Long-term Particulate Matter Modeling for Health Effects Studies in California – Part I: Model Performance on Temporal and Spatial Variations

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Abstract

For the first time, a ~decadal (9 years from 2000 to 2008) air quality model simulation with 4 km horizontal resolution over populated regions and daily time resolution has been conducted for~~in~~ California to provide air quality data for health effects studies. Model predictions are compared to measurements to evaluate the accuracy of the simulation with an emphasis on spatial and temporal variations that could be used in epidemiology studies. Better model performance is found at longer averaging times, suggesting that model results with averaging times ≥1 month should be the first to be considered in epidemiological studies. The UCD/CIT model predicts spatial and temporal variations in the concentrations of O_3, $PM_{2.5}$, elemental carbon (EC), organic carbon (~~,~~OC),~~,~~ nitrate, and ammonium that meet standard modeling performance criteria when compared to monthly-averaged measurements. Predicted sulfate concentrations do not meet target performance metrics due to missing sulfur sources in the emissions. Predicted seasonal and annual variations of $PM_{2.5}$, EC, OC, nitrate, and ammonium have mean fractional biases that meet the model performance criteria in 95%, 100%, 71%, 73%, and 92% of the simulated months, respectively. The base dataset provides an improvement for predicted population exposure to PM concentrations in California compared to exposures estimated by central site monitors operated one day out of every 3 days at a few urban locations.

Uncertainties in the model predictions arise from several issues. Incomplete understanding of secondary organic aerosol formation mechanisms leads to OC bias in the model results in summertime but does not affect OC predictions in winter when concentrations are typically highest. The CO and NO (species dominated by mobile emissions) results reveal temporal and spatial uncertainties associated with the mobile emissions generated by the

EMFAC 2007 model. The WRF model tends to over-predict wind speed during stagnation events, leading to under-predictions of high PM concentrations, usually in winter months. The WRF model also generally under-predicts relative humidity, resulting in less particulate nitrate formation, especially during winter months. These limitations mustissues will be recognized when using dataimproved in healthfuture studies. All model results included in the current manuscript can be downloaded free of charge at http://faculty.engineering.ucdavis.edu/kleeman/.

Key Words: particulate matter, chemical transport models, temporal variation, spatial variation

1. Introduction

Numerous scientific studies have demonstrated associations between exposure to ambient airborne particulate matter (PM) and a variety of health effects, such as cardiovascular diseases (Dockery, 2001; Ford et al., 1998; Franchini and Mannucci, 2009; Langrish et al., 2012; Le Tertre et al., 2002), respiratory diseases (Gordian et al., 1996; Hacon et al., 2007; Hughes and Tolsma, 2002; Willers et al., 2013), low birth weight and birth defects (Barnett et al., 2011; Bell et al., 2010; Brauer et al., 2008; Laurent et al., 2014; Laurent et al., 2013; Stieb et al., 2012), lung cancer (Beelen et al., 2008; Beeson et al., 1998; Pope et al., 2002; Vineis et al., 2006), mortality and life expectancy (Chen et al., 2013; Correia et al., 2013; Dockery et al., 1993; Franklin et al., 2007; Goldgewicht, 2007; Kan and Gu, 2011; Laden et al., 2000; Ostro et al., 2006; Pope et al., 2009). Recently a few studies have investigated the associations between particle composition and health effects (Bell et al., 2010; Bell et al., 2007; Burnett et al., 2000; Cao et al., 2012; Franklin et al., 2008; Ito et al., 2011; Krall et al., 2013; Levy et al., 2012; Mar et al., 2000; Ostro et al., 2007; Ostro et al., 2010; Son et al., 2012). However, there remains large uncertainty about which PM components are most responsible for the observed health effects, possibly due to the

fact that central site monitoring measurements used in the PM composition studies have limited temporal, spatial, and chemical resolution, which could potentially lead to misclassification of exposure estimates and mask some detailed correlations. Central site PM measurements typically have a collection schedule of 1 sample every 3 or 6 days at a few sites used to represent an entire population region. Important~~In addition, important~~ particle size distribution and chemical composition information is not always routinely measured. Additional information relating PM composition to health effects would provide a solid foundation to design effective PM control strategies to protect public health at a reduced economic and social cost.

Chemical transport models (CTMs) have recently been used as one of the alternative approaches to address the limitations of central site monitors (Anenberg et al., 2010; Bravo et al., 2012; Sarnat et al., 2011; Tainio et al., 2012)~~; Bravo et al., 2012; Sarnat et al., 2011; Tainio et al., 2012)~~. The latest generation of CTMs represents a "state-of-the-science" understanding of emissions, transport and atmospheric chemistry. CTM predictions provide more detailed composition information and full spatial coverage of air pollution impacts with a typical temporal resolution of 1 hour. CTMs have great potential to fill the time and space gaps in the central site monitoring dataset for PM measurements leading to improved exposure assessment in epidemiological studies.

The CTM applications in epidemiology studies to date have generally used relatively coarse spatial resolutions in order to reduce computational burden. Global CTMs have used horizontal resolutions of over 100 km and regional CTMs have used resolution of 12-36 km. These resolutions cannot capture fine spatial gradients of PM concentrations, especially in areas

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with diverse topography and demography. Previous CTMs predictions used in epidemiology studies have also been limited to time periods less than one year. Recently Zhang et al. (Zhang et al., 2014a) evaluated the performance of the Community Multiscale Air Quality (CMAQ) model over a 7-year period in the Eastern United States (U.S.), but no other long-term CTMs studies for health effects analyses have been published to date. As a further limitation, previous epidemiology studies based on CTM predictions have mostly used predicted particles with aerodynamic diameter less than 2.5μm ($PM_{2.5}$) mass concentrations without taking full advantage of the ability of CTMs to simultaneously estimate population exposure to multiple particle size fractions, chemical components, and source contributions. The variation in CTM prediction bias as a function of space and time due to uncertainties in model inputs (emissions, meteorological fields, mechanism parameters) is often not sufficiently characterized to understand potential impacts on health effects estimates. Detailed analyses are needed to assess the temporal and spatial features of CTM predictions to identify accurate and/or unbiased information for exposure assessment before such information can be applied in health effect studies (Beevers et al., 2013).

The objective of the current study is to develop and apply advanced source-oriented CTMs to predict the concentrations and sources for enhanced PM exposure assessment in epidemiological studies over a long-term period with high spatial resolution in California. California is chosen as the focus area for the current study because it has extensive infrastructure to support CTM studies, and it has one of the largest populations in the U.S. that is experiencing unhealthy levels of PM pollution. In 2013, 104 U.S. counties with a population of 65 million people are in non-attainment with the National Ambient Air Quality Standards (NAAQS) for

$PM_{2.5}$ (EPA, 2013). Approximately half of that population (31 million people) lives in 29 California counties meaning that California suffers a disproportionately large share of U.S. PM-related mortality (Fann et al., 2012). The California Air Resources Board (CARB) estimates that 14000 – 24000 California residents die prematurely each year due to particulate air pollution (Tran, 2008). The severity of this problem has motivated extensive investments to support air pollution studies. California has the densest ambient PM measurement network, accurate emissions inventories, and the most health effects study groups of any state in the United States. Rich datasets are available to support model application and evaluation.

The current study is the first attempt to address the sparse PM data problem in exposure assessment using CTM results over a ~decadal time period (9 years from 2000 to 2008) over a domain spanning ~1000 km at a spatial resolution of 4 km. Companion studies have modeled primary $PM_{2.5}$ and $PM_{0.1}$ (particles with aerodynamic diameter less than 0.1μm) concentrations and sources in California (Hu et al., 2014a; Hu et al., 2014b). The current paper, as the third in the series, focuses on model evaluation of total (=primary+secondary) $PM_{2.5}$ and major components elemental carbon (EC), organic carbon~~compounds~~ (OC), nitrate, sulfate, ammonium), emphasizing the aspects of temporal and spatial variations, to identify the features of the CTM results that could add skill to the exposure assessment for epidemiological studies. A future study will investigate the model capability for PM source apportionment of primary and secondary organic aerosols~~OC~~, which is currently an area with great uncertainty.

2. Methods

2.1 Air Quality Model Description

The host air quality model employed in the current study is based on the Eulerian source-oriented University of California-Davis/California Institute of Technology (UCD/CIT) chemical transport model (Chen et al., 2010; Held, 2004; Held et al., 2005; Hu et al., 2012; Hu et al., 2010; Kleeman and Cass, 2001; Kleeman et al., 1997; Kleeman et al., 2007; Mahmud, 2010; Mysliwiec and Kleeman, 2002; Rasmussen et al., 2013; Ying, 2008; Ying et al., 2007; Ying and Kleeman, 2006; Zhang and Ying, 2010). The UCD/CIT model includes a complete description of atmospheric transport, deposition, chemical reaction, and gas-particle transfer. The details of the standard algorithms used in the UCD/CIT family of models have been described in the above references and therefore are not repeated here. Only the aspects that are updated during the current study are discussed in the following section.

The photochemical mechanism used by the UCD/CIT model was updated to reflect the latest information from smog-chamber experiments. The SAPRC-11 photochemical mechanism (Carter and Heo, 2012a; Carter and Heo, 2013) was used to describe the gas-phase chemical reactions in the atmosphere. The secondary organic aerosol (SOA) treatment was updated following the method described in Carlton et al. (Carlton et al., 2010). Seven organic species (isoprene, monoterpenes, sesquiterpenes, long-chain alkanes, high-yield aromatics, low-yield aromatics, and benzene) are considered as precursors for SOA formation. A total of twelve semi-volatile products and seven nonvolatile products are formed from the oxidation of the precursor species. The gas-particle transfer of the semi-volatile and nonvolatile products in the UCD/CIT model is dynamically calculated based on the gas vapor pressures calculated over the particle surface and the kinetic limitations to mass transfer. The explicit chemical reactions and the

parameters for the thermodynamic equilibrium calculation (i.e., enthalpy of vaporization, saturation concentrations, and stoichiometric yields) are provided in Carlton et al. and references therein (Carlton et al., 2010).

Model simulations were configured using a one-way nesting technique with a parent domain of 24 km horizontal resolution that covered the entire state of California (referred to as CA_24km) and two nested domains with 4 km horizontal resolution that covered the Southern California Air Basin (SoCAB) (referred to as SoCAB_4km) and San Francisco Bay Area + San Joaquin Valley (SJV) + South Sacramento Valley air basins (referred to as SJV_4km) (shown in Figure 1). The nested 4 km resolution domains are configured to cover the major ocean, coast, urban, and rural regions that influence California's air quality and, most importantly, to cover most of the California's population for the purpose of health effects analyses. Over 92% of California's population lives in the 4 km domains based on the most recent census information. 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 U.S. 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 (Ying et al., 2007) 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 (Zhang et al., 2014b).

