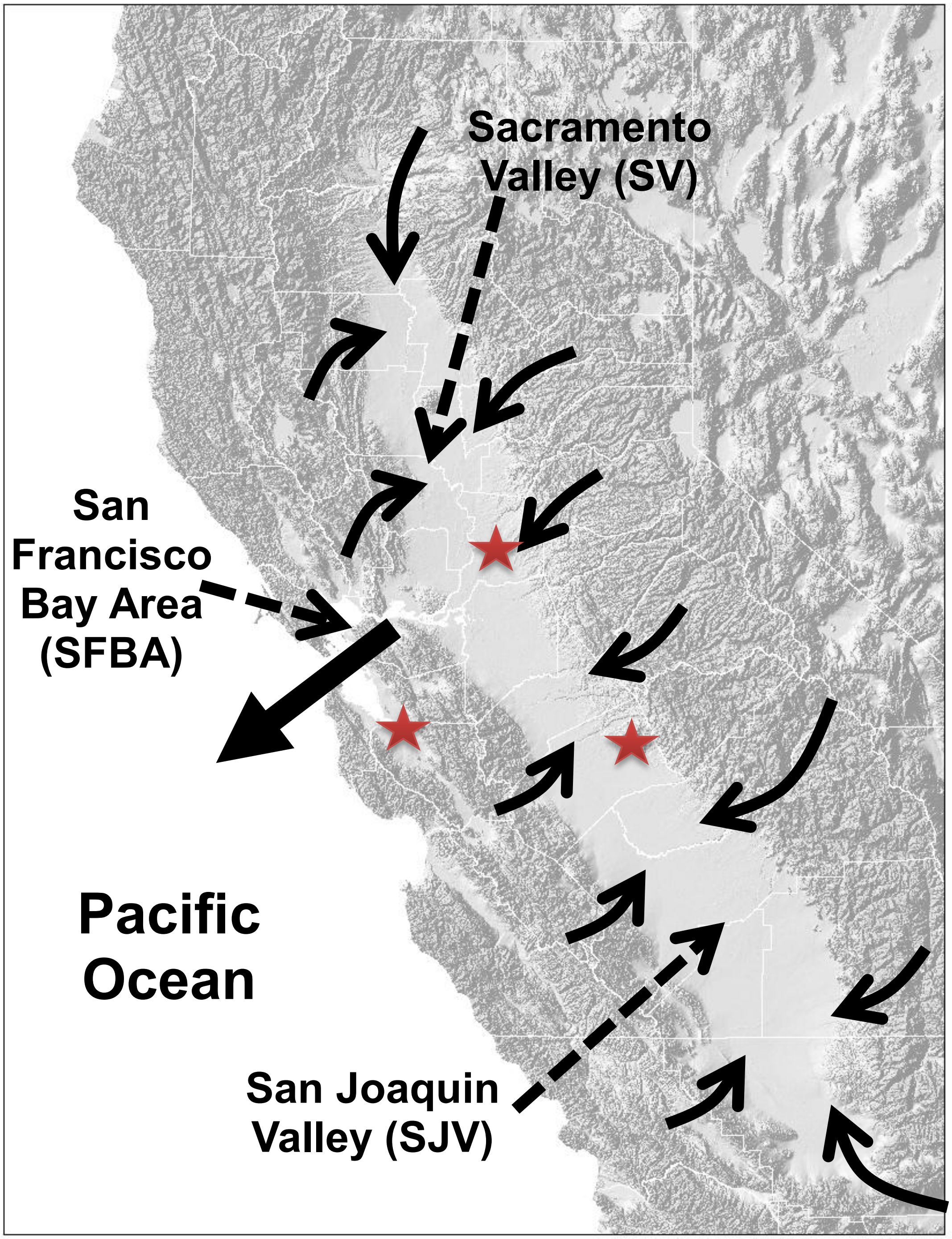


Figure 1. Central California air quality modeling domain (extent of map) showing three major air basins using dashed arrows: the SFBA, the SV, and the SJV. The Central Valley (CV) comprises the SV and the SJV. Red stars indicate positions of Speciation and Trends Network (STN) air quality monitors at San Jose (in the SFBA), Sacramento (in the SV), and Fresno (in the SJV). Solid arrows indicate conceptual model for $PM_{2.5}$ episodes occurring under an aloft high pressure system. Downslope flows over the CV rims converge toward the CV floor, and channeled flow exits the CV from east to west through the SFBA and toward the Pacific Ocean.

