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PM2.5 Data Analysis and Modeling in the San Francisco Bay Area

Final Report

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PM2.5 Data Analysis and Modeling in the San Francisco Bay Area

FINAL REPORT TO THE

Coordinating Research Council
Project # A-96

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Table of Contents

List of Figures

List of Tables

ABSTRACT

Routine ambient measurements of PM2.5 mass and chemical composition collected in the San Francisco Bay Area (SFBA) were analyzed for source contributions using the Chemical Mass Balance (CMB) model and the Positive Matrix Factorization (PMF) model. CMB source contributions appear plausible, with high contributions from biomass burning sources during winter months, and more consistent contributions from other sources throughout the year (slight attenuation due to seasonal changes in atmospheric mixing). Diesel engines, food cooking, gasoline engines, road dust, and brake wear are predicted to be the most significant sources of PM2.5 in addition to biomass burning. PMF source contributions suffered from lack of unique molecular markers and/or unique elements consistently above detection limits. Primary source contributions from biomass and road dust were resolved by PMF, but a single factor was predicted for diesel engines, gasoline engines, brake wear, and cooking. It is uncertain if this combined PMF factor can be deconvoluted given the available measurement data. At present, the CMB source contributions appear to provide a reasonable comparison point for predicted source contributions to PM2.5 produced by chemical transport model.

Wood smoke contributions predicted by UCD/CIT model with the emissions inventory prepared by the BAAQMD are significantly lower than contributions predicted using the emissions inventory prepared by CARB. CMB predictions for wood smoke contributions do not entirely agree with estimates produced by either emissions inventory, but results do suggest that the BAAQMD emissions inventory may slightly under predict wood smoke contributions at the monitoring sites considered. Gasoline contributions predicted by the UCD/CIT model using the BAAQMD 2012 inventory and the ARB 2010 inventory are 0.09 μg/m3 and 0.14 μg/m3 at Livermore, 0.15 μg/m3 and 0.37 μg/m3 at West Oakland, 0.12 μg/m3 and 0.14 μg/m3 at Vallejo, 0.16 μg/m3 and 0.33 μg/m3 at San Jose, and 0.02 μg/m3 and 0.01μg/m3 at Point Reyes, respectively. Diesel contributions predicted by the UCD/CIT model using the BAAQMD 2012 inventory and the ARB 2010 inventory are 0.16 μg/m3 and 0.42 μg/m3 at Livermore, 1.87 μg/m3 and 1.42 μg/m3 at West Oakland, 0.13 μg/m3 and 0.18 μg/m3 at Vallejo, 0.41 μg/m3 and 0.72 μg/m3 at San Jose, and 0.02 μg/m3 and 0.02μg/m3 at Point Reyes, respectively. This reflects the shift of heavy duty diesel emissions on some transportation corridors in the BAAQMD inventory and the daily variability in the CARB inventory. The predicted response of PM2.5 concentrations to emissions controls applied to different pollutants (NOx, VOC, SOx, NH3, PM) was similar for CMAQ and the UCD/CIT model. Changes to primary PM2.5 emissions were predicted to have the greatest effect on ambient PM2.5 concentrations in the Bay Area. PM2.5 improvement from reduction of gas pollutants such as NOx, VOC, NH3 and SOx was very limited in the Bay Area.

EXECUTIVE SUMMARY

The San Francisco Bay Area (SFBA) experiences exceedances of the 24-hour PM2.5 (particles with aerodynamic particle diameters below 2.5μm) National Ambient Air Quality Standard (NAAQS) during winter months. The objectives of this study are to conduct data analysis and modeling in the SFBA, compare results with analysis performed by the Bay Area Air Quality Management District (BAAQMD), and make recommendations on using modeling and analysis tools for air quality decision making. This is the Final Draft and the modeling results are in this report.

Routine PM2.5 mass and chemical species measurement data from 2012 were analyzed at 5 sampling locations in the SFBA: Livermore, West Oakland, San Jose, Vallejo, and Point Reyes. CMB modeling (EPA CMB8.2) and Positive Matrix Factorization (EPA PMF 5.0) modeling was carried out using standard methods. CMB source contributions appear plausible, with high contributions from biomass burning sources during winter months, and more consistent contributions from other sources throughout the year (slight attenuation due to seasonal changes in atmospheric mixing). Diesel engines, food cooking, gasoline engines, road dust, and brake wear are predicted to be the most significant sources of PM2.5 in addition to biomass burning. PMF source contributions suffered from lack of unique molecular markers and/or unique elements consistently above detection limits. Primary source contributions from biomass and road dust were resolved by PMF, but a single factor was predicted for diesel engines, gasoline engines, brake wear, and cooking. It is uncertain if this combined PMF factor can be deconvoluted given the available measurement data.

Predictions of $PM_{2.5}$ species (total mass, OC, EC, sulfate, nitrate) from the CMAQ model using the BAAQMD 2012 emissions inventory, the UCD/CIT model with ARB 2010 emission inventory and the UCD/CIT model with BAAQMD 2012 emission inventory are in general agreement. Predictions from all models have MFBs within ±0.5 and MFE less than 0.80 for PM2.5 total mass and PM2.5 OC, indicating good agreement between predictions and measurement. All models generally under predicted nitrate concentrations. The UCD/CIT model consistently overestimated sulfate concentration, while the CMAQ model slightly under-predicted high concentrations of sulfate and overestimated low concentrations of sulfate. Despite the limitations in predicting secondary species, the general agreement between the predicted and observed concentrations at 5 locations for $PM_{2.5}$ species builds confidence that the major chemical, physical and transport processes are reasonably represented by the model simulation so that the results are suitable for further $PM_{2.5}$ source contribution analysis and control strategies evaluations.

Source contributions predicted by the UCD/CIT air quality model with ARB 2010 inventory and BAAQMD 2012 inventory were compared to receptor-oriented source apportionment results produced by the Chemical Mass Balance (CMB) model at 5 sites in the Bay Area. Wood smoke

contributions predicted by UCD/CIT model with the BAAQMD 2012 inventory were significantly lower than wood smoke contributions predicted with the ARB 2010 inventory. Gasoline and diesel contributions from the UCD/CIT model with the BAAQMD 2012 inventory and with the ARB 2010 inventory differ by as much as ~70%. All of these trends can be explained by the modifications to the emissions inventory carried out by BAAQMD staff, and the current results provide feedback that will focus efforts for further work in this area.

The PM2.5 concentration response to emissions controls for different pollutants, i.e. NOx, VOC, SOx, PM, NH3 predicted by the UCD/CIT model generally matches the predicted response from the CMAQ model. Ambient PM2.5 concentrations in the Bay Area respond most strongly to controls on primary PM2.5 emissions, with more subtle effects from controls on gas phase emissions.

1. Introduction

The San Francisco Bay Area (SFBA) experiences exceedances of the 24-hour PM2.5 (particles with aerodynamic particle diameters below 2.5μm) National Ambient Air Quality Standard(NAAQS) during winter months. Even though the region began meeting both the 24-hour and annual average PM2.5 NAAQS in recent years, PM2.5 still significantly impacts public health in the region. Epidemiological and toxicological studies show that PM2.5 poses a threat to public health (see for example[1-7]).

To reduce ambient PM2.5 levels, the Bay Area Air Quality Management District (BAAQMD) continually evaluates existing emission reduction programs and adopts new ones based on information from on-going PM2.5 modeling and analysis.

The District has been collecting PM2.5 data from fifteen air monitoring stations-five of which include speciation (Table 1); developed a modeling emissions inventory; performed statistical cluster analyses of relationships among meteorology, emissions and air quality; performed chemical mass balance (source apportionment) analyses; applied MM5/CMAQ and WRF/CMAQ couples; conducted model sensitivity simulations; and estimated the health impacts of PM2.5.

Table 1 Bay Area PM2.5 air monitoring stations

	Site Coordinates			continuous	speciated	Design Values 2012-2014 (ug/m3)	
Site	Latitude	Longitude	In operation	PM2.5 anal	filter sampl	24-hour	annual mean
Concord	37.936	-122.026	*	*		22	7.0
Gilroy	36.9995	-121.575	*	*		18	7.6
Laney College	37.796	-122.263	*	*			
Livermore	37.6875	-121.784	*	*	*	27	7.5
Napa	38.311	-122.296	*	*			
Oakland	37.7431	-122.17	*	*		24	9.4
Oakland West	37.81478	-122.282	*	*	*		
Point Reyes	38.12306	-122.908	*		*		
Redwood City	37.4829	-122.203	*	*		23	8.8
San Francisco	37.766	-122.399	*	*		23	8.6
San Jose	37.3485	-122.895	*	*	*	30	10
San Pablo	37.9604	-122.357	*	*			
San Rafael	37.9723	-122.52	*	*		22	9.8
Santa Rosa	38.4435	-122.71		*			
Sebastopol	38.40377	-122.818	*	*			
Vallejo	38.1027	-122.238	*	*	*	26	9.6
	Laney College and Sebastopol started operation in 2014						
Note	Santa Rosa ceased operation in 2014						
	Napa, Oakland West, San Pablo and Point Reyes had insufficient data t to estimate design values						

The objectives of this study are to conduct data analysis and modeling in SFBA, compare data analysis and modeling results with District's, and make recommendations on using modeling and analysis tools for air quality decision making. The specific tasks used to achieve these objectives were:

1) Obtain meteorological model outputs and emissions inventory for 2012 from the District.

2) Adopt a modeling domain for the UCD/CIT model and conduct the same set of base case and sensitivity simulations as the District.

3) Evaluate the UCD/CIT model. Compare the model's performance and response to changes in emissions to those of CMAQ.

4) Conduct CMB and PMF analysis for PM source apportionment.

5) Corroborate primary and secondary PM2.5 data analysis and CMAQ results using UCD/CIT source apportionment; identify contribution of mobile sources to PM2.5.

6) Evaluate overall BAAQMD modeling and analysis of speciated PM2.5 data to determine model and emissions fidelity. Recommend improvements if any.

2. Methods

2.1 Ambient PM2.5 measurements

In this study, ambient data were used from the four District monitoring sites where requisite data were available, Livermore, West Oakland, San Jose, and Vallejo, and from a non-District site, Point Reyes. Point Reyes is part of the IMPROVE network that operates in National Parks. These sampling locations span the variability in PM2.5 composition throughout the Bay Area. The West Oakland, Vallejo, Livermore and San Jose sites are located in different sub-regions of the highly urbanized Bay Area and are four urban sites. Point Reyes is a remote coastal location, presumably sampling near-background pollution levels. Time-integrated 24-hour PM2.5 samples were collected once every six days from 1/1/2012 to 12/29/2012 in Livermore, West Oakland and Vallejo and once every three days San Jose and Point Reyes. For each site, the set of chemical species measured included higher atomic weight elements; various ions including nitrate, sulfate, ammonium, chloride, sodium, and potassium; and elemental and organic carbon (EC and OC). Species chosen for CMB and PMF analysis were limited to those with a substantial fraction of concentrations greater than the estimated measurement uncertainty (that is, the standard deviation of the measurement error).

2.2 CMB and PMF model analysis

CMB modeling estimates source contributions to ambient PM concentrations. The CMB model searches for a fit that explains measured concentrations for a range of chemical components using a liner combination of measured source profiles. For each ambient sample, the CMB model finds the mix of chemical component sources profiles that best combine to match the

measured concentrations. In this study, CMB modeling (EPA CMB8.2) was performed on the PM2.5 samples collected during the year 2012.The profiles used in the CMB model were obtained from the EPA Speciate [8] databases, and BAAQMD databases that had been used in a previous study [9]. The source profiles utilized in the current study included new and aged marine air, geological dust, residential woodsmoke, wildfire smoke, meat cooking, diesel emissions, gasoline emissions, tire/brake wear, and fireworks. Ammonium nitrate and ammonium sulfate were also included.

Positive Matrix Factorization (EPA PMF 5.0) also was used to analyze PM2.5 data at the five measurement sites. PMF assumes that concentrations at receptor sites are the sum of a linear combinations of source emissions. Source emissions factors are derived by the model rather than provided as model inputs. In this work, measurement uncertainties were used for the error estimates of the measured values; missing data were replaced by the annual geometric mean of the corresponding species and four times the annual geometric mean was taken as the error estimate.

2.3 CTM Model Description

The air quality model employed in the current study is the Eulerian source-oriented University of California, Davis/California Institute of Technology (UCD/CIT) chemical transport model [10-26]. The details of the standard algorithms used in the UCD/CIT models are provided in previous studies and therefore only a summary is presented here. The formulation of advection and diffusion scheme is described by Kleeman and Cass [17], the dry deposition scheme is described by Kleeman et al. [19], the vertical advection scheme is described by Hu et al. [16], the wet deposition scheme is described by Mahmud et al [20]. Concentration fields from model

calculation in the coarse-resolution parent domain for every source type are saved in the boundary grid cells of the finer-resolution nested domains, and then are used as the boundary conditions for the model calculation in the nested domains. The UCD/CIT model was configured with 16 vertical layers up to a height of 5 km above ground level in all the mother and nested domains, with 10 layers in the first 1 km. Note that the use of relatively shallow vertical domains is only appropriate in regions with well-defined air basins and would not be appropriate for locations in the eastern USA or other regions with moderate topography. Particulate composition, number and mass concentrations are represented in 15 size bins, ranging from 0.01 to 10 μm in diameter. Primary particles are assumed to be internally mixed, i.e., all particles within a size bin have the same composition. Previous studies [24] have shown that this assumptions provides adequate predictions for total PM concentrations relative to source-oriented mixing treatments in California when feedbacks to meteorology are not considered [27].

2.4.1 Meteorological Fields

The Meteorological fields used in this study were provided by the Bay Area Air Quality Management District. Hourly meteorological fields were simulated with the Weather Research and Forecasting (WRF) model version 3.4. The modeling domain uses a nested grid setting with 36, 12 and 4 km horizontal resolutions. All three grids employ 50 vertical layers. WRF fields have been reformatted as inputs to the UCD/CIT air quality model using established preprocessing software at UC Davis.