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2.2 Meteorology and Emissions

Hourly meteorology inputs (wind, temperature, humidity, precipitation, radiation, air density, and mixing layer height) were generated using the Weather Research and Forecasting model (WRF) v3.1.1 (Wei Wang, January 2010; William C. Skamarock, June 2008). Two-way nesting was used with the outer domain at 12 km resolution and the inner nested domain at 4 km resolution. North American Regional Reanalysis (NARR) data with 32 km resolution and 3-hour time resolution was used as initial and boundary conditions of the coarse 12 km domain. The WRF model was configured with 31 vertical layers up to 100 hPa~~hpa~~ (around 16 km). Four-dimensional data assimilation (FDDA) was used. The YSU boundary layer scheme, thermal diffusion land-surface scheme, and Monin-Obukhov surface layer scheme were used based on results from a previous study in California (Mahmud, 2010; Zhao et al., 2011). The surface wind was over-predicted with the original version of WRF, especially for wind speed less than 3 m/s, consistent with other studies in California (Angevine et al., 2012; Fast et al., 2014; Michelson et al., 2010a). Over-prediction of the slow winds caused under-prediction of concentrations during high pollution events. A recent study (Mass, C.F, personal communication) found that increasing the surface~~Surface~~ friction velocity (u*) ~~was increased~~ by 50% reduced~~to improve~~ the bias in surface wind predictions in a complex-terrain domain. This technique was tested and adopted in previous studies (Hu et al., 2012; Hu et al., 2014a) where it improved the accuracy of air quality predictions. In the current study, a 1-year ~~. A~~ sensitivity simulation ~~conducted~~ for California in the year of 2000 revealed that increasing u* by 50% improved the mean wind bias from 1.15 m/s to −0.50 m/s, and lowered the root-mean-square error from 2.95 to 2.20 m/s (Hu et

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al., 2014a).~~.~~ Hourly average meteorology outputs at the air quality model vertical layer heights were created. The meteorology predictions were evaluated against meteorological observations (CARB, 2011a). The meteorological statistical evaluation over the period 2000-2006 has been presented in a previous study (Hu et al., 2014a)~~(Hu et al., 2014a)~~, and the results in the period 2007-2008 are consistent with those years. In summary, meteorology predictions of temperature and wind speed generally meet benchmarks suggested by Emery et al. (2001). Mean fractional biases (MFBs) of temperature and wind are generally within ±0.15, root mean square errors (RMSEs) of temperature are around 4 $^{\circ}$C, and RMSEs of wind are generally lower than 2.0 m/s, especially in the SoCAB and SJV air basins which are the focus of the current study. Relative humidity is under-predicted, consistent with findings in other studies in California (Bao, 2008; Michelson et al., 2010). Wind, temperature and humidity are the major meteorological factors that influence the PM concentrations. Further discussions of the uncertainties in meteorology predictions on PM predictions are included in the Results ~~and Discussions~~ section.

Hourly gridded gas and particulate emissions were generated using an updated version of the emissions model described by Kleeman and Cass (Kleeman and Cass, 1998). The standard emissions inventories from anthropogenic sources (i.e., point sources, stationary area sources, and mobile sources) were provided by CARB. Size and composition resolved particle emissions were specified using a library of primary particle source profiles measured during actual source tests (Cooper, 1989; Harley et al., 1992; Hildemann et al., 1991a; Hildemann et al., 1991b; Houck, 1989; Kleeman et al., 2008; Kleeman et al., 1999, 2000; Robert et al., 2007a; Robert et al., 2007b; Schauer et al., 1999a, b, 2001, 2002a, b; Taback et al., 1979). A few studies have revealed some uncertainties associated with the standard emissions inventories. Millstein and

Harley (Millstein and Harley, 2009) found that PM and NOx emissions from diesel-powered construction equipment were over-estimated by a factor of 3.1 and 4.5, respectively. Countess (Countess, 2003) suggested that a scaling factor of 0.33 – 0.74 should be applied to the fugitive dust emissions in the California's San Joaquin Valley. Therefore, scaling factors of 0.32 for off-road diesel sources and 0.50 for dust emissions were applied in the current study. The EMFAC 2007 model (CARB, 2008) was used to scale the mobile emissions using predicted temperature and relative humidity fields through the entire nine-year modeling episode. Biogenic emissions were generated using the Biogenic Emissions Inventory System v3.14 (BEIS3.14), which includes a 1-km resolution land cover database with 230 different vegetation types (Vukovich and Pierce, 2002). Sea-salt emissions were generated on-line based on the formulation described by de Leeuw et al. (de Leeuw et al., 2000) for the surf zone and the formulation described by Gong (Gong, 2003) for the open ocean. Emissions from wildfires and open burning at 1 km × 1 km resolution were obtained from the Fire INventory from NCAR (FINN) (Hodzic et al., 2007; Wiedinmyer et al., 2011). The FINN inventory provides SAPRC99 speciated daily emissions of gaseous and particulate emissions (EC, organic matter (OM), ~~OC,~~ $PM_{2.5}$ and PM_{10}) based on satellite observations of open burning events. Each open burning event is allocated to model grid cells of each domain based on the reported longitude/latitude of the event and the area burned. The emissions were injected at the height of the atmospheric mixing layer (PBL). The temporal variation of wildfire emissions was obtained from the Western Regional Air Partnership (WRAP) report (WRAP, 2005). A size distribution profile was calculated based on assumptions described in Hodzic et al. (Hodzic et al., 2007).

2.3 Ambient Air Quality Measurements

The evaluation dataset was compiled from several measurement networks, including CARB's "2011 Air Quality Data DVD" (CARB, 2011b) and the database maintained by the Interagency Monitoring of Protected Visual Environments (IMPROVE). The data DVD includes daily average mass concentrations of $PM_{2.5}$, EC, OC, nitrate, sulfate, ammonium, and trace metals every 3 or 6 days at the sites of the $PM_{2.5}$ Speciation Trends Network (STN) and the State and Local Air Monitoring Stations (SLAMS). There are a total 13 $PM_{2.5}$ speciation sites included in the DVD covered in the 4 km domains during the modeling periods. The precision of STN measurements is estimated to be 3.5%, 8.6%, and 3.9% for sulfate, nitrate, and ammonium, respectively (Sickles Ii and Shadwick, 2002). Measured EC concentrations at 5 sites are found to be exactly 0.5 µg/m3 on > 80% of the measurement days, suggesting corrupt or missing data at these locations. Therefore these 5 sites were excluded in the evaluation for EC, but still included in the evaluation for other PM components. The OC data were not blank corrected, resulting in a positive artifact by the NIOSH5040 method that is equivalent to approximately 1 $\mu g/m^3$. Measured OC concentrations were blank corrected in the current study by subtracting 1 $\mu g/m^3$ from all OC measurements. The IMPROVE network provides daily average mass concentrations every 3 days for $PM_{2.5}$, EC, OC, nitrate, sulfate, and soil. There are a total of 9 IMPROVE sites covered in the 4 km domains. The precision of IMPROVE measurements is estimated to be 4–6% for $PM_{2.5}$ mass, nitrate, and sulfate, and to be > 15% for EC and OC (http://vista.cira.colostate.edu/improve/Publications/OtherDocs/IMPROVEDataGuide/IMPROVEDataGuide.htm). Daily average PM_{10} mass measurements and hourly measurements of several key gaseous pollutants (ozone, CO, NO, NO_2, and SO_2) are also included in the data DVD. There are a total of 66 $PM_{2.5}$ Federal Reference Method (FRM) sites covered in the 4 km domains.

Frank (Frank, 2006) found that FRM $PM_{2.5}$ mass measured using STN monitors was within ± 30% of reconstructed fine mass (RCFM) concentrations measured using IMPROVE monitors.

3. Results and Discussion~~Discussions~~

3.1 Statistical evaluation

Statistical measures of MFB and mean fractional error (MFE) were calculated to evaluate the accuracy of model estimates in space and time. Boylan and Russell (Boylan and Russell, 2006) proposed concentration dependent MFB and MFE performance goals and criteria, realizing 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.

Figures 2 and 3 show the monthly MFB and MFE values, respectively, of predicted daily average EC, OC, nitrate, ammonium, sulfate and total $PM_{2.5}$ mass in the 4 km domains. Measured EC, OC, nitrate, ammonium, and total $PM_{2.5}$ mass concentrations follow similar seasonal patterns with high concentrations occurring in winters (indicated by blue colors in figures) and low concentrations occurring in summers (indicated by red colors in figures). These patterns are driven by the meteorological cycles (i.e., lower mixing layer and wind speed providing less dilution, and lower temperature encouraging partitioning of ammonium nitrate to the particle phase) and the emissions variations (i.e., additional wood burning emissions for home heating in winters). The opposite seasonal variations in sulfate concentrations are observed, due to higher oxidation rates from S(IV) to S(VI) and higher sulfur emissions from natural sources in summer (Bates et al., 1992).

EC predictions are in excellent agreement with measurements. MFBs in all months and MFEs in 107 months out of the total 108 months are within the model performance goal. EC MFBs and MFEs show no significant difference among months/seasons, indicating consistently good EC performance during the entire 9-year modeling period. OC, nitrate, sulfate, and ammonium, the PM components that include the secondary formation pathways, meet the MFBs model performance criteria in 71%, 73%, 46%, and 92% of the simulated months, respectively. These components generally have good agreement between predictions and measurements in winter months, with only a few months not meeting the performance criteria. When analyzing by season, predicted concentrations of these species are found to be more biased in summer months, especially for sulfate and nitrate. Different factors influence the seasonal profile of each species. The more significant OC under-prediction in summertime is mainly associated with the under-prediction of SOA due to incomplete knowledge of SOA formation mechanism at the present time. Similar patterns have been reported in other modeling studies outside California (Matsui et al., 2009; Volkamer et al., 2006; Zhang et al., 2014a; Zhang and Ying, 2011). Measured nitrate concentrations in summertime (1-5 $\mu g/m^3$) are factors of 2-5 lower than concentrations in wintertime (5-12 $\mu g/m^3$). Model predictions tend to underestimate the low particle phase nitrate concentrations in summer, especially when temperatures exceed 25 °C. Model predictions for particulate nitrate are usually less than 1 $\mu g/m^3$ under these conditions, while 2-3 $\mu g/m^3$ nitrate concentrations are still observed in the ambient air. Similar under-predictions of summertime nitrate have been reported in other regional modeling studies (Appel et al., 2008; Tesche et al., 2006; Yu et al., 2005; Zhang et al., 2014a). Model calculations reflect thermodynamics and kinetic gas-particle transfer for ammonium nitrate in mixed particles, suggesting that some other

form of nitrate is present in the real atmosphere, such as organo-nitrates (Day et al., 2010). Sulfate concentrations are consistently under-predicted throughout the modeling period at all locations, especially in southern California where the measured sulfate concentrations are highest. Under-prediction of sulfate has also been reported by other regional modeling studies in California (Chen et al., 2014; Fast et al., 2014), using different air quality models (e.g., CMAQ, WRF-Chem). This consistent behavior suggests that the specific model is not the cause of the sulfate under-prediction. A global model study that included ocean DMS emissions showed a better sulfate performance in California (Walker et al., 2012). Therefore,~~because of~~ missing emissions sources such as the sulfur emitted as dimethyl sulfide (DMS) from the Pacific Ocean likely contribute to the sulfate under-predictions in the current study. The sulfate concentrations at the sites in southern California are ~2 to 3 times higher than in northern California, and are under-predicted by an even larger amount (with MFBs around -1.0). It is therefore likely that anthropogenic sulfur sources are missing in southern California in addition to background DMS sources.~~,~~ Ammonium is drawn to acidic particles and so ammonium concentration predictions reflect the combined trends of nitrate and sulfate predictions.