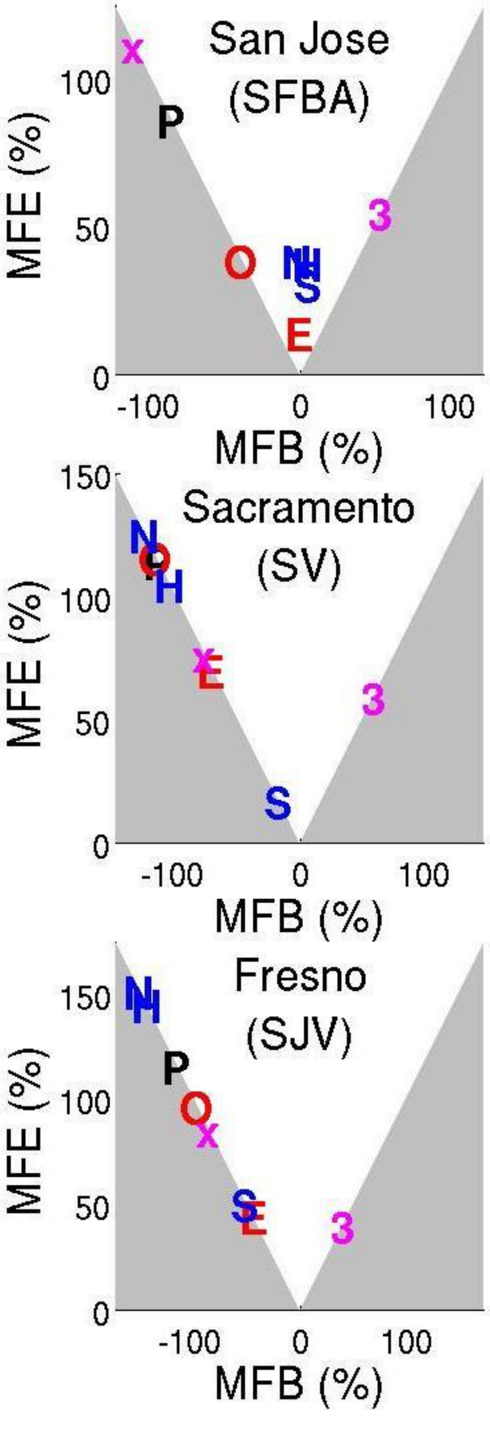


Figure 2. Model performance statistics on soccer goal plots for three locations. Statistics were computed for total $PM_{2.5}$ (black P), EC (red E), OC (red O), nitrate ion (blue N), ammonium ion (blue H), sulfate ion (blue S), ozone (magenta 3), and NO_x (red x). Statistics were computed between simulated and observed 24-h pollutant levels paired in space and time for the 17 SFBA 24-h $PM_{2.5}$ exceedance days from the 2006-07 simulation period.