2.4.2 Emissions

Two different emission inventories for the San Francisco Bay Area were used in this study as a sensitivity analysis. The first set of emissions was developed by the BAAQMD for the year 2012 based on the regulatory inventory provided by the California Air Resources Board for that same year. The BAAQMD updated the wood smoke emissions based on wintertime surveys for the past 10+ years to determine trends in wood burning activities and their characteristics. The recent surveys yielded information on the amounts of fuel (wood) burned by zip code, among other metrics. BAAQMD staff estimated annual total emissions based upon this information and developed spatial allocation surrogates from this information in combination with detailed Census data on home heating fuel and number of households. Staff further adjusted the surrogate for San Francisco County to account for new multi-family construction since the Census was taken. The end results of these modifications were that emissions were greatly reduced compared to previous estimates and emissions were moved away from densely populated urban core areas. BAAQMD staff also modified the mobile emissions inventory to exclude heavy duty trucks from I-580 from Foothill Blvd. in San Leandro to Grand Ave. in Oakland per the restrictions on these vehicles, and modified the spatial allocation of shipping emissions near the Port of San Francisco. The inventory was provided in an IDA format and processed using the SMOKEv3.7 software package provided by US EPA. SMOKE was configured to separately tag emissions from on-road gasoline vehicles, off-road gasoline vehicles, on-road diesel vehicles, off-road diesel vehicles, food cooking, biomass burning, natural gas, and all other sources. Mobile and non-road source sectors were treated as area sources for simplicity. VOCs within each of these source types were specified based on the native VOC profiles in the SMOKE system for the SAPRC99/SAPRC07/SAPRC11 chemical mechanism. PM profiles for each source type were

specified as weighted averages from each of the detailed sources within each broad category as summarized in Table 2 below.

Table 2 PM source profile averaging used with BAAQMD emissions

Source Type	PM Source Profile
On-road gasoline vehicles	10% Non-catalyst vehicle + 90% Catalyst Vehicle
Off-road gasoline vehicles	100% Non-catalyst vehicle
On-road diesel vehicles	100% On-road diesel vehicle
Off-road diesel vehicles	90% 1970's diesel vehicle + 7% 1980's diesel vehicle + 3% on-road diesel vehicle
Food Cooking	85% meat frying + 15% charbroiling
Biomass burning	95% residential wood smoke + 5% waste burning
Natural gas combustion	100% natural gas combustion
Other	70% construction & demolition + 10% paved road travel + 4% farming ops + 3% brake wear + 2% cattle feedlot + 2% mining ops + 1% process heaters + 1% cement manufacturing + 3% wood processing + 1% solid waste disposal + 2% mineral processing + 1% asphalt production + 1% organic solvent

The second emissions inventory was provided by the California Air Resources Board (CARB) for the year 2010 in support of the CalNex field project. The inventory was provided in MEDS format that was processed with individual EIC codes assigned to appropriate source profiles. VOC source profiles were obtained from CARB for the SAPRC chemical mechanism, while PM

source profiles were based on the library maintained at UC Davis. Biogenic emissions for all simulations were calculated using the latest version of the MEGAN model.

3. Results

3.1 Source apportionment of $PM_{2.5}$ using PMF model

To identify the potential sources of $PM_{2.5}$ in the Bay Area, the chemical composition and its uncertainty were used as input to the EPA PMF5.0 model. The data set included 24 variables (PM2.5 total mass, Cl^-, $NO3^-$, $SO4^=$, $NH4^+$, Na+, K^+, OC1, OC2, OC3, OC4, OPTR, OC, EC1, EC2, EC, TC, Si, S, Cl, K, Ca, Fe, Cu) at Livermore, West Oakland, Vallejo sites and 18 variables (missing OC and EC fractions) at San Jose and Point Reyes. All these variable have nomenclature Signal-to-Noise Ratio (S/N ratio) larger than 0.4. When the S/N ratio of the variable is larger than 1.0, we categorize it as “Strong”, when the S/N ratio is smaller than 1.0 and larger than 0.4, we categorize is as "Weak". The PM2.5 total mass is categorized as "Weak" since it should not have a large influence on the solution. Random seed number was chose for every site and the results for the run with the lowest Objective Function (Q) (robust) are chosen. Five different random seed values were tested and similar results were obtained. The seed value was eventually set at 89, 76, 87, 74, 86 at Livermore, West Oakland, Vallejo, San Jose and Point Reyes, respectively. Bootstrapping on the base solution reported stable results, with > 80 out of 100 bootstrap factors mapped with those in the base run for all the sites. In this study, PMF was able to resolve seven factors for the four urban sites (Livermore, West Oakland, Vallejo and San Jose) and four factors for the remote rural site (Point Reyes). PMF factors were identified as

mobile, ammonium sulfate (AmSul), ammonium nitrate (AmNit), biomass burning (Bioburning), Transport/Background, road dust (Roaddust) and marine.

Figure 1 shows annual averaged source contributions (%) to PM2.5 mass at the 5 measurement sites. Mobile and wood burning are the two major primary sources at urban sites. The former contributes 7-23% and the latter contributes 16-20% . The contribution from transport/background also plays an important role at both urban and rural sites. This factor mainly contains sulfate, nitrate, OC and aged sea salt. It may reflect the atmospheric transporting, mixing and condensation of oxidized compounds. PMF results represent aged sea salt mixing with ship emission or other secondary pollutants transported from outside California. Other results vary between sites. Road dust contributes 2-7% and marine contributes 7-13%. At the Point Reyes background site, marine dominates, representing 51% of total PM2.5. Nitrate and sulfate are major secondary source factors at both urban and rural sites.

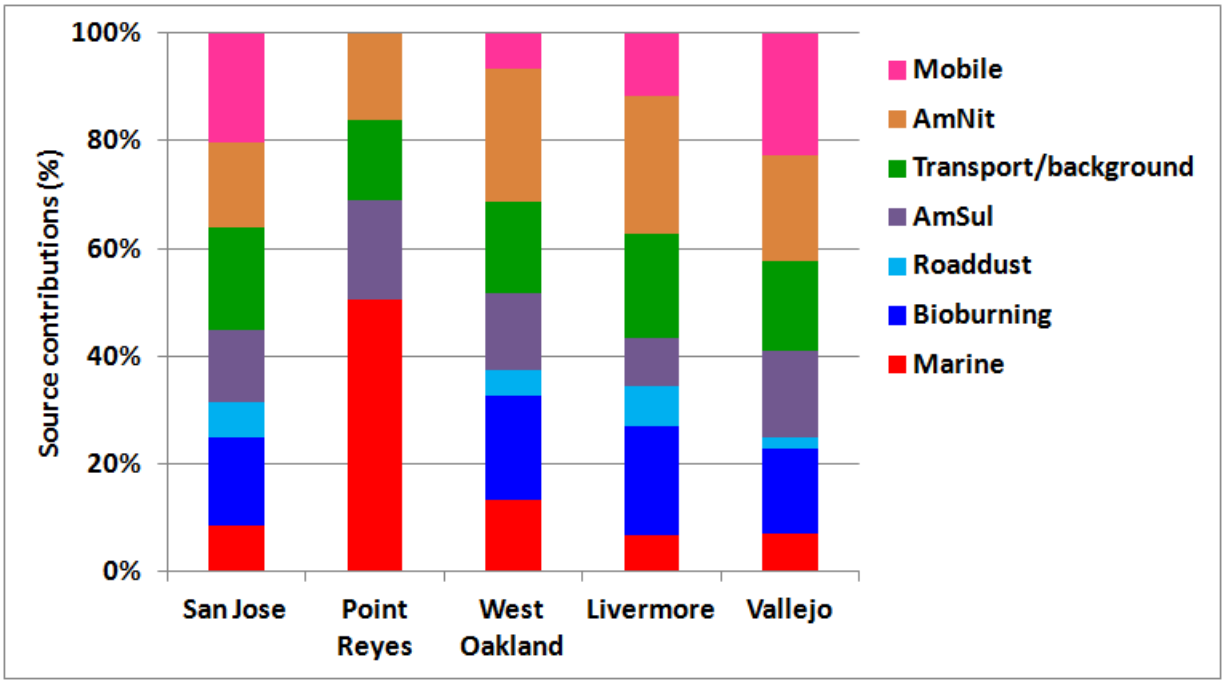


Figure 1. Annual averaged source contributions (%) to PM2.5 mass calculated using PMF, by site.

3.2 Source apportionment of PM2.5 using CMB model

Figure 2 shows annual average source contributions from 9 categories to PM2.5 concentrations at the 5 sampling sites. Table 3 shows the percentages for each site and the averaged percentages at the four urban sites. In general, the major categories among 4 urban sites are direct, combustion-related, largely carbonaceous sources – Biomass burning and fossil fuel (diesel and gasoline); and secondary, combustion-related sources – ammonium nitrate and ammonium sulfate. Biomass burning and ammonium nitrate are the two largest contributors at every urban site, averaging 25% and 23% of the total, respectively; marine air dominates at the Point Reyes background site, representing 48% of its total. Among all the anthropogenic sources, ammonium sulfate is the third greatest urban source category, representing 13% of the total. Diesel is next at 14%, followed by cooking (10%) and Gasoline (4%). Road dust contributions averaged 2%, and brake contributions averaged 2% at urban sites.

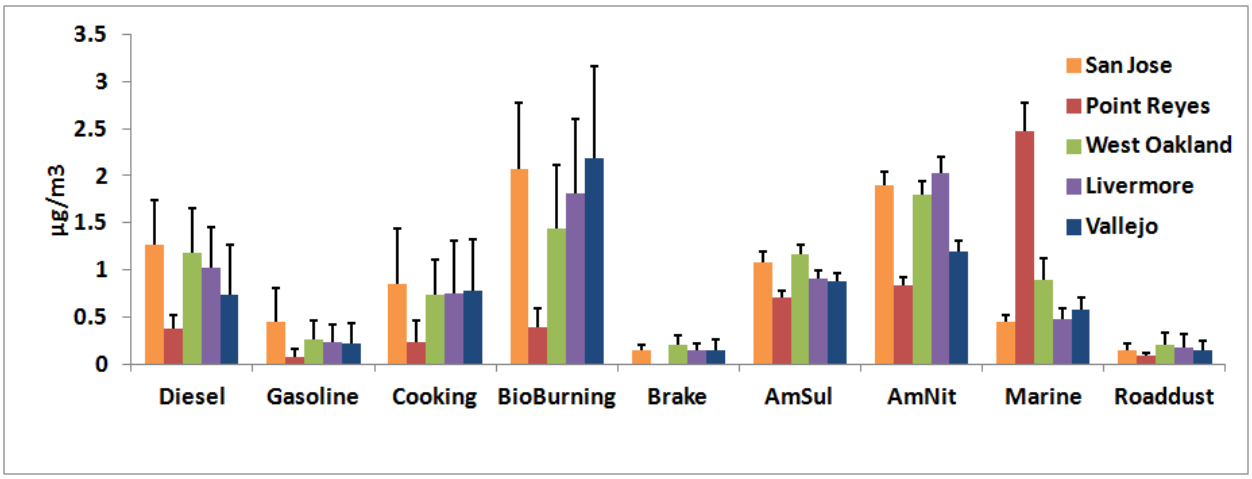


Figure 2. Annual averages of concentrations for 9 source categories estimated using CMB.

Table 3 Annual source category contribution percentages, by site.

	Diesel	Gasoline	Cooking	BioBurning	Brake	AmSul	AmNit	Marine	Roaddust
San Jose	15%	5%	10%	25%	2%	13%	23%	5%	2%
West Oakland	15%	3%	9%	18%	3%	15%	23%	11%	3%
Livermore	14%	3%	10%	24%	2%	12%	27%	6%	2%
Vallejo	11%	3%	11%	32%	2%	13%	17%	8%	2%
4 Site Ave	14%	4%	10%	25%	2%	13%	23%	8%	2%
Point Reyes	7%	1%	5%	8%	0%	14%	16%	48%	2%

Figures 3-7 show CMB results by season for each site. PM2.5 concentrations are highest in November-Feb. We defined this period as the winter season. Other seasons are defined as follows: spring as March-April, summer as May-August, fall as September-October. At every site, ammonium nitrate and biomass burning are much higher in the winter season due to the low temperature which increases emissions of NOx associated with heating and encourages the partitioning of nitrate to the particle phase. Diesel and gasoline are also highest in winter due in part to greater atmospheric stability in the winter. Marine is lowest in winter, peaking in summer. This may be related to the seasonal wind pattern in the Bay Area. Easterly winds are common in

winter months, with westerly wind carrying marine air from the Pacific Ocean more typical during the rest of the year. Ammonium sulfate and cooking are less variable from season to season.

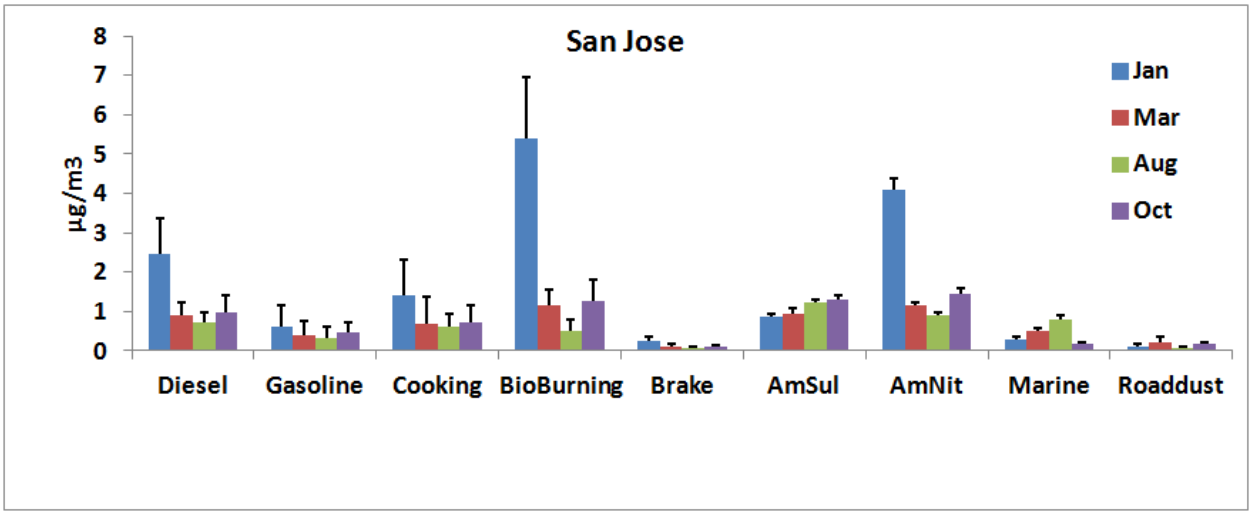


Figure 3 PM2.5 source contributions by season, San Jose, 2012.