The model predictions of total ~~mass of~~ $PM_{2.5}$ mass, as a summation of all components, show very good agreement with measurements, with only 3 summer months and 2 spring months~~month~~ (5% of all simulated months) not meeting the performance criteria, and 78% and 75% of months within the performance goals for MFB and MFE, respectively. The largest biases in the total $PM_{2.5}$ mass occur in summer. Under-prediction in summer sulfate and OC contribute to negative biases in the total $PM_{2.5}$ mass predictions. Sulfate and OC concentrations in summer accounted for ~18% and ~37% of the total $PM_{2.5}$ mass. Sulfate~~Therefore, sulfate~~ and

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OC under-prediction contributed to a combined ~37% under-prediction of total $PM_{2.5}$ mass. However, positive biases in predicted dust concentrations rich in crustal elements such as aluminum and silica (Hu et al., 2014a)~~(Hu et al., 2014a)~~ compensate for the under-predictions in carbonaceous components and water-soluble ions described above.

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Figure 4 shows the MFB and MFE values of particulate species of $PM_{2.5}$ total mass, EC, OC, nitrate, sulfate, ammonium and gaseous species of O_3, CO, NO, NO_2, SO_2 using daily averages across all measurement sites during the entire modeled 9-year period. $PM_{2.5}$ total mass, EC, OC, ammonium and gaseous species of O_3, CO, NO_2 have MFBs within ±0.3 and MFE less than 0.75, indicating general agreement between predictions and measurement for these species. Nitrate and NO have MFBs of -0.4 and -0.28, respectively, but MFEs of 0.8 and 1.07, respectively. The relatively moderate or small bias combined with relatively large error indicates that the daily predictions miss the extremely high and low concentrations. Sulfate and SO_2 have high MFBs of -0.7 and -0.5, respectively, and high MFEs of 0.8 and 0.9, respectively, indicating that these species are consistently under-predicted.

Concentrations averaged over longer times, such as 1 month or 1 year, are used in some air pollution-health effects studies. A previous examination of primary particles in California revealed that air quality model predictions are more accurate over longer averaging time because the influence of extreme events and short-term variability is reduced as the averaging period gets longer (Hu et al., 2014a)~~(Hu et al., 2014a)~~. Figure 4 compares the MFB and MFE values for total (=primary+secondary) particulate matter and gaseous species using daily, monthly, and annual averages across all sites in the 4 km domains. The results demonstrate that longer averaging

times produce better agreement between model predictions and measurements (except for sulfate, which is under-predicted due to missing emissions) because they remove the effects of random measurement errors at monitoring stations and variations in actual emissions rates that are not reflected in seasonally-averaged emissions inventories. The reduced errors associated with longer averaging times indicate that model results may be most useful in epidemiological studies that can take advantage of~~require~~ averaging times ≥1 month.

3.2 Spatial and temporal variations

Figure 5 panel (a) shows the predicted and measured monthly average concentrations of 1-h peak O_3 at 5 major urban sites (Sacramento, Fresno, Bakersfield, Los Angeles, and Riverside). Strong seasonal variations are observed in measured and predicted 1-h peak O_3. The measured 1-h peak O_3 shows seasonal variation from 100 ppb in summertime to 20 ppb in wintertime. The predicted high 1-h peak O_3 concentrations in non-winter months are in good agreement with, or slightly higher than, ambient measured concentrations at all sites. This is consistent with studies in the eastern U.S. (Zhang et al., 2014a), which found similar slight over-predictions of summer O_3 concentrations. Predicted 1-h peak O_3 concentrations in cold winter months, however, are generally higher than measured values. Photochemical reaction rates in wintertime months are slow and the predicted O_3 concentration at the surface mostly reflects downward mixing of the aloft background O_3 followed by titration by surface NO emissions. The STN measurement sites in California are located in urban areas that are close to major freeways (see the site locations and nearby sources information in (Hu et al., 2014a)~~(Hu et al., 2014a)~~). The 4 km × 4 km model grid cells that contain both freeways and monitors dilute the high NO concentrations around the measurement sites leading to an under-prediction of O_3

370 titration and an over-prediction of O_3 concentrations. EPA recommends a threshold O_3 value of
371 60 ppb for model O_3 evaluations (U.S.EPA, 2007), which means that wintertime O_3
372 concentrations at the urban sites will generally not be considered in the formal model evaluation.
373
374 Figure 5 panels (b) and (c) show the predicted and measured monthly average CO and
375 NO concentrations. Strong seasonal variations in CO and NO can be observed, with wintertime
376 concentrations that are a factor of 3-5 higher than summertime concentrations. Model predictions
377 generally reproduce the seasonal variations except at the Riverside site where predicted seasonal
378 variations are weaker than measurements. The model performance varies by simulation year and
379 location. At the Sacramento and Fresno sties, predicted CO is in good agreement with measured
380 concentrations in all months of 2002 through 2006, but CO is under-predicted in winter months
381 of 2000-2001 and slightly over-predicted in most months of 2007-2008. At the Bakersfield site,
382 CO is under-predicted in 2000-2003 and in good agreement with measurements in 2004-2005
383 (after which further measurements are not available). At the Los Angeles site, CO is in good
384 agreement in 2000-2003, and over-predicted in the later years. At the Riverside site, CO is
385 under-predicted in all months of 2000-2003, under-predicted in non-summer months in 2004-
386 2006, and in general agreement with measurements in 2007-2008. NO predictions generally
387 agree well with measured NO concentrations in 2000-2004 at Sacramento, Fresno, Bakersfield
388 and Los Angeles, and then are over-predicted in the later years. NO at Riverside is under-
389 predicted in the winter months of 2000-2003, and over-predicted in the summer months of 2004-
390 2008. Mobile emissions are the dominant sources of CO and NO in California, contributing >
391 80% of total anthropogenic emissions (CARB, 2012). The results of the current modeling study
392 suggest that uncertainties in the mobile emissions exist both in time and space.

A clear and similar decreasing trend is apparent in measured CO and NO concentrations from 2000-2008. This inter-annual trend is not well captured by the model predictions due to the uncertainties in the emissions. An adjusted NO prediction (NO_adj) can be calculated using CO as a tracer for the mobile emissions and dilution according to the equation:

NO_adj = NO_noadj * CO_predicted / CO_measured

where NO_noadj is the NO predictions before the adjustment (i.e., the concentrations showing in Figure 5(c)). NO_adj has higher correlation coefficient (R^2) with measured NO concentrations than the NO_noadj prediction at all the five monitoring sites (as shown in Figure 7~~6~~) and NO_adj has a regression slope closer to 1.0 than NO_noadj at 3 out of 5 sites. This suggests that either emissions or physical dilution processes in the model contribute to the errors observed in Figure 5 (in addition to the possibility of errors in model chemistry). Unfortunately, the large variation in the correction factor among different locations suggests that these scaling factors cannot be simply interpolated/extrapolated from the indicated five monitoring sites to the full modeling domain.

Figure 5 panels (d) and Figure 6 (a~~(e~~) show the predicted and measured monthly average ammonium and nitrate concentrations. Ammonium nitrate is a major $PM_{2.5}$ component in California, especially in wintertime when the low temperature and high relative humidity favor partitioning to the condensed phase. The monthly average ammonium and nitrate results demonstrate similar model performance. The predicted concentrations agree reasonably well with measured ambient concentrations and seasonal variations. Model predictions are lower than measured values in the early years, especially during winter months when concentrations are

highest. This pattern is very consistent with CO model performance, suggesting mobile emissions are under-estimated for the early years of the simulation period. Nitrate is formed through NO oxidation to nitric acid but NO concentrations are not under-predicted, suggesting that the chemical conversion of NO to nitric acid is too slow. Carter and Heo (Carter and Heo, 2012b) suggested that SAPRC11 mechanism systematically under-predicts OH radical concentrations by ~30%, which would be consistent with the observed trends.

Gas-particle partitioning of ammonium nitrate depends on temperature and relative humidity. While there is no systematic bias in WRF temperature, relative humidity is generally under-predicted by up to 40% over California. A one-year sensitivity analysis was conducted with RH increased uniformly by +30% (but not to exceed 95%, and all other meteorological parameters were kept the same)~~%)~~ in 2008 to investigate the impact of the relative humidity bias on particulate nitrate predictions. The arbitrary increase in RH by 30% in the air quality model simulations yields an upper bound estimate of the nitrate sensitivity to RH. Figure 8~~Figure 7~~ compares the monthly average nitrate concentrations predicted with the original RH (denoted as "RH_ori" case) and the enhanced RH (denoted as "RH+0.3" case) at Sacramento and Fresno. Nitrate predictions are generally higher in the "RH+0.3" case due to more particle phase water available to absorb nitrate into the condensed phase. The nitrate predictions at Sacramento are significantly improved during most months in 2008, suggesting this area suffers from the low RH bias in the WRF predictions. Nitrate at Fresno is improved mostly in the winter and spring, but is still under-predicted during the time period with peak winter concentrations, indicating this area is influenced by other factors besides RH. Nitrate predictions at Fresno in summer and fall are lower when RH is enhanced, due to faster deposition caused by larger particle sizes with

more particle phase water. The uniform RH increase of 0.3 in this region is likely unrealistically large during~~in~~ these months.

