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**Climate impact on
airborne particulate
matter
concentrations**

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Climate impact on airborne particulate matter concentrations in California using seven year analysis periods

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Abstract

The effect of global climate change on the annual average concentration of fine particulate matter (PM_{2.5}) in California was studied using a climate – air quality modeling system composed of global through regional models. Output from the NCAR/DOE Parallel Climate Model (PCM) generated under the “business as usual” global emissions scenario was downscaled using the Weather Research and Forecasting (WRF) model followed by air quality simulations using the UCD/CIT airshed model. The air quality simulations were carried out for the entire state of California with a resolution of 8-km for the years 2000–2006 (present climate) and 2047–2053 (future climate). The 7-year windows were chosen to properly account for annual variability with the added benefit that the air quality predictions under the present climate could be compared to actual measurements. The climate – air quality modeling system successfully predicted the spatial pattern of present climate PM_{2.5} concentrations in California but the absolute magnitude of the annual average PM_{2.5} concentrations were under-predicted by ~35–40% in the major air basins. The majority of this under-prediction was caused by excess ventilation predicted by PCM-WRF that should be present to the same degree in the current and future time periods so that the net bias introduced into the comparison is minimized.

Surface temperature, relative humidity (RH), rain rate, and wind speed were predicted to increase in the future climate while the ultra violet (UV) radiation was predicted to decrease in major urban areas in the San Joaquin Valley (SJV) and South Coast Air Basin (SoCAB). These changes resulted in a ~0.6–1.9 $\mu\text{g m}^{-3}$ decrease in predicted PM_{2.5} concentrations in coastal and central Los Angeles. Annual average PM_{2.5} concentrations were predicted to increase at certain locations within the SJV and the Sacramento Valley due to the effects of climate change, but a corresponding analysis of the annual variability showed that these predictions are not statistically significant (i.e. the choice of a different 7-year period could produce a different outcome for these regions). Overall, virtually no region in California outside of coastal and cen-

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tral Los Angeles experienced a statistically significant change in annual average $PM_{2.5}$ concentrations due to the effects of climate change in the present study.

The present study employs the highest spatial resolution (8 km) and the longest analysis windows (7 years) of any climate-air quality analysis conducted for California to date, but the results still have some degree of uncertainty. Most significantly, GCM calculations have inherent uncertainty that is not fully represented in the current study since a single GCM was used as the starting point for all calculations. Ensembles of GCM results are usually employed to build confidence in climate calculations. The current results provide a first data-point for the climate-air quality analysis that simultaneously employ the fine spatial resolution and long time scales needed to capture the behavior of climate- $PM_{2.5}$ interactions in California. Future downscaling studies should follow up with a full ensemble of GCMs as their starting point.

1 Introduction

Exposure to elevated concentrations of airborne particles with aerodynamic diameter less than $2.5\ \mu\text{m}$ ($PM_{2.5}$) has serious health consequences (see for example Pope, 2000). The National Ambient Air Quality Standard for $PM_{2.5}$ is currently $15\ \mu\text{g m}^{-3}$ and the more stringent California State Air Quality Standard for $PM_{2.5}$ is currently $12\ \mu\text{g m}^{-3}$. Despite the adoption of a more stringent state standard, annual average $PM_{2.5}$ concentrations measured in California's San Joaquin Valley (SJV) ($21.5\ \mu\text{g m}^{-3}$) and South Coast Air Basin (SoCAB) ($19.7\ \mu\text{g m}^{-3}$) were the highest in the nation during the years 2007–2008. Some estimates predict that more than 18 000 California residents die prematurely each year due to air pollution (Tran et al., 2008). Meteorological parameters including temperature, precipitation, clouds, atmospheric water vapor, boundary layer height, wind speed, and wind direction influence the atmospheric chemistry and transport processes that determine $PM_{2.5}$ concentrations (see for example Kleeman, 2008; Aw and Kleeman, 2003; Sillman and Samson, 1995). Climate change will modify weather patterns in California with unknown consequences for $PM_{2.5}$ concentrations.