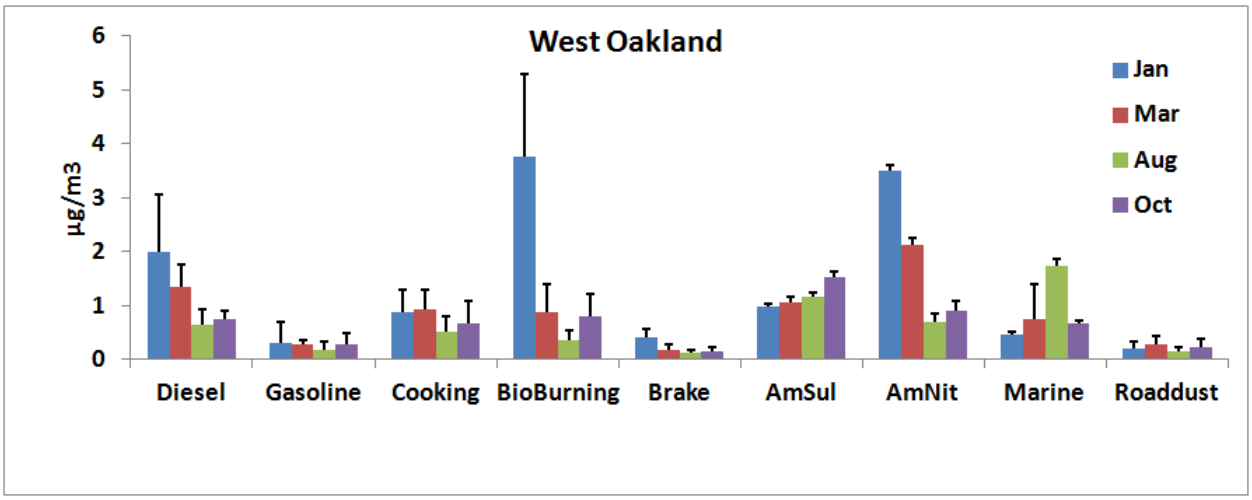


Figure 4. PM2.5 source contributions by season, West Oakland, 2012.

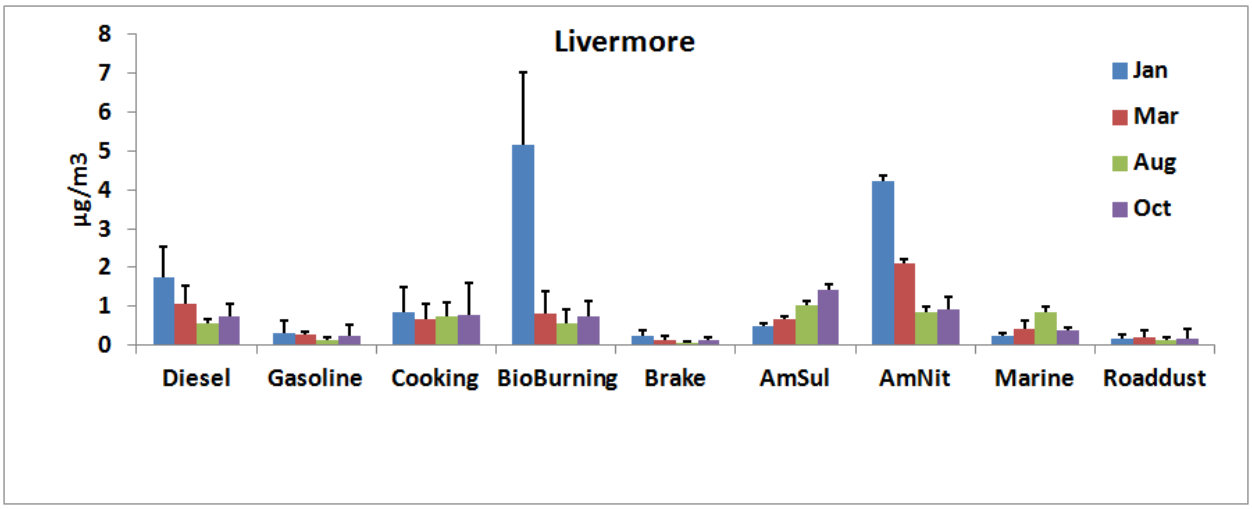


Figure 5. PM2.5 source contributions by season, Livermore, 2012.

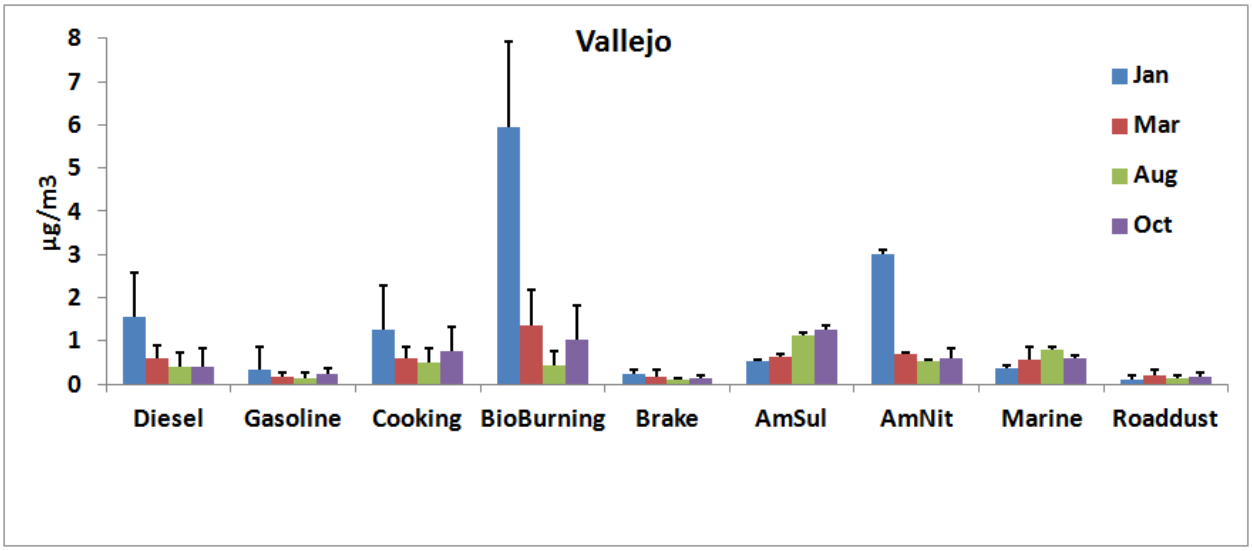


Figure 6. PM2.5 source contributions by season, Vallejo, 2012.

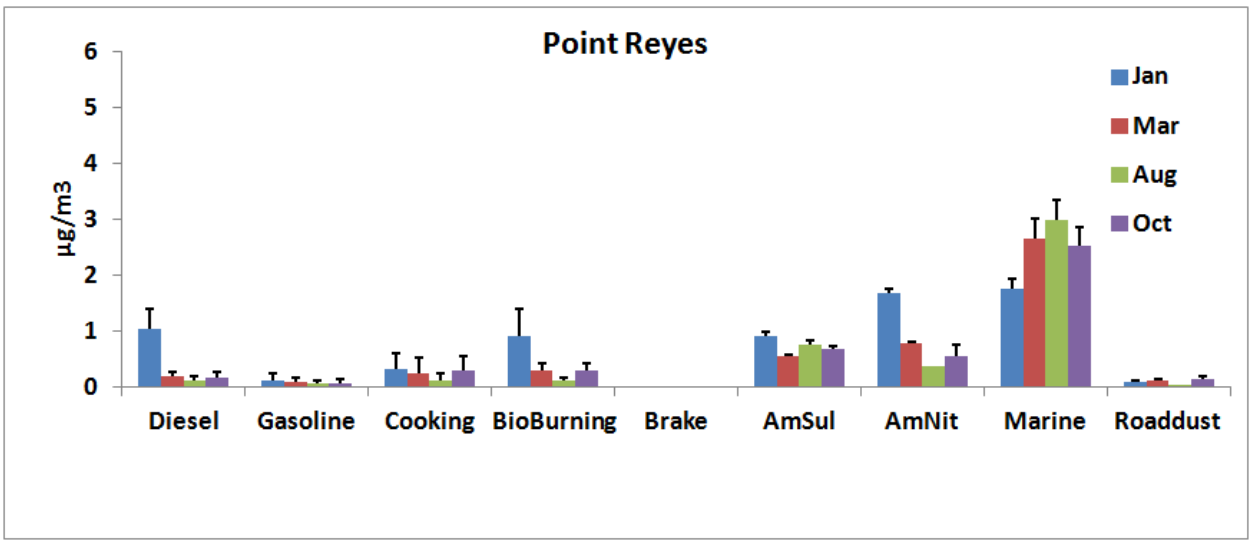


Figure 7. PM2.5 source contributions by season, Point Reyes, 2012.

3.3 Comparison of the PMF and CMB results

Figure 8 shows annual averaged source contributions (%) to PM2.5 mass estimated by PMF and CMB at each sampling site. Mobile source contributions are not split into diesel and gasoline in PMF model due to lack of unique element signatures for these different sources in the measurement dataset. The Transport /Background factor in the PMF results and the unresolved residual in CMB results are also excluded from the comparison in Figure 8 since they are not present in both sets of model results.

Both CMB and PMF identify ammonium sulfate and ammonium nitrate as major secondary sources at both urban sites and rural sites. Good agreement is found between CMB and PMF model results for the Marine source. The differences in estimated contributions from mobile sources and other sources (mostly primary) may be attributed to several issues. Resolved source profiles "as determined" in PMF have more processed (or aged) characteristics resulting in part

from atmospheric mixing and condensation of oxidized compounds, whereas source profiles used "as input" in CMB are obtained from measurements of emission sources with minimum atmospheric processing. The omission of possible known or unknown sources due to lack of specific measurements for unique elements or molecular ''marker'' species may also cause the differences between CMB and PMF estimates.

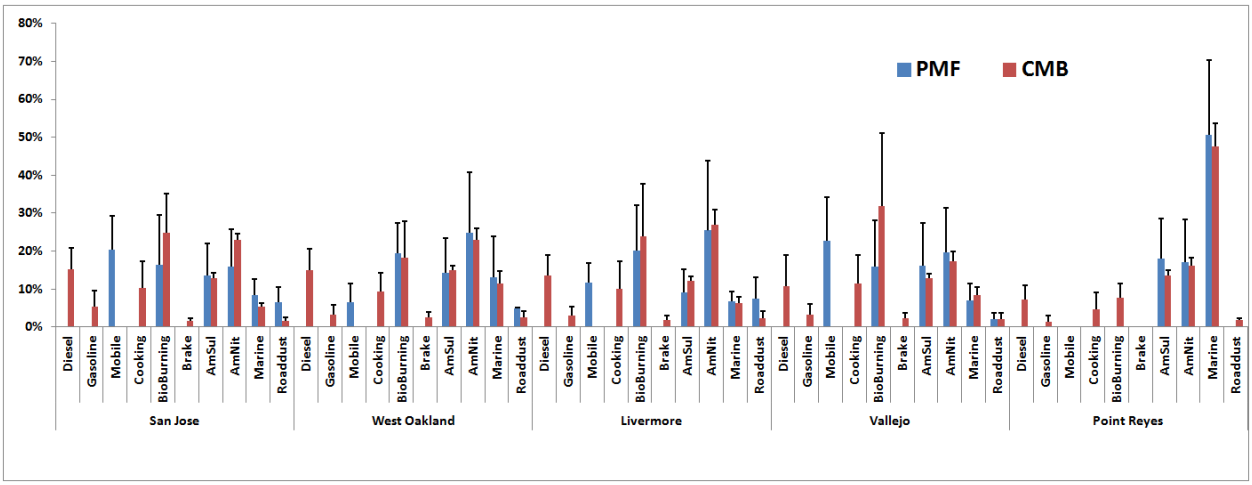


Figure 8. Annual averaged source contributions (%) to PM2.5 mass estimated by PMF and CMB at different sites in the Bay Area.

3.4 Performance evaluation of CMAQ and UCD/CIT model

In this study, 6 model periods in 2012 corresponding to periods simulated by BAAQMD were selected for further analysis: Jan 1-31 (winter stagnation); Mar 1-15 (spring convection); May 1-15 (early summer); August 1-15 (mid summer); Oct 1-15 (fall convection); December 1-31 (winter stagnation). Statistical measures of mean fractional bias (MFB) and mean fractional error (MFE) were calculated to evaluate the accuracy of model predictions in this study.

Concentration-dependent MFB and MFE performance goals and criteria were proposed by Boylan and Russell [28] to account for the fact that lower concentrations are more difficult to accurately predict. The performance goals are the level of accuracy close to the best that a model can be expected to achieve, while performance criteria are the level of accuracy acceptable for standard modeling applications.