Figure 6~~5~~ panel (b~~f~~) shows the OC predictions and measurements. Organic aerosol in California it is typically the second most abundant species, after ammonium nitrate. In the comparison, an OM/OC ratio of 1.6 (Turpin and Lim, 2010) is applied to convert primary organic aerosol OM back to OC for comparison to measured concentrations. The conversion ratios for SOA species are taken from Table 1 in Carlton et al. (Carlton et al., 2010). Predicted OC agrees reasonably well with measured concentrations, but is lower than the wintertime high concentrations in the early years, similar to other PM components. Predicted OC in summers is also in good agreement with measurements at the indicated monitoring sites. As mentioned previously, these sites are all near major freeways and therefore OC is dominated by primary organic aerosols. Larger bias is found at sites distant from local sources where SOA becomes more important. More analysis about the concentrations and sources of the OC results are included in a companion paper (Hu, Manuscript in preparation).

Figure 6~~5~~ panel (c~~g~~) shows that predicted EC concentrations agree well with measured concentrations. High measured EC concentrations in a few winter months in the early years are under-predicted, but EC concentrations in the summer months are generally over-predicted.

Figure 6~~5~~ panel (d~~h~~) shows that monthly average predictions for $PM_{2.5}$ mass concentrations agree well with observations, and seasonal trends are generally captured with high concentrations in winter, and low concentrations in summer. $PM_{2.5}$ is over-predicted in

summer months when nitrate, sulfate, and ammonium are found to be under-predicted. These trends reflect the over-prediction of the primary components, mostly dust particles, in the model calculations (Hu et al., 2014a)~~(Hu et al., 2014a)~~. This result suggests that a uniform scaling factor of 0.5 for dust emissions may not be appropriate. A smaller factor (for example, a factor of 0.25 was used in the eastern U.S. (Tesche et al., 2006)) or a spatially resolved method that accounts for the land-use types (Pace, 2005) should be used for future studies in California.

California experiences the highest $PM_{2.5}$ concentrations in wintertime, caused by stagnant meteorological conditions characterized by low wind speed and shallow atmospheric mixing layer. The WRF model tends to over-predict wind speed during low wind speed events ($\leq$ 2 m/s) in California (Zhao et al., 2011). Increasing u* by 50% improves the WRF wind prediction but still over-predicts wind speed during events when measured wind speed is <1.5 m/s. A zero-order approximation of air pollutant concentration (Mahmud, 2010) is:

$$C = \frac{E}{V} = \frac{E}{u \times \mathrm{H}} \quad (1)$$

where C is the pollutant concentration, E is the source pollutant emission rate, V is the air ventilation rate which is equal to (wind speed × mixing height), u and H are the horizontal wind speed and mixing height, respectively. ~~.~~ The concentration is linearly dependent on the inverse wind speed ($1/u$). Figure 9~~8~~ shows the MFBs of the predicted atmospheric inverse wind speed ($1/u$) as a function of the observed atmospheric inverse wind speed. Also shown in Figure 9~~8~~ are the MFBs of PM component concentrations as a function of the observed concentrations. The MFBs decrease when the inverse wind speed or concentrations increase, indicating low inverse wind speed/concentrations are over-predicted, but high inverse wind speed /concentrations are under-predicted. The trends of inverse wind speed and concentrations are well correlated,

indicating that simple wind bias ~~effects on the ventilation rates~~ leads to bias in PM predictions, especially during the events with high PM pollution. The correlation with $1/u$ MFB is stronger for primary PM component(s) than for secondary components, indicating that additional processes affect the secondary PM, such as chemistry, gas-particle partitioning, etc. Sulfate bias has the weakest~~least~~ correlation to inverse ventilation bias, because sulfate bias~~it~~ is mainly driven by the bias in sulfur~~SO_2~~ emissions.

Figure 10~~9~~ shows the predicted 9-year average concentrations of $PM_{2.5}$, EC, OC, nitrate, sulfate, and ammonium, compared with measured average concentrations over California. High concentrations of all PM pollutants occur in the urban areas with large population, indicating that most of the PM is generated by anthropogenic activities. The predicted spatial distributions generally agree well with measurements, but provide much more detailed information. $PM_{2.5}$ concentrations are over-predicted in the SJV air basin due to an over-prediction of agricultural dust. High OC concentrations were measured at two sites in northern California due to intense wood burning. The two sites are in the 24 km model domain but outside the 4 km, therefore the predicted OC concentrations in the 24 km grids do not agree well with the measurements at this location. This finding confirms that 24 km resolution is probably too coarse for health effects studies and justifies the use of~~thee~~ 4 km grids over the majority of California's population in the current work. Background sulfate concentrations at IMPOVE sites were measured to be 0.6-1 $\mu g/m^3$ but higher concentrations of 2~3 $\mu g/m^3$ were measured in Southern California. Model calculations do not reproduce this concentration enhancement, leading to an under-prediction in the concentrations of this $PM_{2.5}$ species.

3.3 Discussion

In general, the reasonable agreement between model predictions and measurement builds confidence that the model predictions can provide a reasonable estimate of exposure fields in locations with no available measurements. The detailed analysis described in the previous section identifies several aspects that must be considered when applying the data in the health effect studies. For the gaseous pollutants, daily maximum O_3 predictions are in good agreement with measurements across the entire modeling domain. Seasonal and annual variations are captured accurately. Therefore daily maximum O_3 predictions can be used in health analyses with high confidence. The predictions also capture the seasonal variations in NO and CO, but do not reflect the long-term trends, especially in southern California. Predicted monthly averages of NO and CO in northern California are preferred over daily averages for use in health analyses. For the PM pollutants, daily concentrations and spatial distributions of EC and total $PM_{2.5}$ mass generally agree well with observations, but monthly averages should be considered first in health studies as they are in better agreement with observations than shorter averages. Predicted OC in winter is also reasonably accurate, but OC in summer should be used with caution. Sulfate and nitrate are both under-predicted. Sulfate has greater bias in southern California than in northern California, while nitrate has consistent bias throughout the modeling domain. This suggests that the spatial distribution information of nitrate might still be useful for health effect studies that use contrasts in exposure as a function of location, but sulfate data are likely not useful in health effects studies at the present time.

Predicted monthly averages for PM concentrations are more accurate than daily averages, suggesting that the PM exposure predictions will be most useful in studies that can take

advantage of averaging times ≥ 30 days. Longer averaging times smooth out short-term PM variations that could be useful in some epidemiological studies that focus on short term changes in health effects. To get more accurate pollutant predictions at shorter timescales would require more accurate representation of emissions, meteorological conditions, and atmospheric chemistry at these time scales. Many intensive studies that manually corrected input data have focused on high temporal resolution for short periods (generally less than 1 month), such as the California Regional PM_{10}/$PM_{2.5}$ Air Quality Study (CRPAQS) (Ying, 2008). It is currently impractical to carry out such efforts for a ~10 year modeling period in which there are a large number of special events that are not represented by automated meteorology and emissions models. The atmospheric modeling community continues to refine tools that can capture and accurately represent these special cases. For example, the current study includes automatic detection and incorporation of wildfire emissions into the modeling system based on satellite observations. This automated feature was not generally available in previous studies. Future advances will detect transportation patterns responding to traffic accidents or holiday traffic jams, drought effects on biogenic emissions, etc. These future advances will improve models to have more accurate predictions in both short- (<1 month) and long- (> 1 month) averaging timesprovide a reasonable estimate of exposure fields.

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4. Conclusions

For the first time, a ~decadal (9 year) CTM air quality model simulation with 4 km horizontal resolution over populated regions has been conducted in California to provide air quality data for health effects studies. Model predictions are compared to measurements in order to evaluate both the spatial and temporal accuracy of the results. The performance of the source-oriented UCD/CIT air quality model is satisfactory for O_3, $PM_{2.5}$, and EC (both spatially and

554 temporally). Predicted OC, nitrate, and ammonium are less satisfactory, but generally meet
555 standard model performance criteria. OC bias is larger in summertime than wintertime mainly
556 due to an incomplete understanding of SOA formation mechanisms. Bias in predicted
557 ammonium nitrate is associated with uncertainties in emissions, the WRF predicted relative
558 humidity fields, and the chemistry mechanism. Predicted sulfate is not satisfactory due to
559 missing sulfur sources in the emissions. The CO and NO (species dominated by mobile
560 emissions) results reveal significant temporal and spatial uncertainties associated with the mobile
561 emissions generated by the EMFAC 2007 model. The WRF model tends to over-predict wind
562 speed during stagnation events, leading to under-predictions of high PM concentrations, usually
563 in winter months. The WRF model also generally under-predicts relative humidity, resulting in
564 less particulate nitrate formation especially during winter months. Despite the issues noted
565 above, predicted spatial distributions of PM components are in reasonably good agreement with
566 measurements. Predicted seasonal and annual variations also generally agree well with
567 measurements. Better model performance with longer averaging time is found in the predictions,
568 suggesting that model results with averaging times ≥1 month should be first considered in
569 epidemiological studies. All model results included in the current manuscript can be
570 downloaded free of charge at http://faculty.engineering.ucdavis.edu/kleeman/.

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Figures and Tables

Figure 1. Modeling domains (blue lines outline the CA_24km domain, and red lines outline the SoCAB_4km (bottom) and SJV_4km domains (up)) and PM measurement sites (dots). Blue dots represent the sites of the $PM_{2.5}$ Speciation Trends Network (STN) and the State and Local Air Monitoring Stations (SLAMS), green dots represent the Interagency Monitoring of Protected Visual Environments (IMPROVE) sites, and gray dots represent the $PM_{2.5}$ Federal Reference Method (FRM) sites.

Figure 2. Monthly mean fractional bias (MFB) of $PM_{2.5}$ EC, OC, nitrate, ammonium, sulfate, and total mass. Solid lines represent the MFB criteria, and the blue dash lines represent the MFB goals.

Figure 3. Monthly mean fractional errors (MFE) of $PM_{2.5}$ EC, OC, nitrate, ammonium, sulfate, and total mass. Solid lines represent the MFE criteria, and the blue dash lines represent the MFE goals.

Figure 4. Mean fractional bias (MFB) and mean fractional errors (MFE) of PM and gaseous species when calculated using daily, monthly and annual averages.

Figure 5. Predicted (red lines) vs. observed (dark dots) monthly average O_3 (a),~~,~~ CO (b),~~,~~ NO (c),~~, ammonium, nitrate, OC, EC,~~ and $PM_{2.5}$ ammonium (d)~~total mass~~ at Sacramento, Fresno, Bakersfield, Los Angeles, and Riverside.

Figure 6. Predicted (red lines) vs. observed (dark dots) monthly average $PM_{2.5}$ nitrate (a), OC (b), EC (c), and $PM_{2.5}$ total mass (d) at Sacramento, Fresno, Bakersfield, Los Angeles, and Riverside.