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The effect of global climate on regional PM_{2.5} concentrations can be studied using a dynamic downscaling approach where Global Climate Models (GCMs) provide initial and boundary conditions for Regional Climate Models (RCMs) with subsequent analysis using regional chemical transport models. California has several unique features that make this type of detailed analysis challenging. California's topographic features transition abruptly between mountains, valleys, deserts, lakes and oceans over distances of 10's of km. Emissions patterns in California also change quickly over small spatial scales due to the complex arrangement of residential, commercial, industrial, agricultural, and natural landuse types. Intricate meteorological features that strongly influence air pollution concentrations such as the land-sea breeze system and mountain-valley flows must also be modeled using high spatial resolution. Regional air quality studies in California are usually carried out at spatial scales finer than 10 km to resolve these features. Long time periods must also be considered when studying climate-air quality interactions in California. Natural annual variation in meteorology such as the El-Nino Southern Oscillation (ENSO) has a strong effect on air quality. The period of the ENSO cycle in recent decades has been 3–8 years suggesting that air quality analysis must be carried out over a similar time scale in order to be climatologically relevant. The combined need for fine spatial scales and long simulation times results in a computationally challenging analysis for climate-air quality interactions in California.

Several recent studies have examined the effects of global climate change on regional air quality across the entire United States including California (see for example Hogrefe et al., 2004; Tagaris et al., 2007; Jacobson, 2008; Avise et al., 2009; Chen et al., 2009; Dawson et al., 2009; Liao et al., 2009; Tagaris et al., 2009). The GCMs employed in the studies to date include the Goddard Institute for Space Studies (GISS) GCM (Russell et al., 1995), the NCAR/DOE Parallel Climate Model (PCM) (Washington et al., 2000), and the Stanford GATOR-GCMOM model (Jacobson, 2008). Most of the regional climate analysis was conducted using the fifth generation Penn State/NCAR Mesoscale Model (MM5) (Grell et al., 1994) although the GATOR-GCMOM model pro-

vides a unified framework from global to regional scales. Of those studies that compared $PM_{2.5}$ concentrations in present and future climates, Tagaris et al. (2009) simulated air quality for the entire United States using 36 km resolution for the years 2001 and 2050. The results of their study indicate that that the annual average $PM_{2.5}$ concentration in some parts of California is likely to decrease by $\sim 1.5 \mu g m^{-3}$ in the future due to the effects of climate change alone under the A1B emissions scenario. Avise et al. (2009) also conducted a climate – air quality study for the entire United States using 36 km resolution but they focused their analysis on a single month (July) in the years 1990–1999 and 2045–2054. Results were averaged within the administrative regions defined by the United States Environmental Protection Agency (USEPA). Avise et al. (2009) predicted that 24-hr average concentrations of $PM_{2.5}$ mass, $PM_{2.5}$ sulfate, $PM_{2.5}$ nitrate, and $PM_{2.5}$ ammonium for EPA’s Region 9 (containing California) would decrease by $\sim 0.4 \mu g m^{-3}$, $\sim 0.1 \mu g m^{-3}$, $\sim 0.1 \mu g m^{-3}$, $\sim 0.1 \mu g m^{-3}$, respectively, due to the effects of climate change alone. Dawson et al. (2009) modeled the climate effects on 24-h average total mass and speciated $PM_{2.5}$ concentrations for the eastern United States under the A2 emissions scenario. In this study, the authors used 36-km resolution and modeled January and July in 5 present years (1990s) and 5 future years (2050s) to report that on average the total mass and speciated $PM_{2.5}$ concentrations are likely to decrease in January due to increased precipitation and increase in July due to decreased ventilation in the future.