Figure 9 shows the MFB and MFE values of particulate species of $PM_{2.5}$ total mass, EC, OC, nitrate, and sulfate using daily averages across all measurement sites during the entire modeled period. In this report, the 'CMAQ' case refers to results generated by BAAQMD staff using the CMAQ model; the 'BA2012' case refers to results from the UCD/CIT model based on the BAAQMD 2012 emission inventory; and the 'ARB2012' case refers to results also from UCD/CIT model but based on ARB 2010 emission inventory. All cases use 2012 meteorology. The difference between the CMAQ case and the BA2012 case represents the impact from using different air quality models. The difference between the BA2012 case and the ARB2012 case quantifies the change caused by different emission years and different version of the emission inventory.

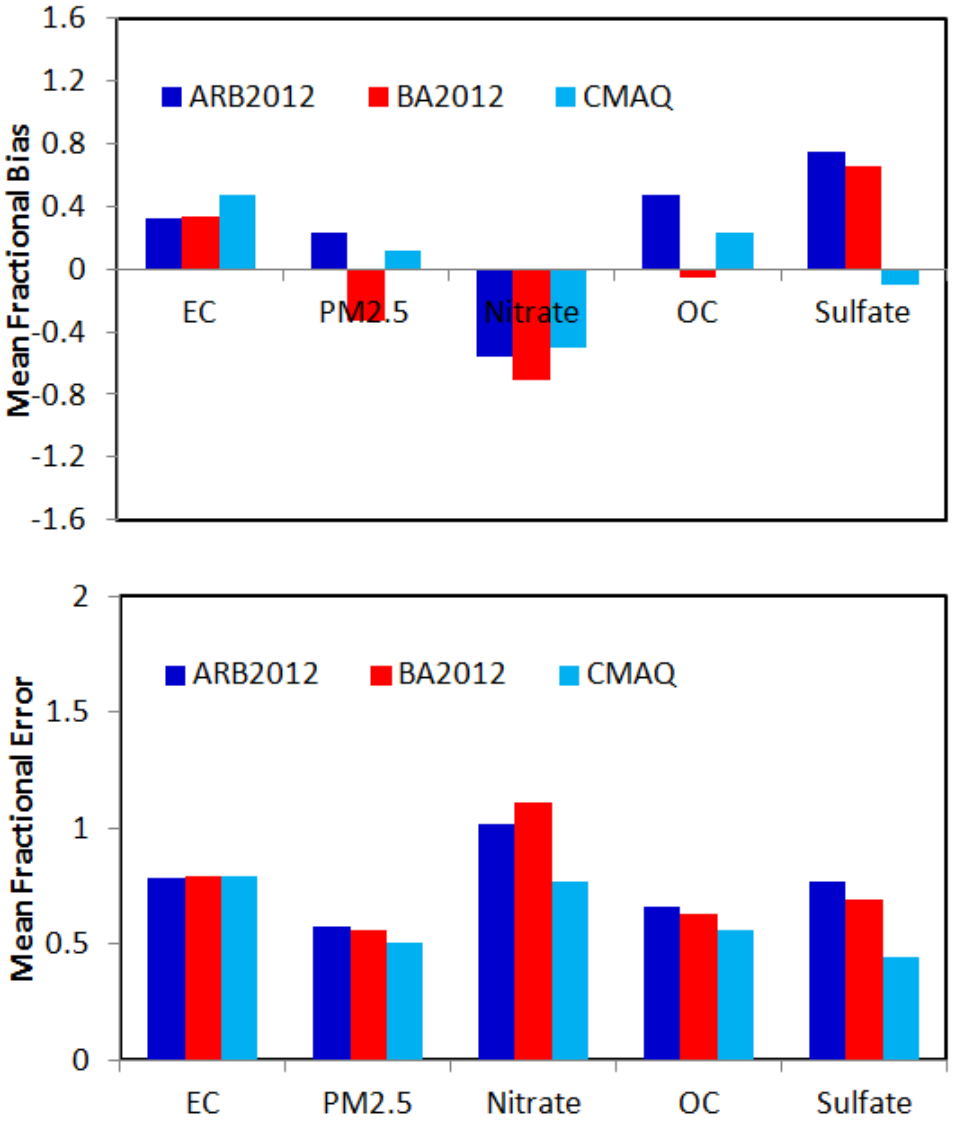


Figure 9 Mean fractional bias and mean fractional errors of PM species calculated using daily averages.

EC and PM2.5 have MFBs within ±0.4 and MFE less than 0.80 for all three cases, indicating general agreement between predictions and measurement. Nitrate has MFBs of -0.56, -0.50, -0.71, with MFEs of 1.01, 0.76, 1.11 for ARB2012, BA2012 and CMAQ case, respectively. The large negative bias combined with large error indicates that the daily nitrate concentrations are

consistently underestimated for both the CMAQ model and the UCD/CIT model using both emissions inventories. OC has MFB of 0.48 0.23,-0.05 with MFE of 0.66, 0.56 and 0.62 for ARB2012, CMAQ and BA2012 cases, respectively. These statistics indicate acceptable agreement between predicted OC and observed OC for the two cases that employed the BAAQMD emissions inventory. All simulations used identical secondary organic aerosol (SOA) models and treated primary organic aerosol (POA) as non-volatile. POA dominates the total predicted organic aerosol in the Bay Area during the current simulations, but SOA makes increasingly larger contributions to total organic aerosol at locations farther inland (Figure 10). Sulfate has MFB of 0.75 and 0.65 with MFE of 0.77 and 0.69 for ARB2012 and BA2012 cases indicating that the UCD/CIT model consistently overestimated sulfate concentration. Sulfate has MFB of -0.09 and MFE of 0.44 in the CMAQ case indicating that high concentration sulfate is slightly under-predicted and low concentration sulfate is slightly overestimated. Both the UCD/CIT model and the CMAQ model obtained boundary conditions for sulfate from MOZART simulations and both models had similar shipping emissions. The UCD/CIT model includes an online sea salt emissions module that includes sea salt sulfate which may produce higher predicted sulfate concentrations.

(a) BAAQMD2012 Emissions Case

(b) ARB2012 Emissions Case

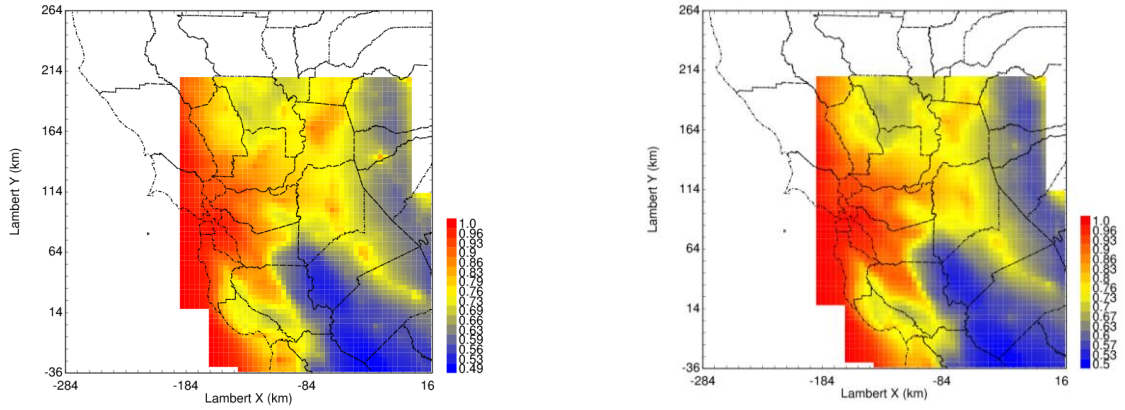
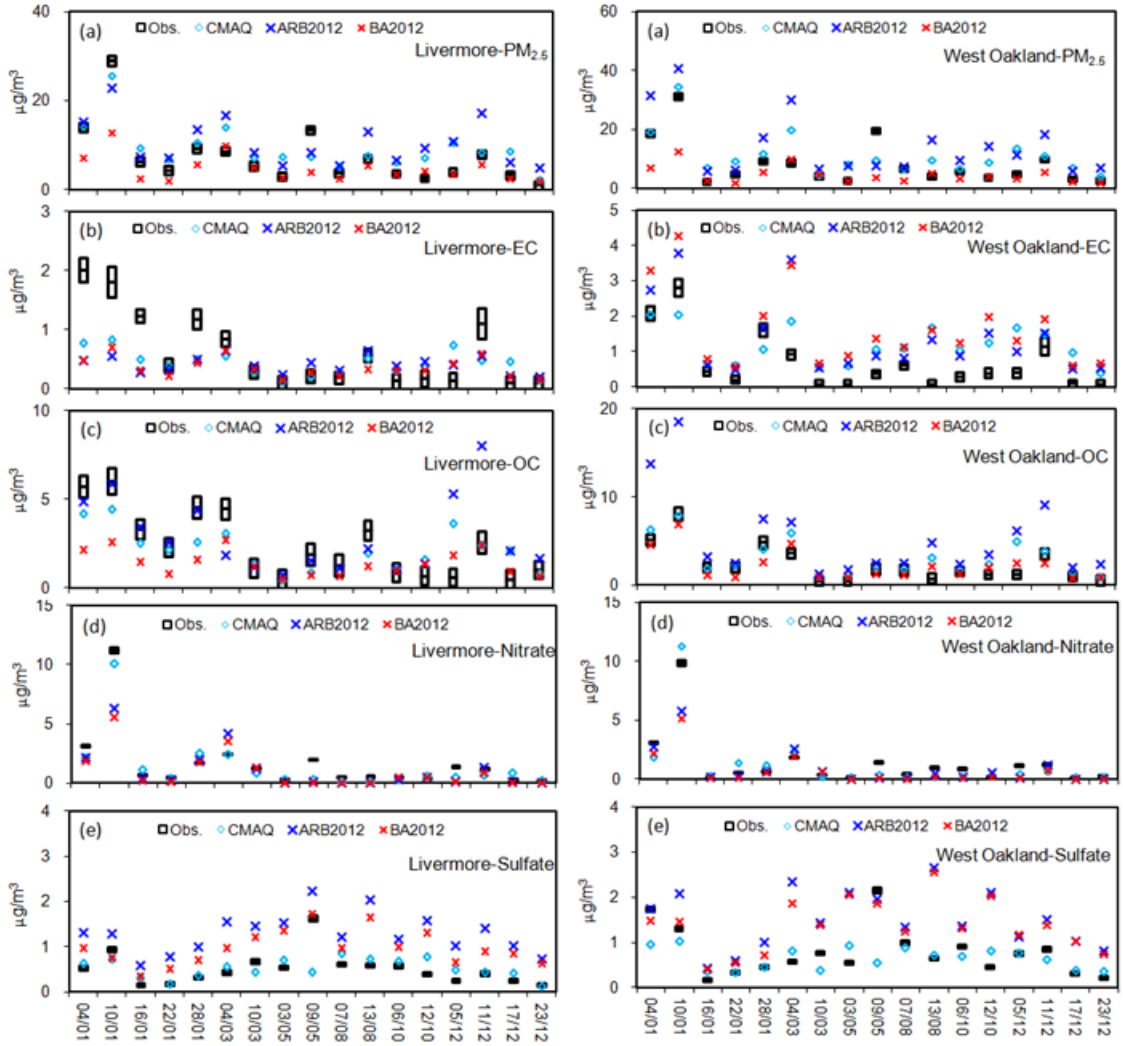
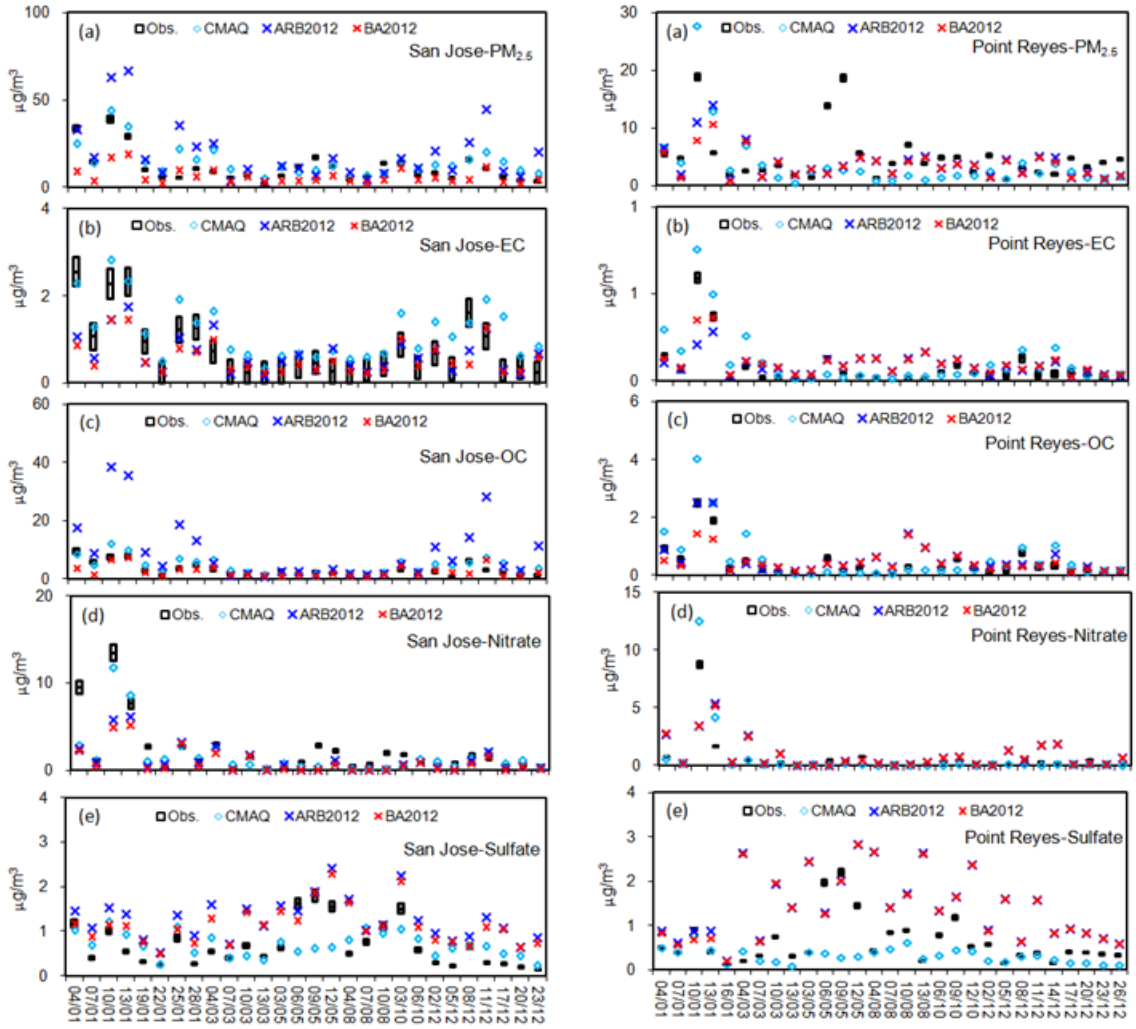


Figure 10: Ratio of primary organic aerosol (POA) to total organic aerosol mass using (a) the BA2012 emissions case and (b) the ARB2012 emissions case averaged over all 2012 simulation days. Organic matter in marine aerosol is counted as POA.