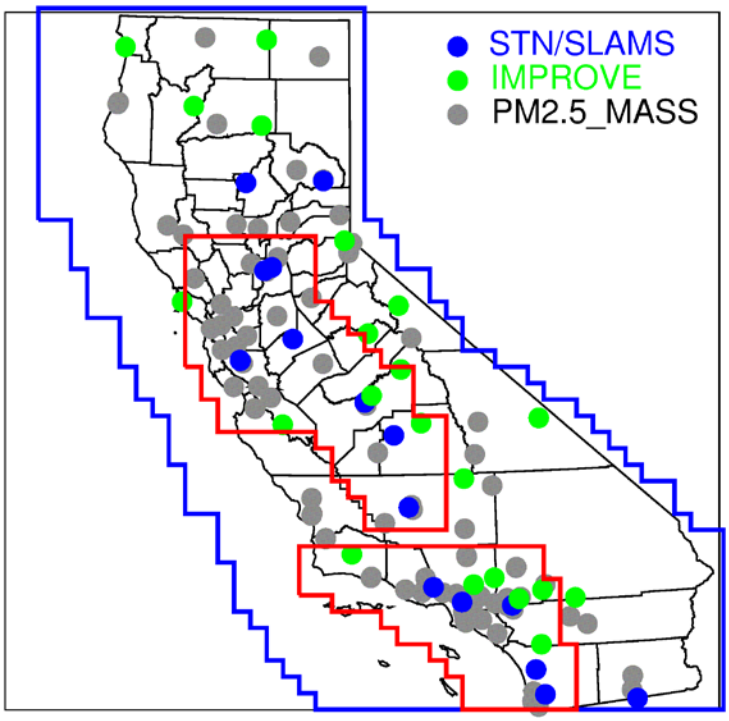
Figure 7.~~Figure 6.~~ Monthly average NO concentrations adjusted with the predicted/observed CO ratios. NO_noadj represents the NO concentrations in the UCD/CIT model predictions, and NO_adj represents the NO concentrations adjusted with observations as:
NO_adj = NO_noadj * CO_predicted / CO_measured

Figure ~~8~~7. Monthly average nitrate concentrations in2008 at Sacramento and Fresno predicted with perturbed relative humidity (RH+0.3), compared to the basecase nitrate predictions (RH_ori) and observed concentrations (Obs)

Figure ~~9~~8. Association between predicted PM concentration bias and wind bias vs. observed values. The observed PM concentrations and 1/u values on the x-axis are expressed in a relative scale of 0-100% of maximum range calculated as x (%) = $(C-C_{min})/(C_{max}-C_{min})*100$. Values for $[C_{min}, C_{max}]$ are listed in the concentration key. Bias between predicted vs. observed values is shown on the y-axis. Ideal behavior is bias of zero at all concentrations & wind speeds.

Figure ~~10~~9. Predicted (1) vs. measured (2) 9-year average $PM_{2.5}$ total mass (a), EC (b), OC (c), nitrate (d), sulfate (e), and ammonium (f) concentrations. The SoCAB_4km and SJV_4km results are overlayed on top of CA_24km results to create the model predicted spatial

992 distributions. Predicted and measured concentrations of the same species are in the same scale
993 showing in the panels of measurements
994



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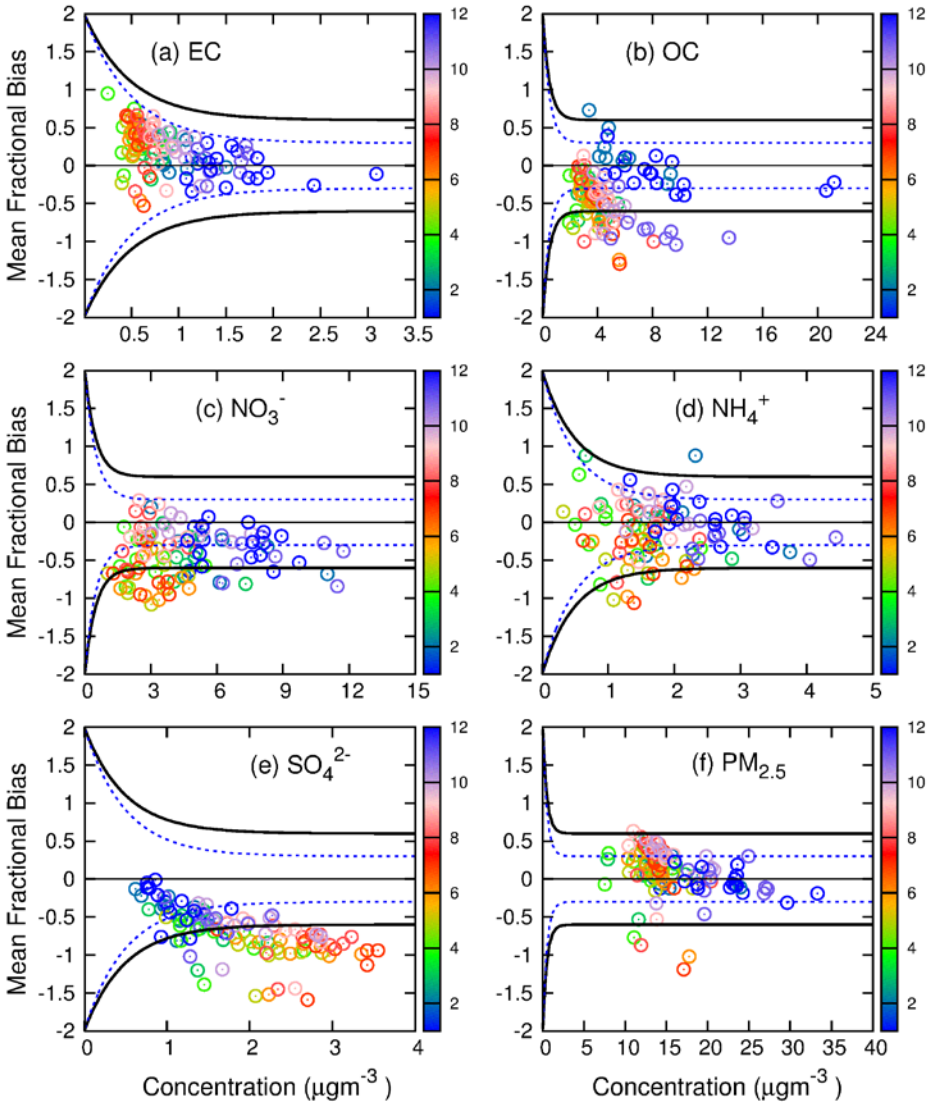

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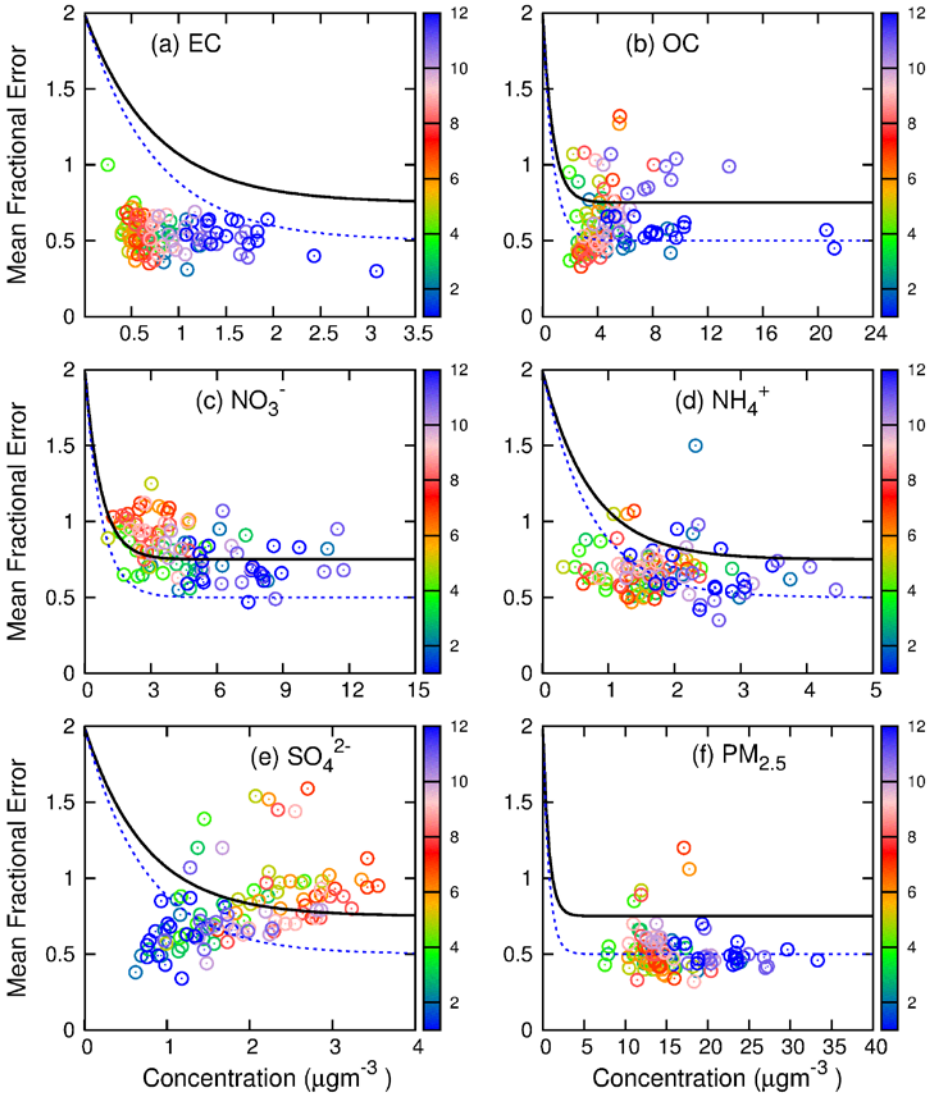


Figure 3. Monthly mean fractional errors (MFE) of $PM_{2.5}$ EC, OC, nitrate, ammonium, sulfate, and total mass. Solid lines represent the MFE criteria, and the blue dash lines represent the MFE goals.