None of the previous studies for the entire United States have simultaneously used spatial resolution finer than 10 km and analysis times longer than 3 years to determine how climate change will influence the annual average $PM_{2.5}$ concentration in California. The current study applies a dynamic downscaling approach with 8-km spatial resolution during 7 years of current climate and 7 years of future climate to address this question. The 7-year analysis period during the current climate is long enough to allow for a meaningful comparison between simulated vs. measured $PM_{2.5}$ concentrations. To the best of our knowledge, this comparison is the most rigorous evaluation of a climate – air quality modeling system ever conducted. The 7-year analysis windows also contain

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enough data to support a statistical analysis of annual variability so that confidence intervals can be calculated for the differences between present vs. future conditions. The results of the current study reduce the uncertainty of likely climate impacts on annual-average PM_{2.5} concentrations in California at the regional scale.

2 Methods

Figure 1 shows a schematic diagram of the regional climate – air quality modeling system employed in this study. The regional climate is simulated by dynamically downscaling output from the Parallel Climate Model (PCM) using the Weather Research and Forecasting (WRF) model, and the air quality is simulated using the latest generation of the UCD/CIT air quality model for the entire state of California. A detailed description of the modeling system is provided below.

PCM is a Global Climate Model (GCM) that was developed at the National Center for Atmospheric Research (NCAR) (Washington et al., 2000). The parent PCM dataset utilized in this study was available in 2.8° (~240-km) × 2.8° (~310-km) horizontal resolution with 18 hybrid vertical layers from ground to ~4 hPa. PCM output was obtained for a 7-year period in the present-day climate (2000–2006) and a 7-year period in the future climate (2047–2053) under the “business as usual” (B06.44) global emissions scenario. Seven year windows were selected to account for the effects of El Nino Southern Oscillation (ENSO) events and the intra-annual variability in the climate data that would have different implications in the final air quality results (see for example, Aw and Kleeman, 2003). The PCM output was processed through the WRF Preprocessing System (WPS) in preparation for regional downscaling.

Version 2.2 of the WRF model (Skamarock, 2004) was utilized to generate the regional climate data in this study. All of the WRF simulations were carried out using an optimized configuration of the modeling system for California (Zhan et al., 2010). The optimization process tested six different configurations of WRF for conditions in central California. The best agreement with measured meteorological parameters was pro-

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duced by the Yonsei University (YSU) planetary boundary layer (PBL) scheme (Hong, Noh and Dudhia, 2006), Grell-Devenyi cumulus parameterization scheme (Grell and Devenyi, 2002), WRF single-momentum 6-class (WSM6) microphysics scheme (Hong, Dudhia and Chen, 2004), Rapid Radiation Transfer Model (RRTM) long-wave scheme (Mlawer et al., 1997), and Dudhia short-wave radiation scheme (Dudhia, 1989). The WRF simulations used a three-domain 1-way nesting technique, where the coarse grid output provided initial and lateral boundary conditions for the fine grid simulations with horizontal resolutions of 36-km, 12-km, and 4-km, respectively. Meteorological fields with 4-km resolution were generated for a domain with 264×264 grid cells in the horizontal plane and 10-vertical layers extending to a height of 5000 m above the ground. The heights of the vertical layers in WRF were matched with those of the air-quality model such that the model used finer vertical resolutions near the surface. All of the variables were saved on the A-grid (i.e. in the center of a grid-cell) except for the u- and v-components of wind speed, which were staggered using the Arakawa C-grid (i.e. on the face center of each grid-cell). The native C-grid output was retained for the wind field in order to achieve better accuracy in the air quality transport calculations. The gridded WRF output was averaged over each 60-min simulation period for every simulated day. A total of nine periods were simulated for each year that each consisted of 17-days followed by 25 days that were not simulated. The first simulation day of each year was set to be 1 January. This approach captures an unbiased sample of predicted meteorology and air quality across the full seven year period that reflects both seasonal variation and annual variability while still keeping the analysis time to a reasonable level.