Figures 11 shows the predicted and measured daily average $PM_{2.5}$ total mass and major PM2.5 species EC, OC, sulfate and nitrate at Livermore, Oakland, San Jose, Point Reyes and Vallejo. The results in Fig 11 illustrate spatial and temporal variations of predicted and measured PM2.5 species. At four urban sites, Livermore, Oakland, Vallejo and San Jose, seasonal trends in observation of $PM_{2.5}$ species (PM2.5 total mass, OC, EC, nitrate) are generally captured with high concentrations in the winter and low concentrations in the summer, but model results do not exactly match measured values, especially for nitrate. At the upwind Point Reyes site, both simulated and observed concentrations of PM2.5 OC, EC and nitrate are higher during the winter season and relatively lower in the summer, but total PM2.5 mass does not follow this trend. This discrepancy may be due to sea salt which is a major component of PM2.5 at Point Reyes site.


(a) Obs. CMAQ ARB2012 BA2012 Livermore-PM2.5
(b) Livermore-EC
(c) Livermore-OC
(d) Livermore-Nitrate
(e) Livermore-Sulfate
(a) West Oakland-PM2.5
(b) West Oakland-EC
(c) West Oakland-OC
(d) West Oakland-Nitrate
(e) West Oakland-Sulfate
μg/m3
04/01 10/01 16/01 22/01 28/01 04/03 10/03 03/05 09/05 07/08 13/08 06/10 12/10 05/12 11/12 17/12 23/12
04/01 10/01 16/01 22/01 28/01 04/03 10/03 03/05 09/05 07/08 13/08 06/10 12/10 05/12 11/12 17/12 23/12


(a) Obs. CMAQ ARB2012 BA2012 San Jose-PM2.5
(b) Obs. CMAQ ARB2012 BA2012 San Jose-EC
(c) Obs. CMAQ ARB2012 BA2012 San Jose-OC
(d) Obs. CMAQ ARB2012 BA2012 San Jose-Nitrate
(e) Obs. CMAQ ARB2012 BA2012 San Jose-Sulfate
(a) Obs. CMAQ ARB2012 BA2012 Point Reyes-PM2.5
(b) Obs. CMAQ ARB2012 BA2012 Point Reyes-EC
(c) Obs. CMAQ ARB2012 BA2012 Point Reyes-OC
(d) Obs. CMAQ ARB2012 BA2012 Point Reyes-Nitrate
(e) Obs. CMAQ ARB2012 BA2012 Point Reyes-Sulfate
μg/m3

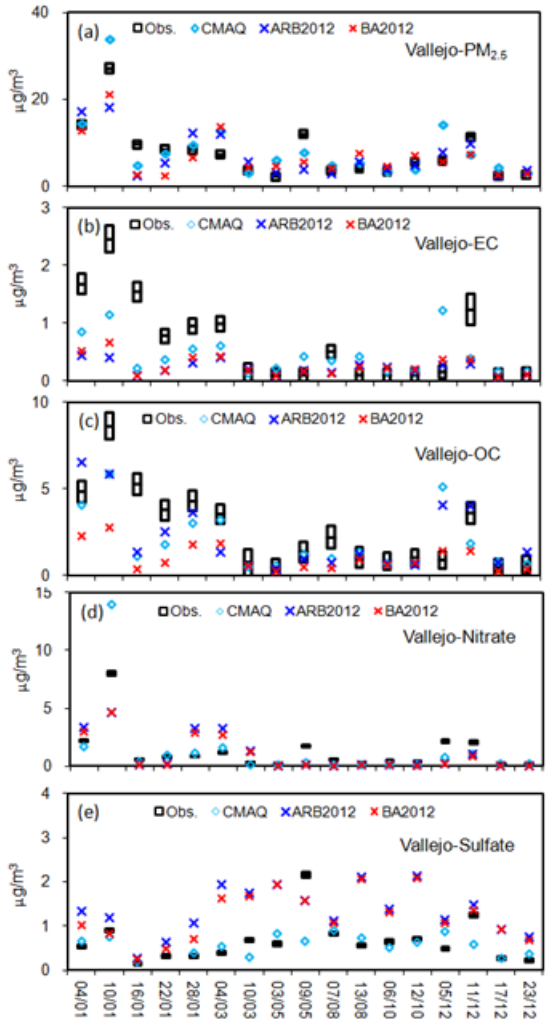


Figure 11 Predicted vs. observed daily average (a) $PM_{2.5}$ total mass, (b) $PM_{2.5}$ EC,(c) OC , (d) Nitrate, and (e) Sulfate at Livermore, West Oakland, San Jose, Point Reyes and Vallejo in Bay Area. Dark box represents observation and uncertainty; light blue diamond represents predicted results from CMAQ model; dark blue cross represents predicted results from UCD/CIT model based on ARB2010 emission inventory; and red cross represents predicted results from UCD/CIT model based on BAAQMD 2012 emission inventory.

3.5 $PM_{2.5}$ source appointment results estimated by UCT/CIT model

Figure 12 compares the average source contributions to primary PM2.5 mass predicted by the CMB receptor model, the UCD/CIT model with ARB 2010 emission (ARB2012), the UCT/CIT

model with BAAQMD 2012 emission and CMAQ model with BAAQMD 2012 emission at four urban site (Livermore, West Oakland, Vallejo and San Jose) and a rural site (Point Reyes) in the Bay Area of California during the 2012 simulation period. Dust sources were excluded from the relative source contribution calculation due to previously identified problems with the raw fugitive dust emission inventory [29]. The source contribution predictions from the UCD/CIT model with ARB 2010 emission (ARB2012) for wood burning, diesel, gasoline vehicles and cooking, generally fall into the uncertainty range of the CMB model at the Livermore site. The ARB2012 case predicts higher contribution from food cooking (1.85 μg m^{-3}) than the CMB model (0.74±0.46 μg m^{-3}) at the Oakland site, and much higher contribution from wood smoke (8.13 μg m^{-3}) than CMB model (3.97±1.59 μg m^{-3}) at the San Jose site. The predictions based on the ARB2012 case are in excellent agreement with predictions from the CMB model for wood smoke at the Oakland site and cooking at the San Jose site. The ARB2012 case predicts much lower contributions than the CMB model from all the sources at Vallejo and Point Reyes. The UCT/CIT model with BAAQMD 2012 emissions (BA2012) generally predicts much lower contributions from wood smoke sources than the CMB model at all the monitoring sites. The BA2012 case also predicts lower contribution from diesel, gasoline and cooking than the CMB model at Vallejo and Point Reyes. However, the predictions from BA2012 for diesel contributions are higher than CMB model at Oakland. In general, the source contribution predictions from the UCD/CIT model with ARB 2010 emission inventory are higher than predictions from the UCD/CIT model with BAAQMD 2012 emission, especially for wood smoke.

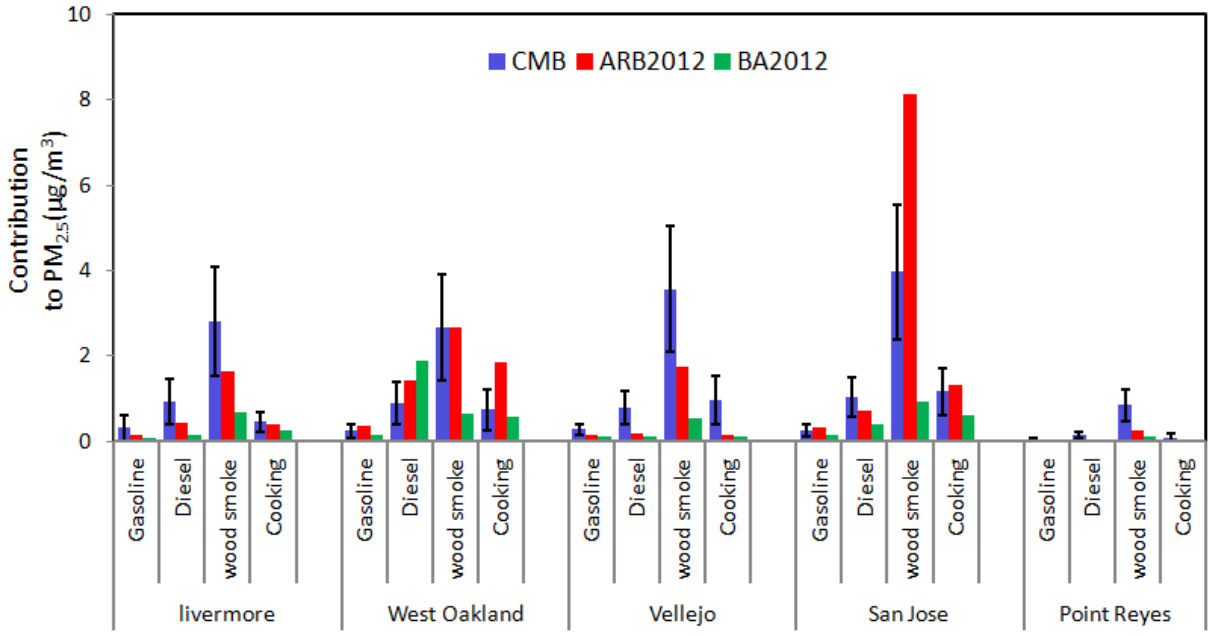


Figure 12 Averaged source contributions to primary PM2.5 concentrations calculated by CMB and UCD/CIT model based on ARB emission inventory and BAAQMD emission inventory, respectively.

The daily averaged source contributions during the 2012 simulation period predicted by the CMB model, the UCD/CIT model with ARB 2010 emissions and the UCD/CIT model with BAAQMD 2012 emissions at Livermore, Oakland, Vallejo, San Jose and Point Reyes sites are shown in Figure 13-17, respectively. At every site, UCD/CIT predictions for wood smoke contributions are much higher during the winter season than during the summer season which matches trends predicted by the CMB model. During the winter season, the predictions of wood smoke contributions from the UCD/CIT model with BAAQMD 2012 emissions are lower than CMB predictions at the Livermore, Vallejo and Point Reyes sites. The predictions of wood smoke contributions from the UCD/CIT model with ARB2012 emissions agree well with CMB model predictions at Livermore, Oakland, Vallejo and Point Reyes during most of the simulation

days, but are higher than CMB model predictions at the San Jose site. During seasons other than winter, the UCD/CIT predictions for wood smoke contributions using both ARB2012 and BA2012 emissions are much lower than CMB model predictions for wood smoke. The predictions of gasoline and diesel contribution from ARB2012 and BA2012 are also higher during the winter season and lower during the summer season. Predictions for cooking contributions to PM2.5 do not follow a clear seasonal pattern, with variability apparent throughout the year at many of the monitoring sites.