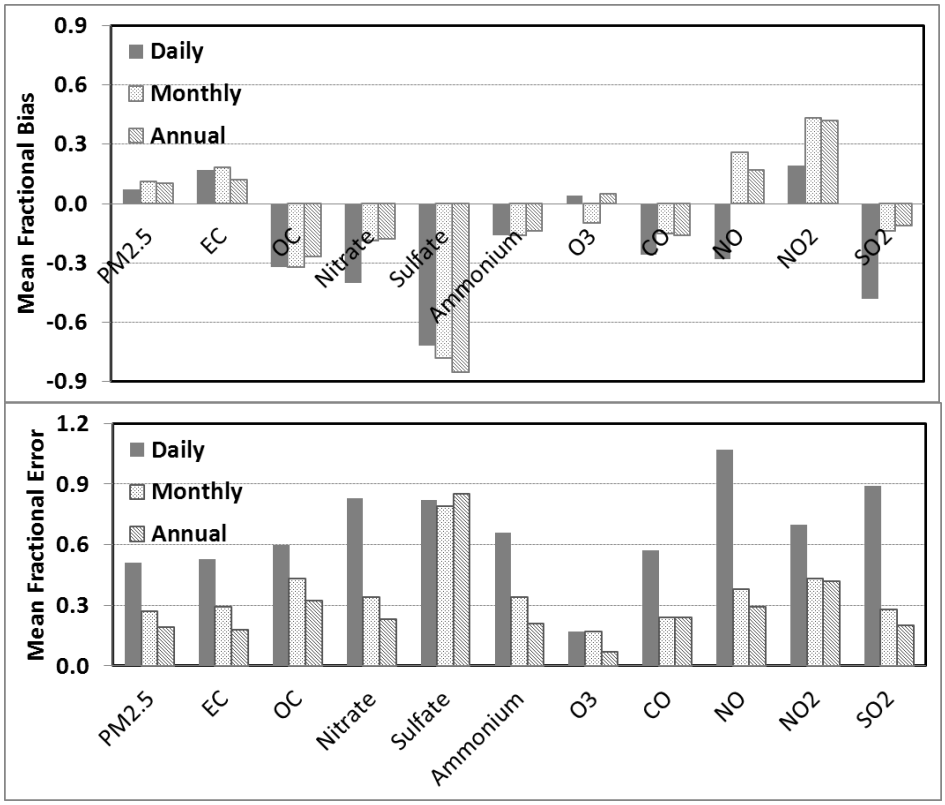
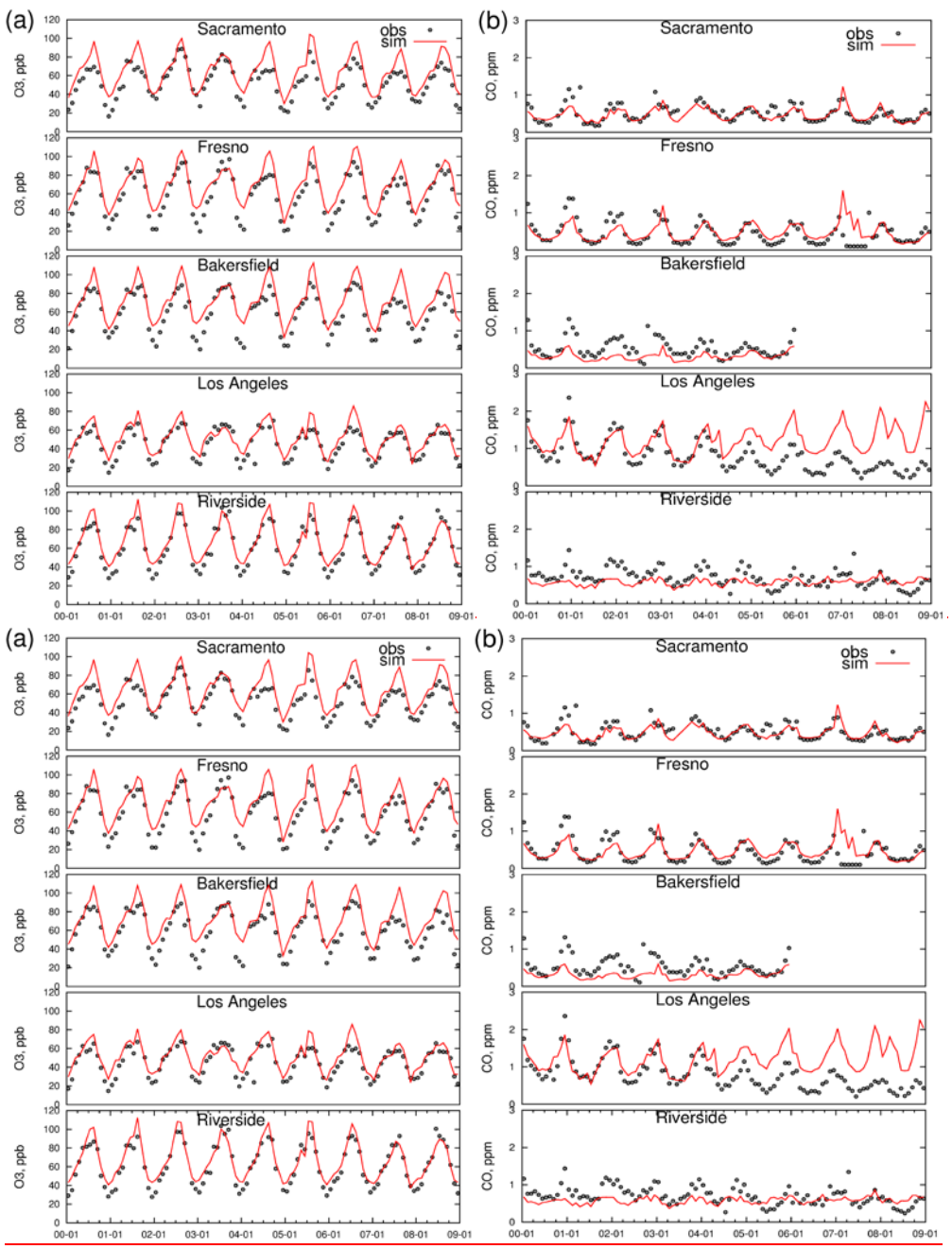