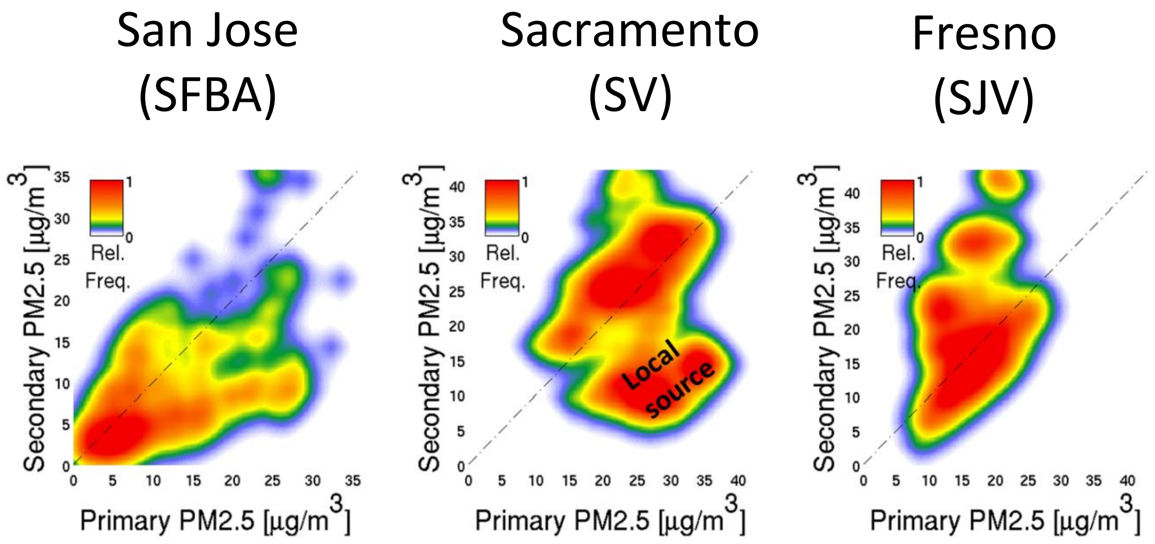
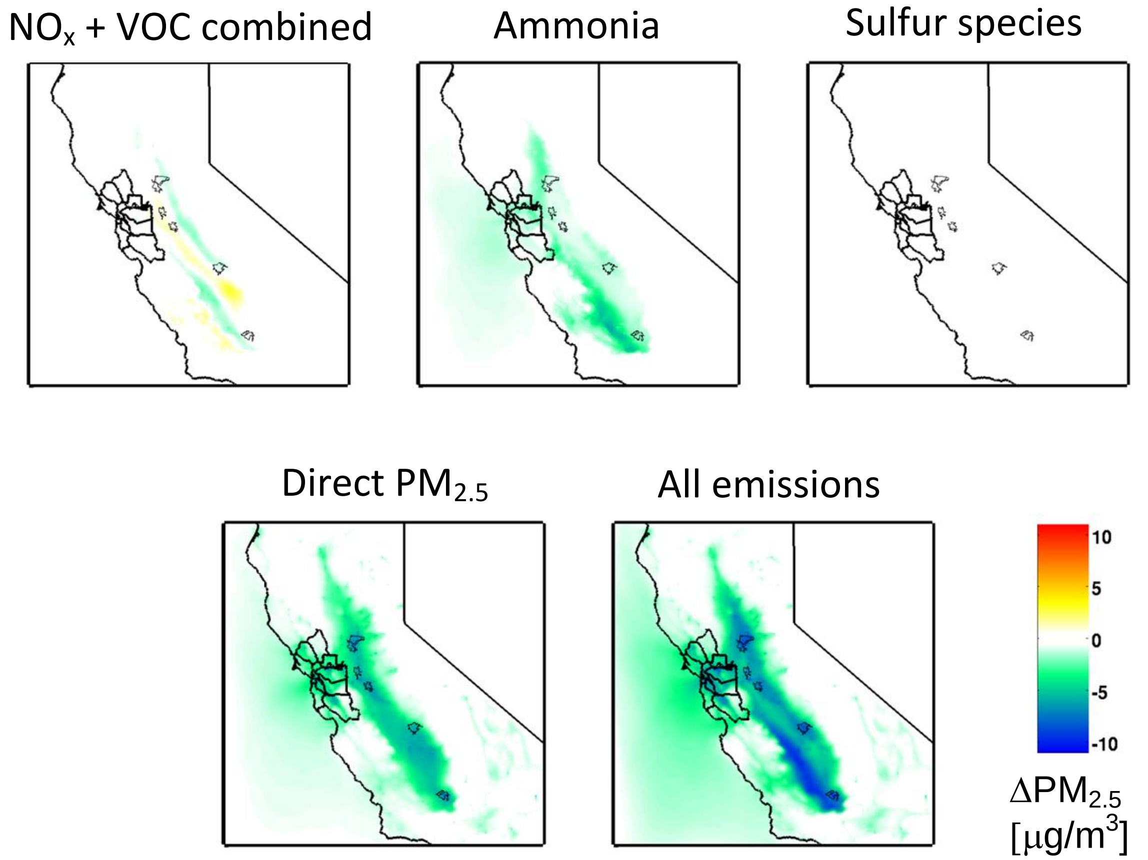


Figure 3. Distribution of simulated primary and secondary $PM_{2.5}$ levels for three monitoring locations. Distributions estimated for 3×3 array of grid cells around each monitoring location, for the 17 SFBA 24-h $PM_{2.5}$ exceedance days from the 2006-07 simulation period. The dashed 1:1 line indicates equal levels of primary and secondary $PM_{2.5}$. The cloud of data below the 1:1 line for Sacramento labeled "Local source" resulted from two grid cells containing a major highway whose emissions were not believed to strongly impact the Sacramento monitoring data.



NO_x + VOC combined

Ammonia

Sulfur species

Direct $PM_{2.5}$

All emissions

Figure 4. Sensitivity run results for five simulations with 20% across-the-board anthropogenic emissions reductions: NO_x and VOC combined, ammonia, sulfur species, direct $PM_{2.5}$, and all emissions. Color scale indicates change in $PM_{2.5}$ level expressed in units $\mu g/m^3$. Negative values, colored green to blue, indicate a benefit in which $PM_{2.5}$ levels were reduced in response to reduced emissions levels. California state lines, county lines within the SFBA, and the major CV cities of Sacramento, Stockton, Modesto, Fresno, and Bakersfield (from north to south) are drawn using black.