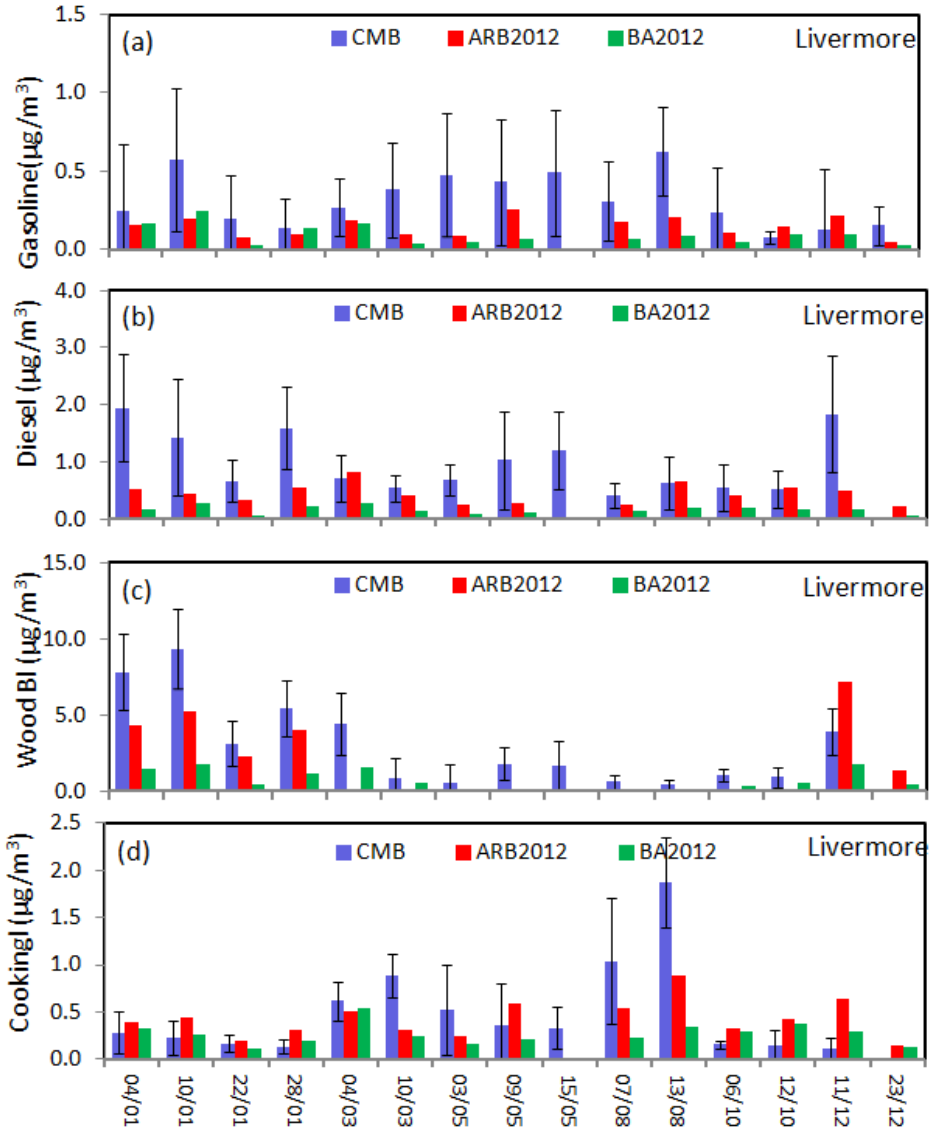


Figure 13 Source contributions to primary PM2.5 mass concentrations in Livermore

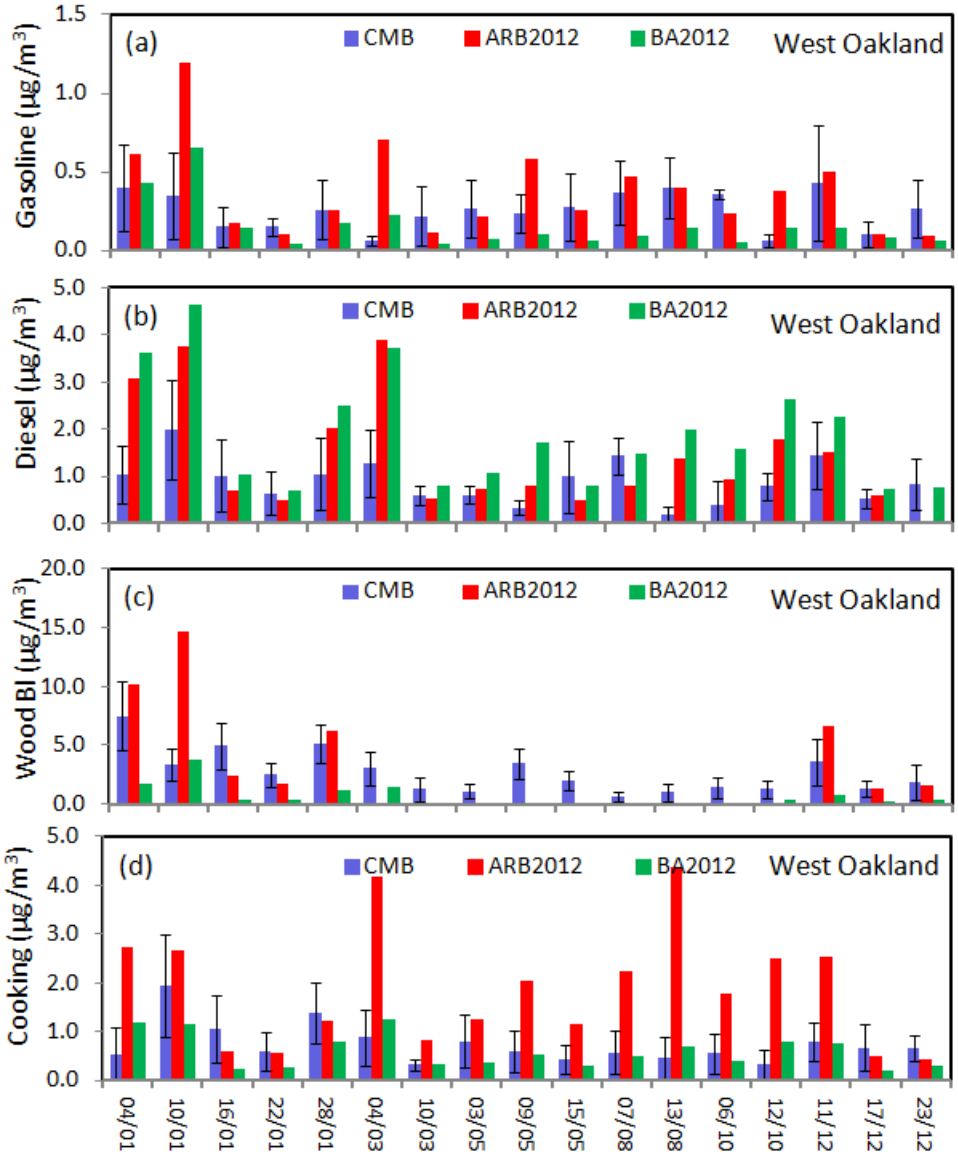


Figure 14 Source contributions to primary PM2.5 mass concentrations in West Oakland

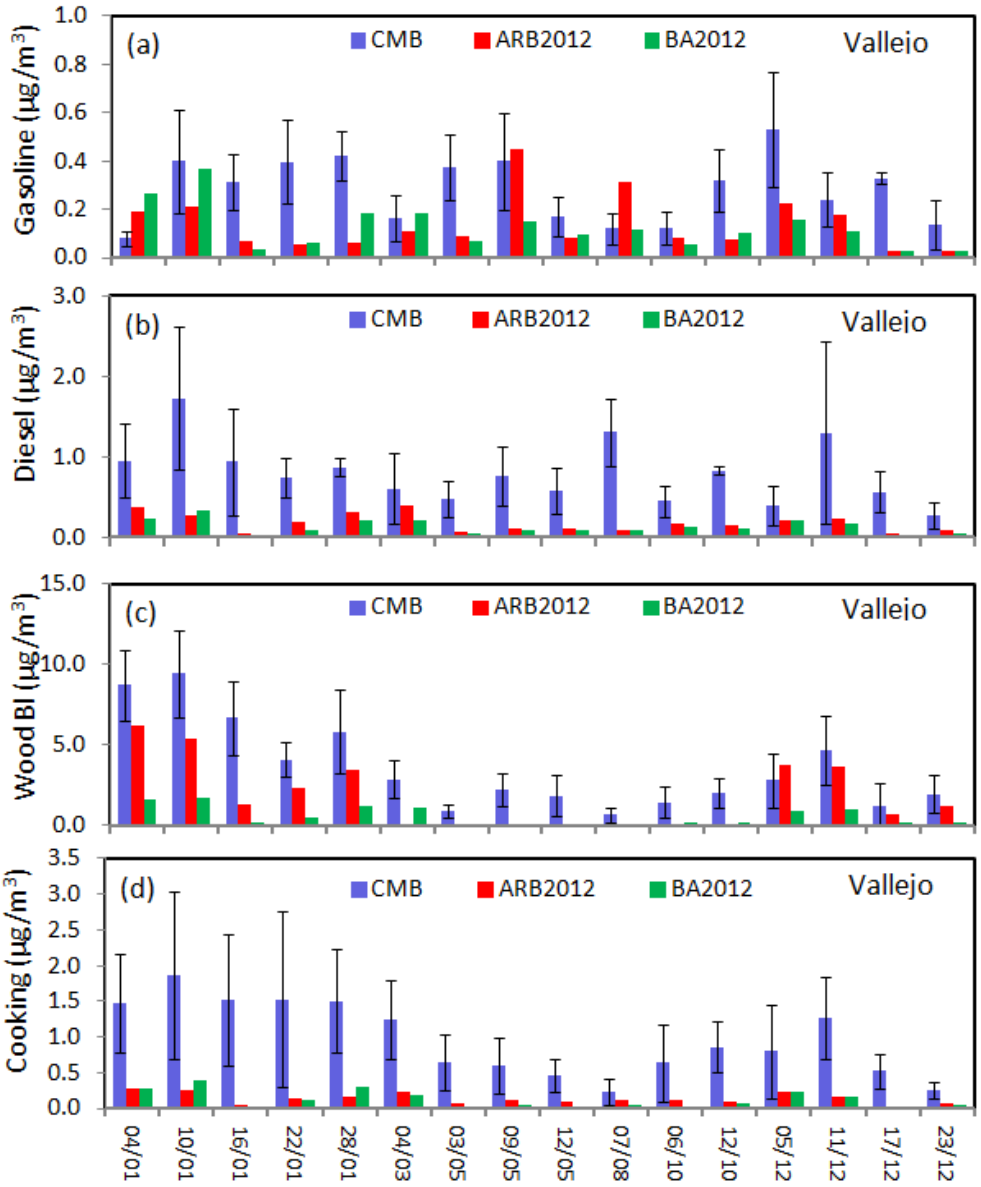


Figure 15 Source contributions to primary $PM_{2.5}$ mass concentrations in Vallejo

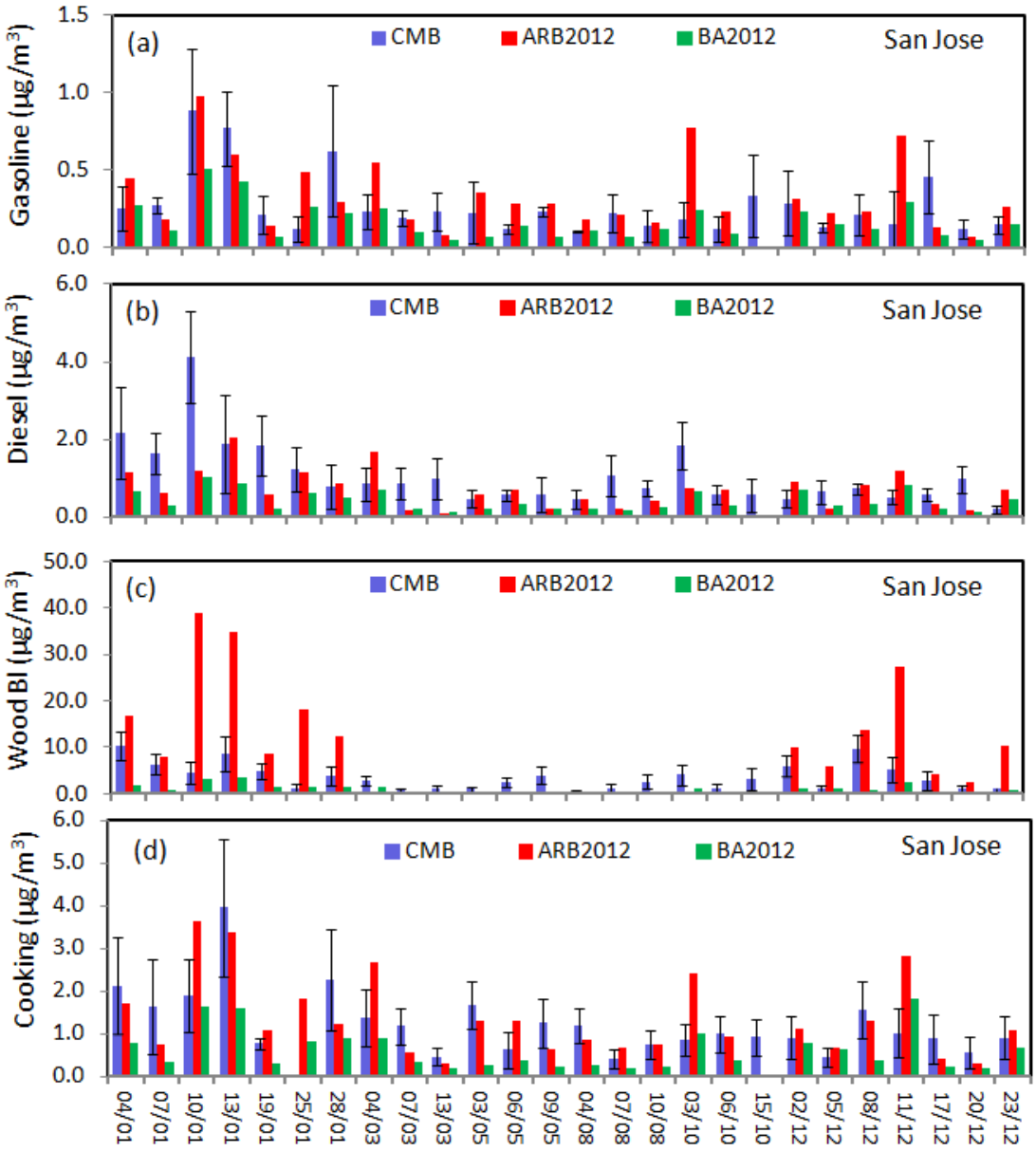


Figure 16 Source contributions to primary $PM_{2.5}$ mass concentrations in San Jose