Figure 4. Mean fractional bias (MFB) and mean fractional errors (MFE) of PM and gaseous species when calculated using daily, monthly and annual averages.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Sacramento
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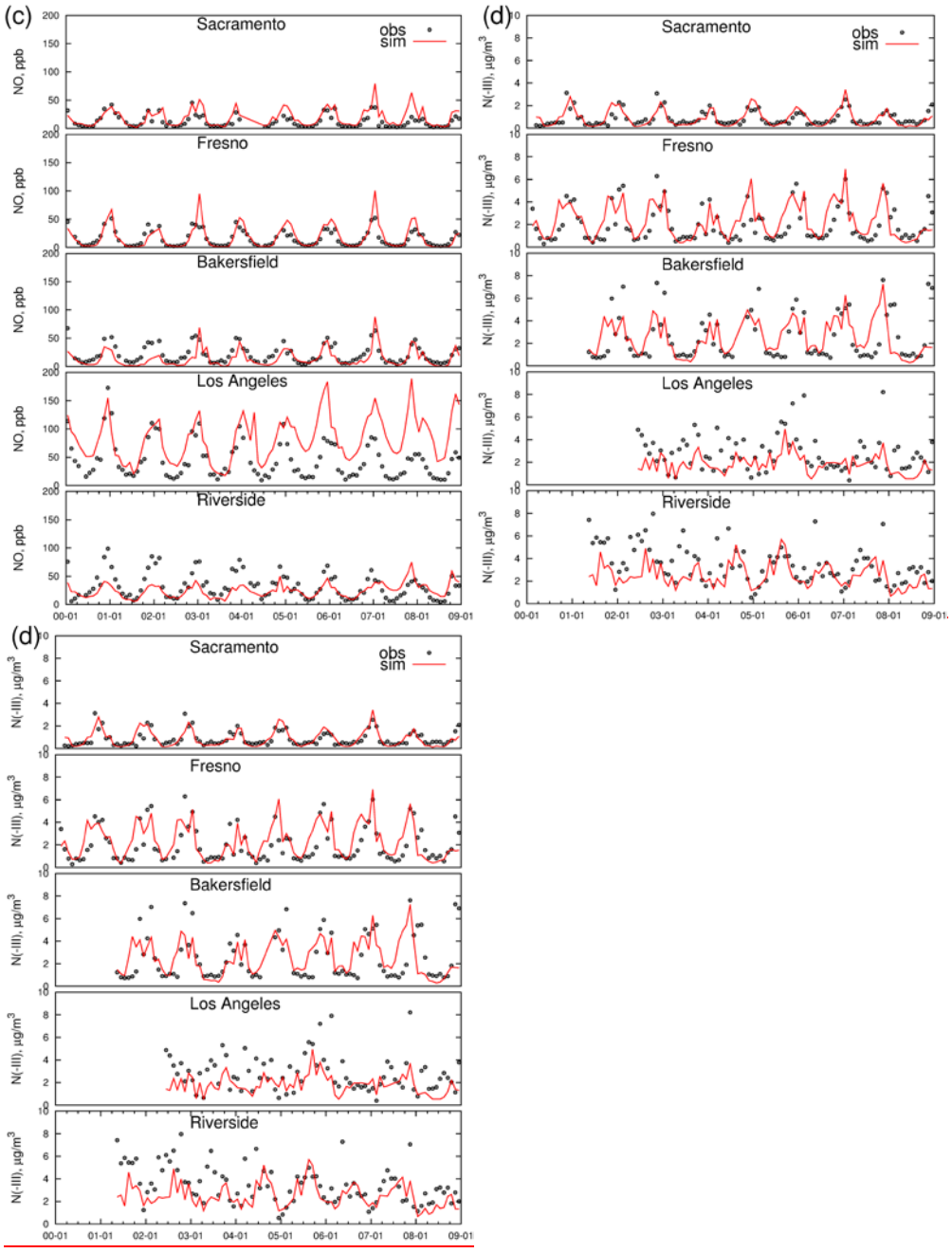
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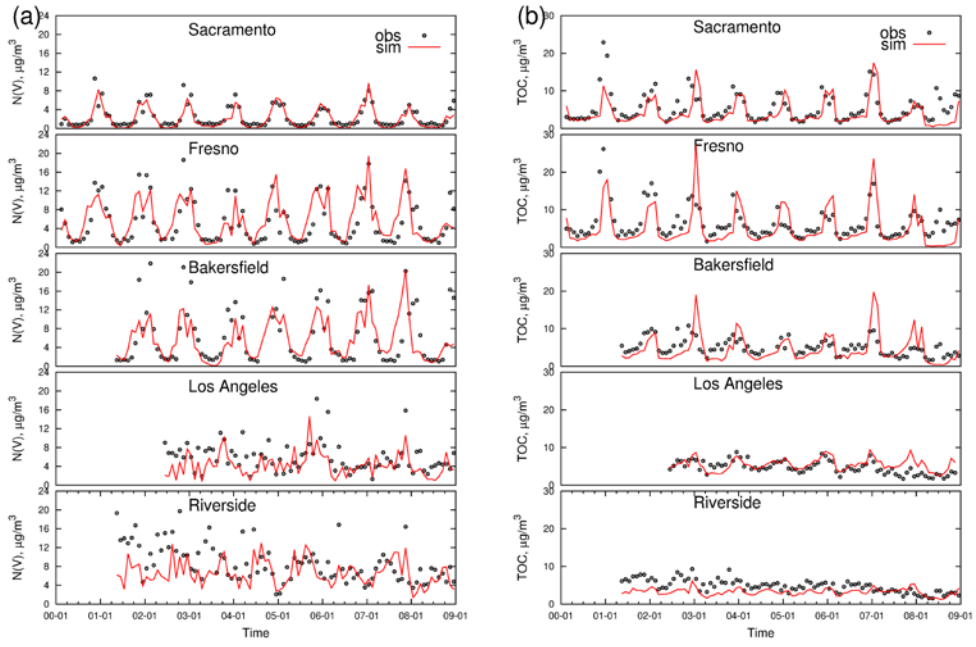
Figure 5. Predicted (red lines) vs. observed (dark dots) monthly average O_3 (a), ~~,~~ CO (b), ~~,~~ NO(c) ~~,~~ ~~ammonium, nitrate, OC, EC~~, and $PM_{2.5}$ ammonium (d)~~total mass~~ at Sacramento, Fresno, Bakersfield, Los Angeles, and Riverside.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TOC, μg/m3
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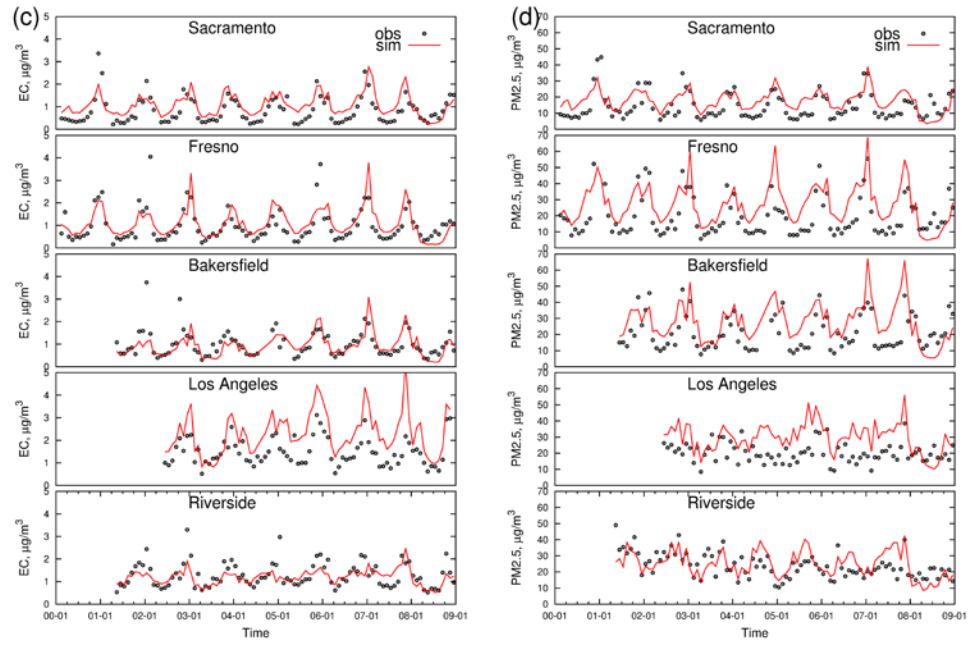
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EC, μg/m3
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PM2.5, μg/m3
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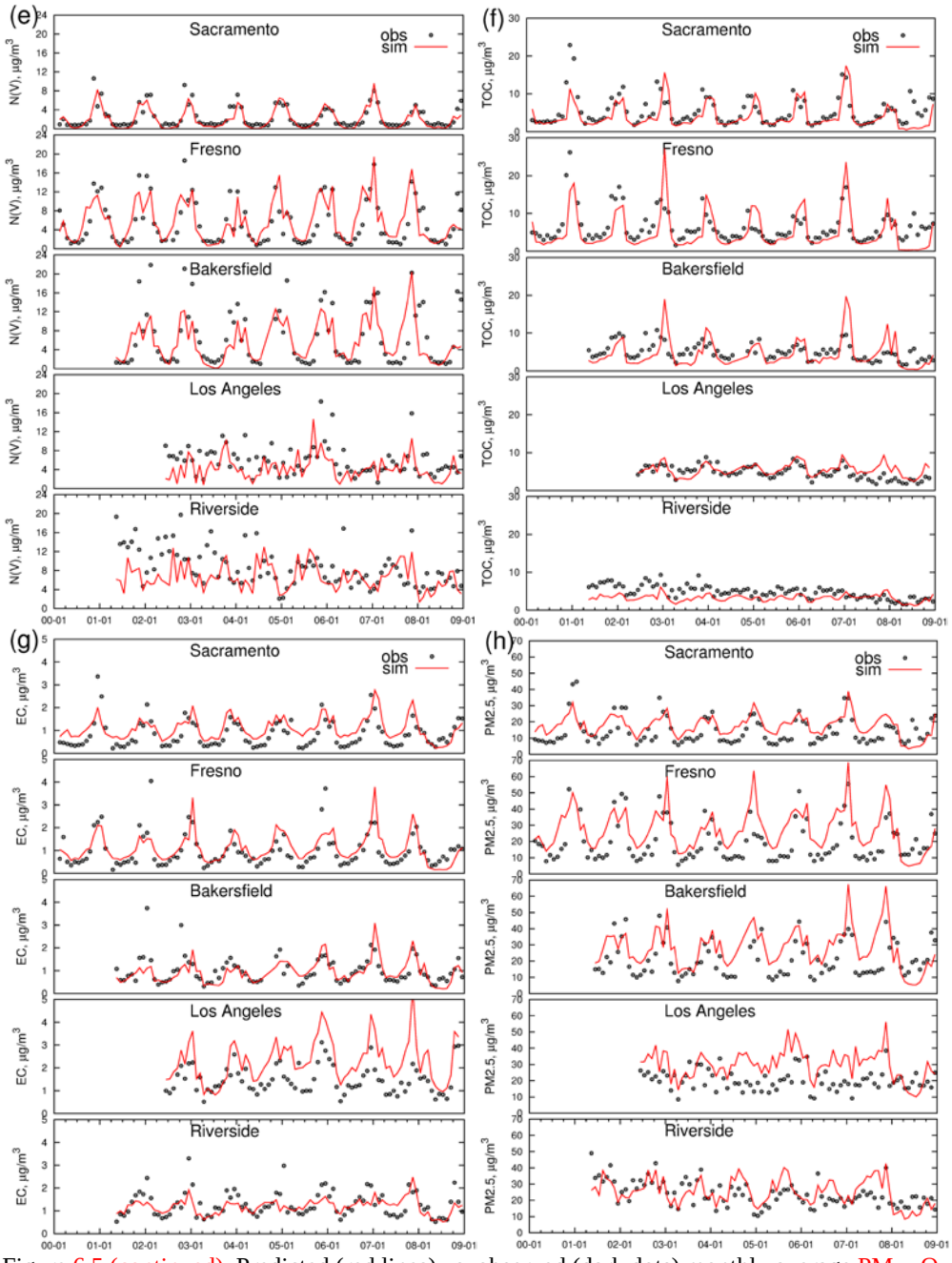
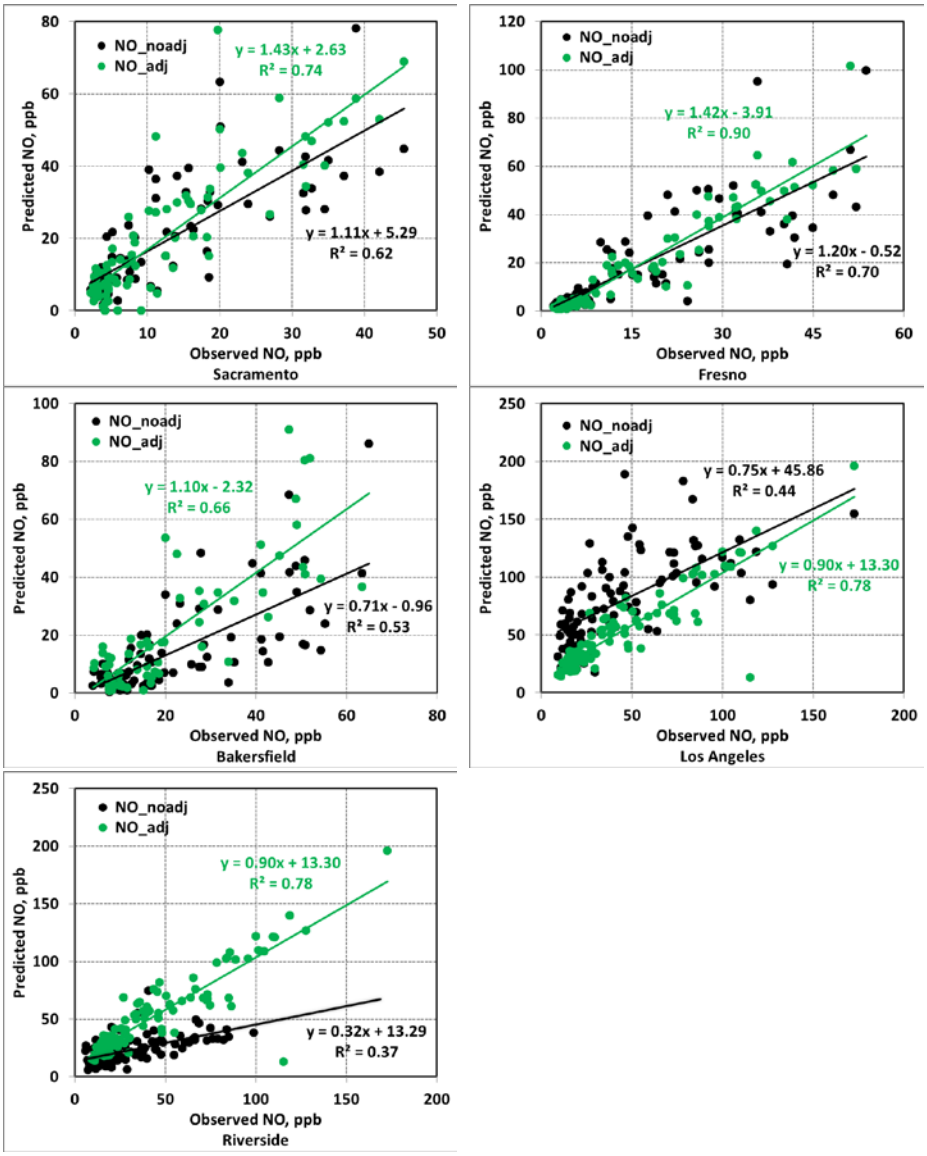


Figure 6.5 (continued). Predicted (red lines) vs. observed (dark dots) monthly average $PM_{2.5}$ ~~O_3, CO, NO, ammonium,~~ nitrate (a), OC (b), EC (c), and $PM_{2.5}$ total mass (d) at Sacramento, Fresno, Bakersfield, Los Angeles, and Riverside.