Finer grid resolution provides a better description of small-scale meteorological dynamics such as sea-land breezes and orographical winds during regional meteorological modeling, but this resolution does not necessarily need to be retained in subsequent regional air quality modeling exercises. In the present study, the 4-km WRF meteorological fields were extracted and averaged to 8-km resolution to generate inputs for the air quality modeling. The coarser resolution of the air quality model reflects a balance

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between accuracy and speed. Each 7-year study period involved 1008 simulation days over a domain composed of $131 \times 128 \times 10$ grid cells requiring approximately 45 days of real time to complete. Ying et al. (2008) showed that increasing the resolution of simulations in the Central Valley of California from 8-km to 4-km does not significantly improve accuracy but it increases the computational effort by a factor of 4. A previous study carried out in Marseilles-Fos-Berre in the south of France has also found that beyond a certain point finer grid resolution does not necessarily yield better air quality predictions (Menut, Coll and Cautenet, 2005). The South Coast Air Quality Management District (SCAQMD) provides emissions inventories with 5-km spatial resolution for the SoCAB and numerous previous air quality studies have been carried out at this scale (Kleeman and Cass, 2001; Griffin et al., 2002; Martien and Harley, 2006) without the perceived need to improve resolution to 4-km. The requirement for uniform grid spacing across the entire state of California combined with the balance between efficiency vs. accuracy produced the compromise of 8-km grid resolution during the air quality modeling carried out in this study.

The initial conditions (ICs) of gas-phase and particle-phase species were generated following the work of (Ying et al., 2008 and Kleeman et al., 2007). Table 1 summarizes the domain average IC concentrations of major model species used in the current study as a function of season. The IC concentrations can be quantified within the UCD/CIT framework through the use of internal tracers. It was found that the influence of IC concentrations on air quality chemistry diminishes by the end of first 3–4 days in the current study. As a result, predicted concentrations from the first 4-days of each simulation period were disregarded in the calculations of periodic/annual average particulate matter (PM) concentrations. The boundary conditions (BCs) of various gas-phase and particle-phase species were adjusted for seasonal variability (Liang et al., 1998; Jaffe et al., 2005). The scaling for vertical boundary conditions was obtained from Ying et al. (2008), and boundary conditions at the top of the model domain were interpolated from the lateral boundary conditions in the top model layer. Table 2 summarizes the average BC concentrations of selected model species along the West Coast as used

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in the current study as a function of season.

The base-case raw emissions inventories for the year 2000 were obtained from the California Air Resources Board (CARB) and the SCAQMD. Area source and point source emissions were used without modification in all simulations. On-road mobile source emissions were adjusted for the variation of meteorological conditions experienced during each simulation using CARB's Emissions Factors (EMFAC) model. EMFAC produces mobile emissions summaries for different pollutants and technology classes in 69 geographical locations spanning the entire state of California. EMFAC is also able to predict how temperature and humidity influence mobile source emissions of nitrogen oxides (NO_x) and reactive hydrocarbons (RHCs). EMFAC results were generated for the base-case year 2000 emissions inventory and for the conditions predicted to occur within each of the 69 geographical regions at each simulated hour. The base-case year 2000 mobile source emissions were then scaled by the ratio of $\text{EMFAC}^{\text{actual}}/\text{EMFAC}^{\text{base-case}}$ for each pollutant of interest in each grid cell of the model domain. WRF predictions for hourly-averaged temperature and surface shortwave radiation in each grid cell were also combined with the MODIS satellite data and fed into CARB's BEIGIS model to predict episode-specific biogenic volatile organic compound emissions. The adjusted mobile source emissions and the newly generated biogenic emissions were then processed with the remaining area and point sources to provide a source oriented model-ready gridded hourly emissions inventory.