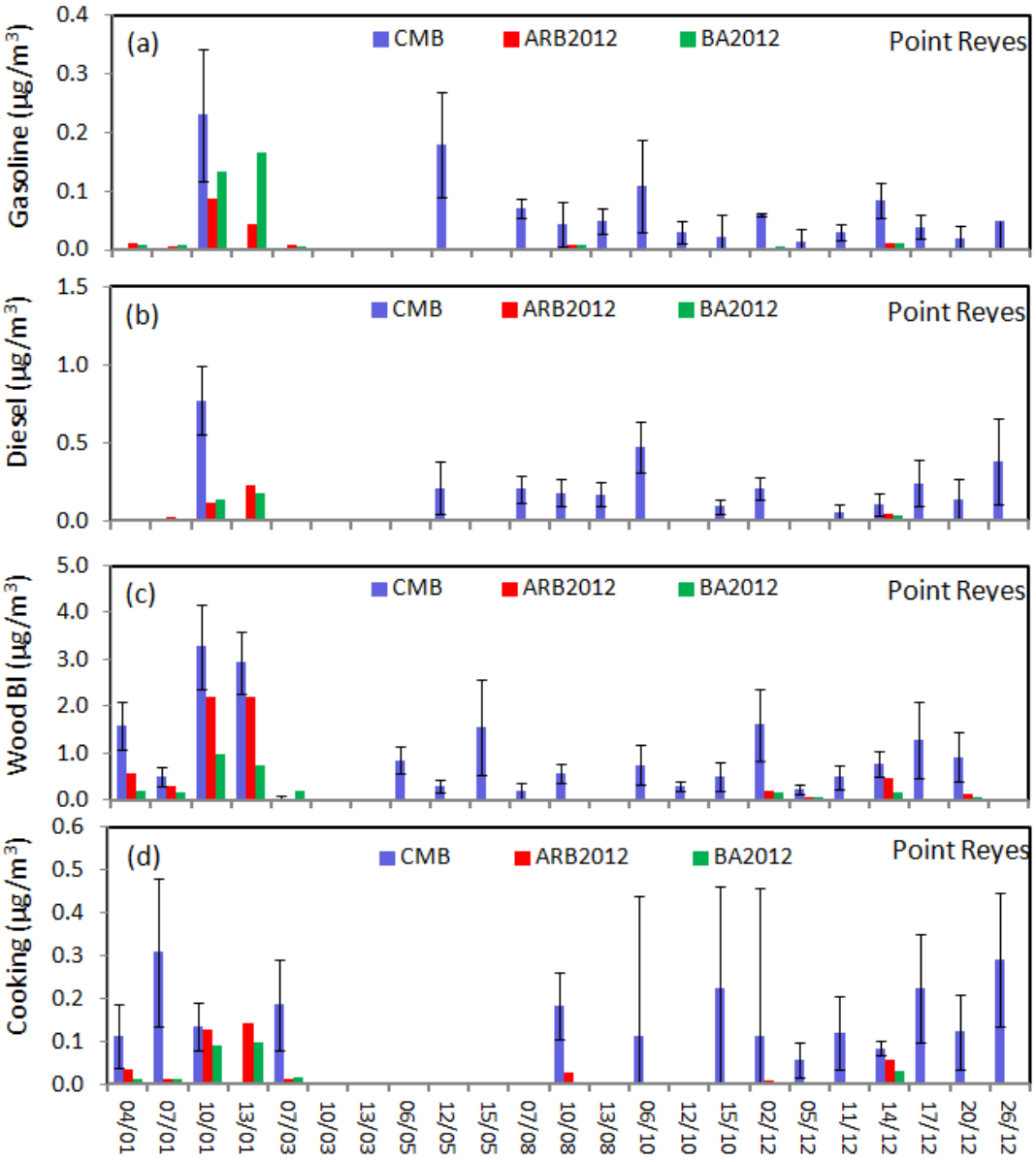


Figure 17 Source contributions to primary $PM_{2.5}$ mass concentrations in Point Reyes.

The fraction of PM2.5 attributed to traffic sources at each of the monitoring sites is shown in Figure 18 and summarized in Table 3. CMB estimates for the traffic fraction of PM2.5 mass vary between 11-17% at inland sites, while UCD/CIT calculations range between 4-12% using the ARB2012 case and 5-30% for the BA2012 case. The UCD/CIT predictions for traffic contributions to PM2.5 mass were lower than CMB estimates at all sites except for the West Oakland site with the BA2012 emissions case.

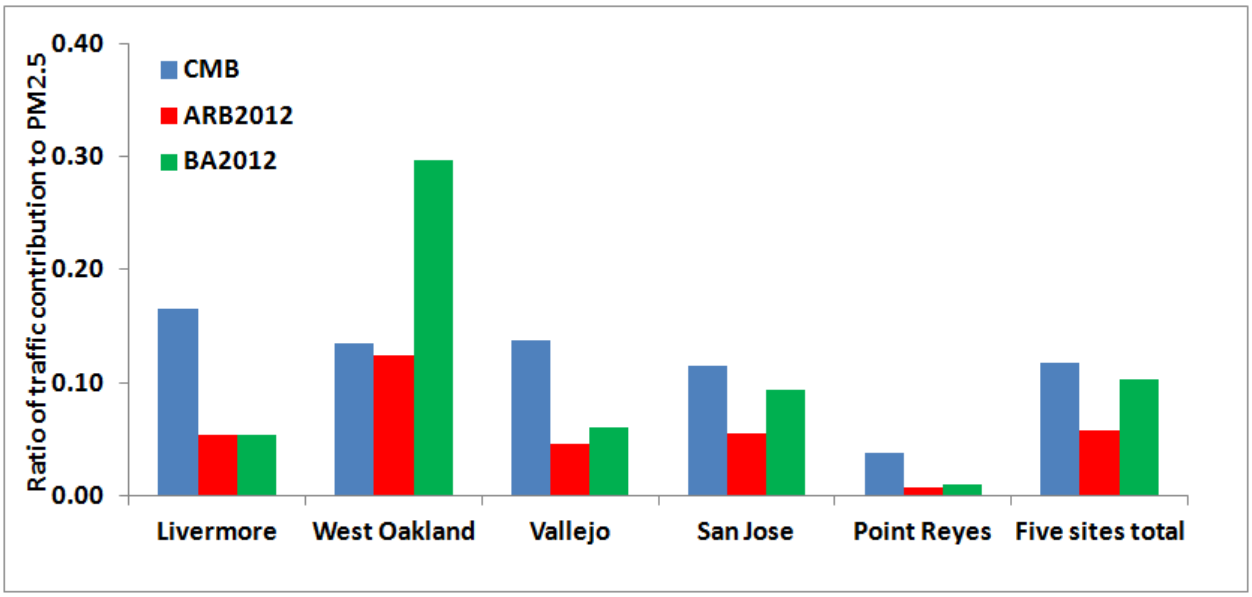


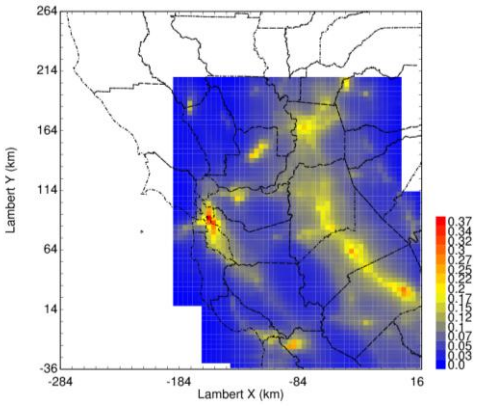
Figure 18 Fraction of total PM2.5 mass attributed to primary PM from traffic sources during 2012.

Table 4 Fraction of total PM2.5 mass attributed to primary PM from traffic sources during 2012.

	Livermore	West Oakland	Vallejo	San Jose	Point Reyes	Five sites total
CMB	0.17	0.13	0.14	0.11	0.04	0.12
ARB2012	0.05	0.12	0.04	0.05	0.01	0.06
BA2012	0.05	0.30	0.06	0.09	0.01	0.10

The fraction of PM2.5 attributed to traffic sources across the entire model domain is shown in Figure 19 using the BA2012 case and the ARB2012 case. As expected, the fraction of PM2.5 mass attributed to traffic sources is highest over major urban areas and transportation corridors. Predicted maximum traffic contributions are 37% over West Oakland using the BA2012 case. These estimates are significantly higher than CMB estimates (see Figure 18 and Table 4). Predicted maximum traffic contributions are 13% over West Oakland using the ARB2012 case. These predictions are more comparable to CMB estimates but regionally they fall below CMB estimates. These results generally suggest that the magnitude and spatial distribution of traffic emissions in the SFBA could benefit from additional research.

(a) BAAQMD2012 Emissions Case



(b) ARB2012 Emissions Case

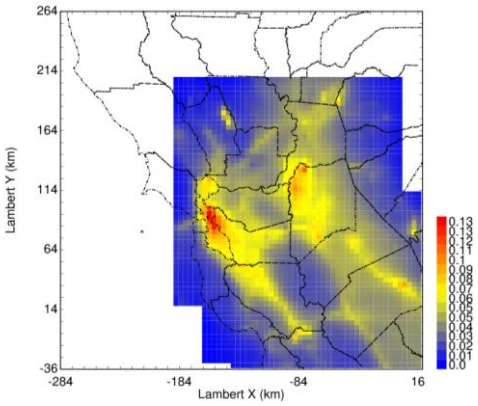


Figure 19: Fraction of total PM2.5 mass attributed to primary PM from traffic sources during 2012.

3.6 Response of PM2.5 mass concentration to the emission reduction from different pollution precursors in Bay Area

To assess the effectiveness of hypothetical control strategies for major pollution precursors in the Bay Area, 5 individual emission reduction scenarios were designed for NOx, VOC, SOx, NH3, and PM alone. A 6^{th} combined emission reduction scenario was also considered for NOx, VOC, SOx, NH3 and PM together. The reduction rate in all scenarios was fixed at 20%. January 2-15, May 2-15, August 2-15 and October 2-15 are selected for study corresponding to periods simulated by BAAQMD. Figures 20-23 show the spatial distribution of PM2.5 mass concentration from UCD/CIT with BAAQMD 2012 emission in base case (center panel) and PM2.5 changes (base case minus control case) during selected simulation periods, respectively. Reductions in primary PM2.5 emissions have the greatest impact on predicted ambient PM2.5 concentrations across all model episodes. The control of gaseous pollutant emissions has limited impact on predicted PM2.5 concentrations in the Bay Area.

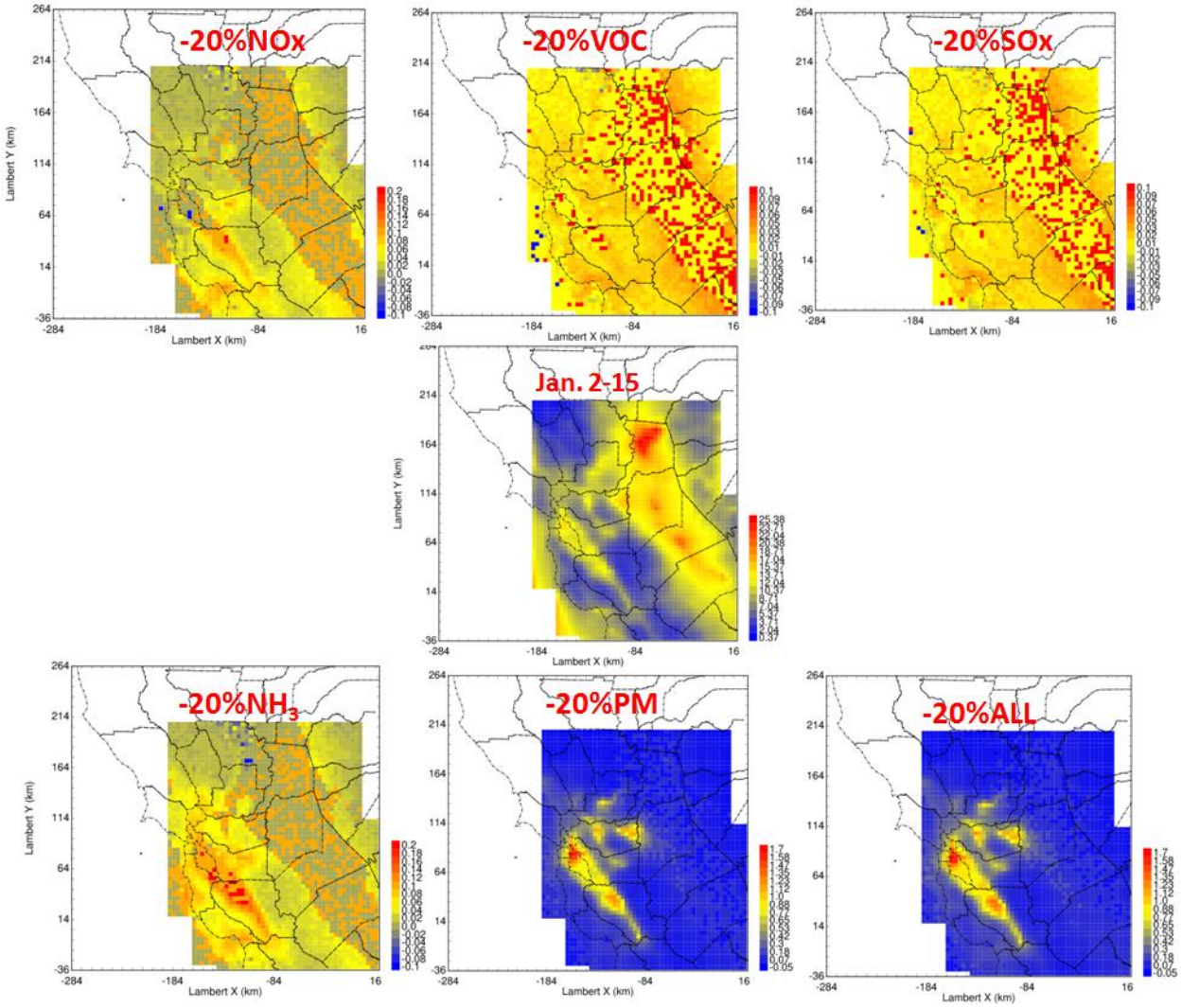


Figure 20 Spatial distribution of average PM2.5 in base case (center) and PM2.5 changes (base case - scenario case) under the six scenarios, during January 2-15.