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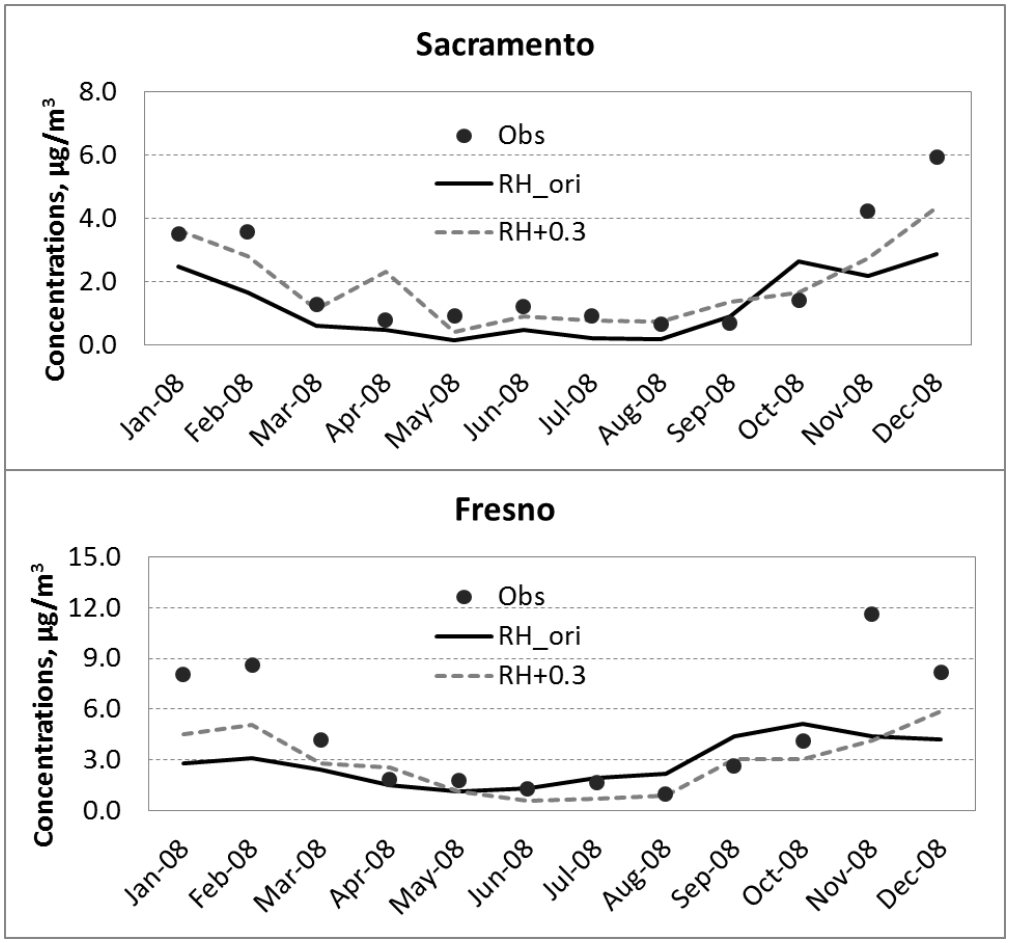

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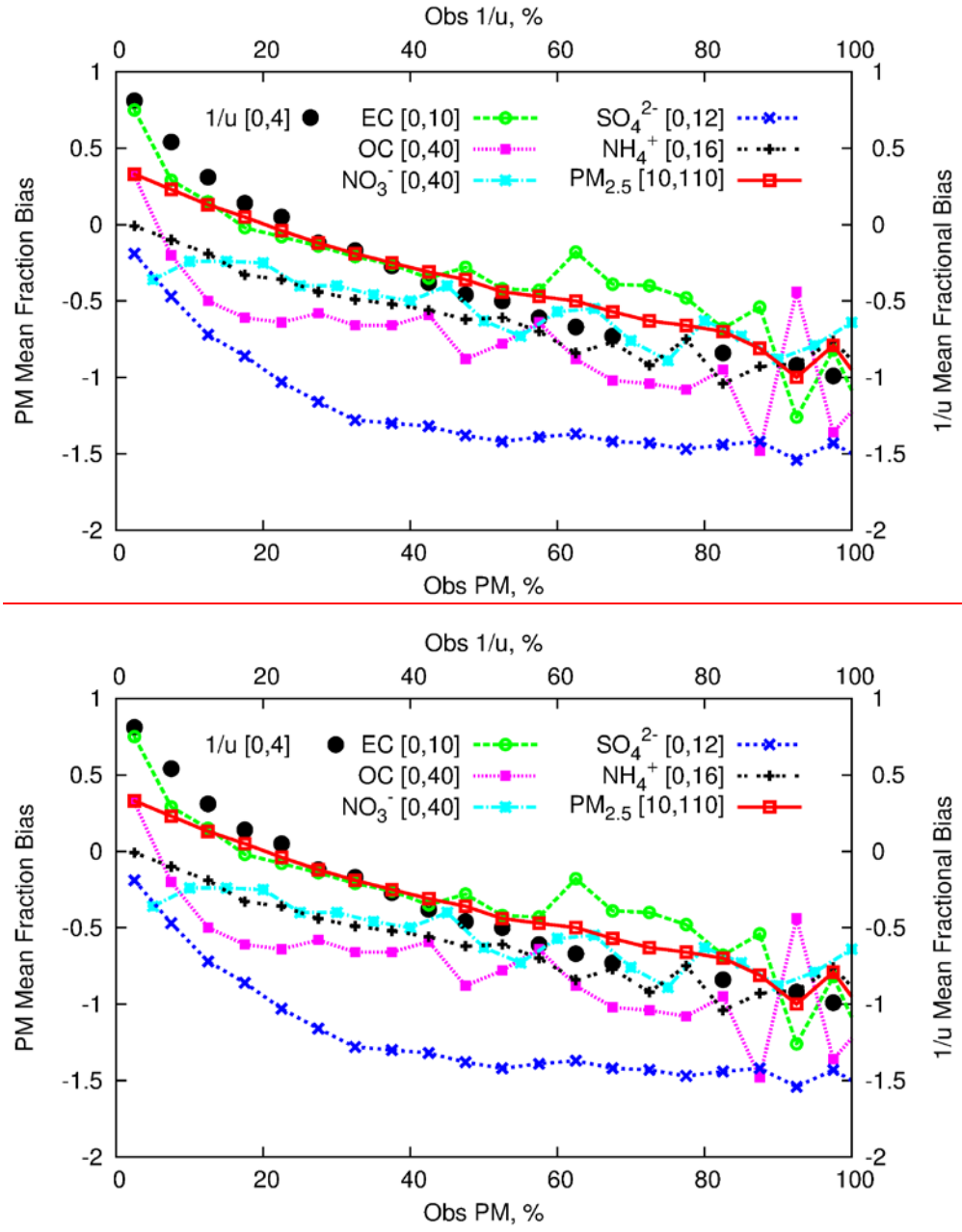

Figure 98. Association between predicted PM concentration bias and wind bias vs. observed values. The observed PM concentrations and 1/u values on the x-axis are expressed in a relative scale of 0-100% of maximum range calculated as x (%) = $(C-C_{min})/(C_{max}-C_{min})*100$. Values for

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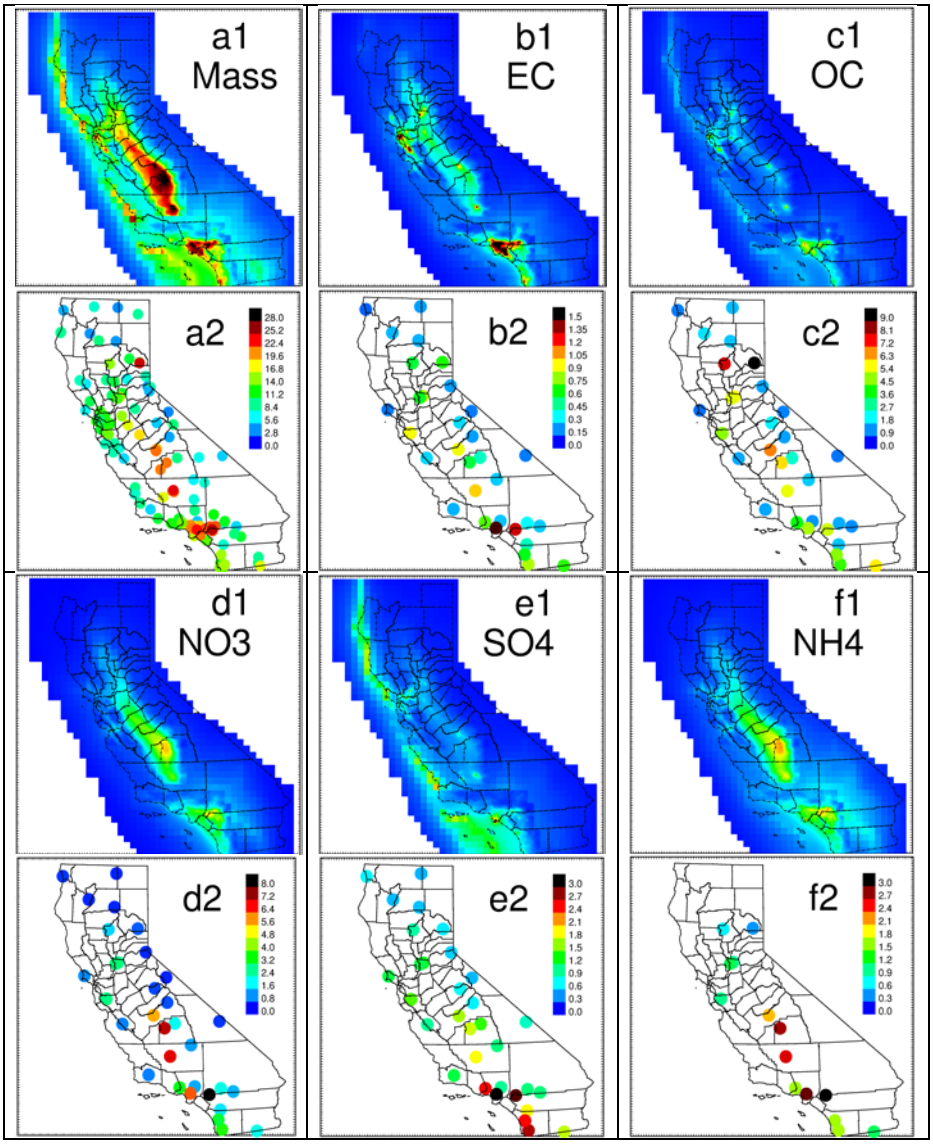

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