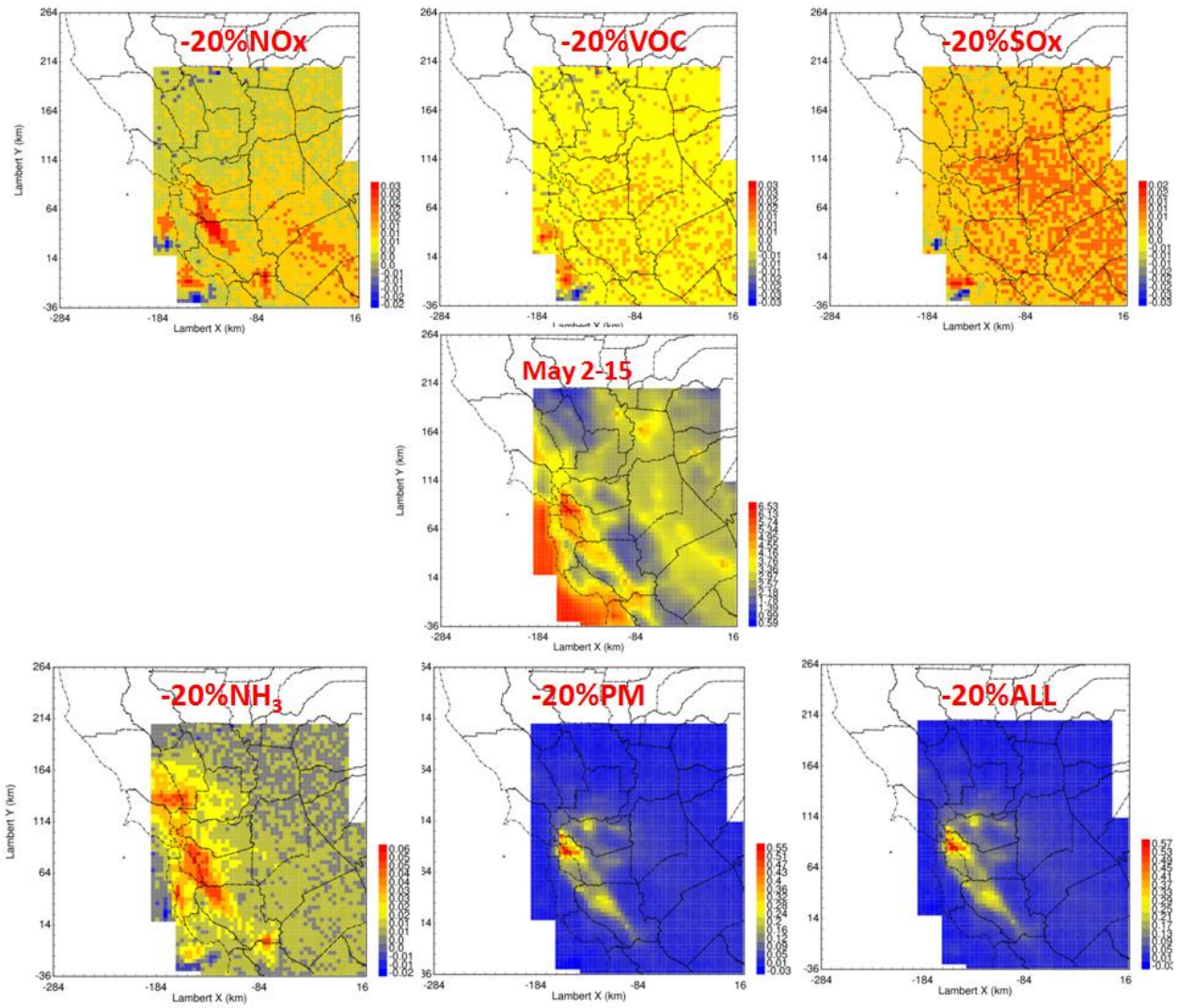


Figure 21 Spatial distribution of average PM2.5 in base case (center) and PM2.5 changes (base case - scenario case) under the six scenarios, during May 2-15.

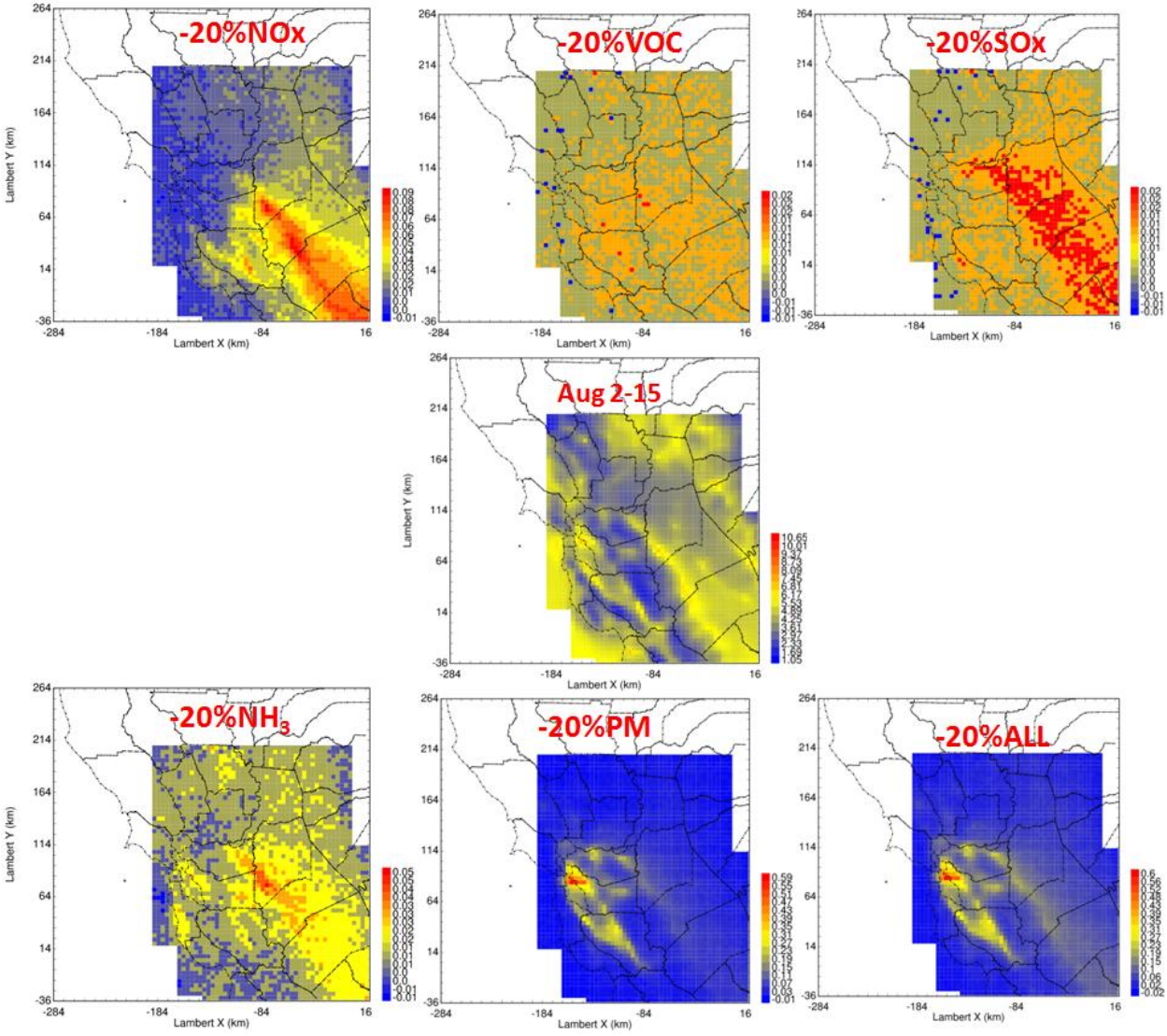


Figure 22 Spatial distribution of average PM2.5 in base case (center) and PM2.5 changes (base case -scenario case) under the six scenarios, during August 2-15.

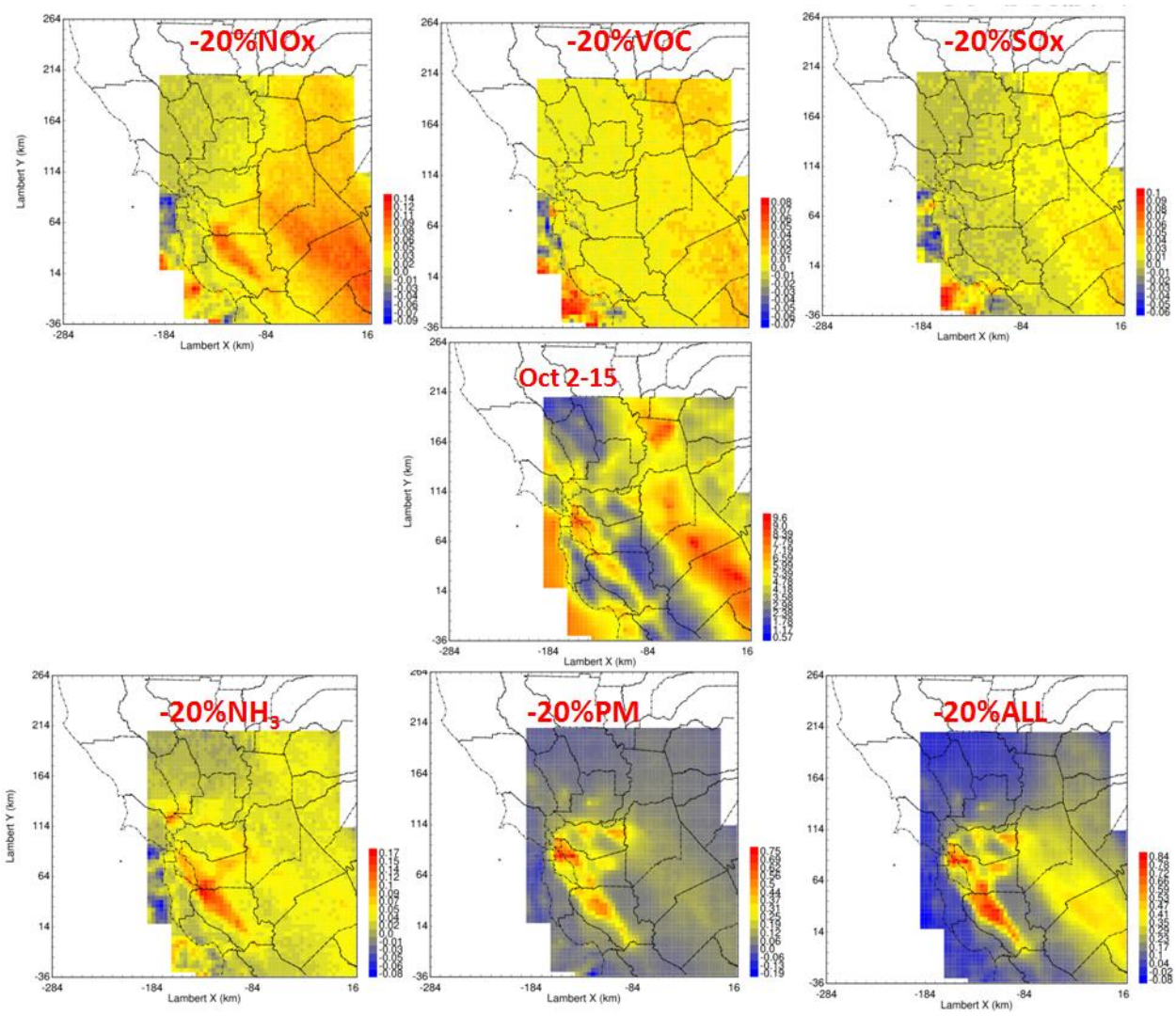


Figure 23 Spatial distribution of average PM2.5 in base case (center) and PM2.5 changes (base case -scenario case) under the six scenarios, during Oct 2-15.

4. Conclusions

CMB calculations carried out on ambient measurements of PM2.5 at 5 sites in the Bay Area are able to predict plausible primary source contributions from diesel tailpipe, gasoline tailpipe, brake wear, road dust, cooking, and biomass burning, as well as generic secondary source contributions from ammonium sulfate, ammonium nitrate, and marine. A successful comparison of these results to independent predictions from regional chemical transport models will build confidence in the results from both CMB and the CTM.

The PMF calculations carried out on the same ambient measurements were only able to resolve primary source contributions from mobile (=diesel+gasoline tailpipe), road dust, biomass burning, and generic secondary source contributions from ammonium sulfate, ammonium nitrate, and marine. A comparison between the common sources in both models shows reasonable agreement for biomass burning, but less agreement for the sum of CMB gasoline+diesel vs. PMF mobile. It seems likely the PMF "mobile" factor is the combination of gasoline, diesel, brake wear, and cooking sources. Additional work is needed to split this profile into these contributing sources.

Predictions of $PM_{2.5}$ species (total mass, OC, EC, sulfate, nitrate) from the CMAQ model using the BAAQMD 2012 emissions inventory, the UCD/CIT model with ARB 2010 emission inventory and the UCD/CIT model with BAAQMD 2012 emission inventory are in general agreement. Predictions from all models have MFBs within ±0.5 and MFE less than 0.80 for PM2.5 total mass and PM2.5 OC, indicating good agreement between predictions and measurement. All models generally under predicted nitrate concentrations. The UCD/CIT model consistently overestimated sulfate concentration, while the CMAQ model slightly under-predicted high concentrations of sulfate and overestimated low concentrations of sulfate.

Source contributions predicted by the UCD/CIT air quality model with ARB 2010 inventory and BAAQMD 2012 inventory were compared to receptor-oriented source apportionment results produced by the Chemical Mass Balance (CMB) model at 5 sites in the Bay Area. Wood smoke contributions predicted by UCD/CIT model with the BAAQMD 2012 inventory were significantly lower than wood smoke contributions predicted with the ARB 2010 inventory. Gasoline and diesel contributions from UCD/CIT model with BAAQMD 2012 inventory and with ARB 2010 inventory are moderately different. All of these trends can be explained by the modifications to the emissions inventory carried out by BAAQMD staff, and the current results provide feedback for further work in this area. Comparison between predicted wood smoke contributions calculated using the CMB model and the UCD/CIT model with the BAAQMD emissions suggests that the BAAQMD emissions inventory may slightly under predict wood smoke contributions at the monitoring sites considered. The BAAQMD should carefully review estimated wood smoke emissions in the Bay Area.

The PM2.5 concentration response to emissions controls for different pollutants, i.e. NOx, VOC, SOx, PM, NH3 predicted by the UCD/CIT model with BAAQMD 2012 emission inventory generally matches the predicted response from the CMAQ model. Ambient PM2.5 concentrations in the Bay Area respond most strongly to controls on primary PM2.5 emissions, with more subtle effects from controls on gas phase emissions.

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