The UCD/CIT air quality model employed in this study is based on the original CIT 3-D photochemical model (Mcrae et al., 1982; Harley et al., 1993) with updates to provide a source-oriented framework (see for example, (Kleeman et al., 1997; Kleeman et al., 2001; Mysliwicz and Kleeman, 2002; Ying and Kleeman, 2003; Held et al., 2004; Ying and Kleeman, 2006; Ying et al., 2008). Only those aspects of the UCD/CIT model that were updated during the current study are discussed here.

Previous studies (Byun, 1999; Odman and Russell, 2000; Lee et al., 2004; Hu et al., 2006; Sportisse et al., 2007) have found mass consistency errors when the meteorological model and the chemistry-transport model do not use the same grid sys-

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tem, the same interpolation strategies and/or the same transport algorithms. (Hu et al., 2006) proved the existence of the mass consistency errors while using the MM5/CMAQ modeling system and recommended an effective way to overcome the problem by recalculating the vertical wind velocity using the horizontal wind components and atmospheric density generated by the meteorology model. Calculation of vertical winds was therefore incorporated into the UCD/CIT modeling framework in the current study. The UCD/CIT model was also updated so that it could work with either prognostic wind fields (C-grid) or diagnostic wind fields (A-grid).

The computational burden of simulating air quality in a large domain over 2016 days motivated revisions to the approach for gas-particle transfer of inorganic species in the UCD/CIT model. The original approach for gas-particle conversion involved fully dynamic transfer of both acidic and basic species between the gas and particle phase, requiring many thousands of calls to the thermodynamics routines during each integration time step within each grid cell. The fully dynamic approach for gas-particle conversion was replaced by the approach proposed by (Jacobson, 2005) whereby acid gases are treated as fully dynamic species while ammonia is considered to be in equilibrium between the gas and particle phases. The original thermodynamic routines were based on the Aerosol Inorganic Module (AIM) developed by (Wexler and Seinfeld, 1992) with updates to correct coding errors and impose minimum water constraints to keep the solution in the continuum region (Clegg et al., 2008). The modified approach used in the present study replaces the AIM thermodynamics module with ISORROPIA II (Nenes et al., 1998; Fountoukis and Nenes, 2007) to calculate the vapor pressure of semi-volatile inorganic species above each particle surface. The combination of these changes increased the speed of the model calculations by roughly a factor of two with less than 10% difference in the final predicted concentrations of gas-phase and particle-phase species.

Wet deposition calculations were added to the UCD/CIT model so that it could correctly predict removal rates for gases and particles during rain events. The rain drop size distribution was calculated as a function of total precipitation rate (Cerro et al.,

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1997) followed by a calculation of the pollutant washout rate (Loosmore and Cederwall, 2004).

The original version of the UCD/CIT model relied on independently generated emissions inventories for sea salt over ocean grid cells. The input data and correlations used to calculate sea salt emissions are relatively simple, and so the sea salt emissions algorithm was embedded directly within the UCD/CIT model in the present study. Sea salt emissions over the open ocean (Gong, 2003) and sea salt emissions in the surf zone (de Leeuw et al., 2000) were both predicted by this new module based on the surface wind speed.

3 Results

Airborne particulate matter concentrations in California are strongly influenced by temperature, relative humidity, UV radiation, wind speed, and mixing height (Aw et al., 2003; Kleeman, 2008). It is instructive to examine the climate-induced changes to these meteorological variables over 7 year windows to gain greater insight into the processes that influence $PM_{2.5}$ concentrations. Figure 2 shows the differences between the future (2047–2053) and present-day (2000–2006) averages for ground-level temperature, humidity, wind speed, precipitation, mixing height, and UV radiation. Figure 2a shows that the surface air temperature is predicted to increase by 1.0–1.7 °C over the Pacific Ocean west of California and increase by 0.2 to 1.2 °C over California inland regions in the future years compared to the present-day. The predicted increase is slightly greater (~ 1.0 °C) in large urban areas such as Los Angeles, Bakersfield, Fresno, San Francisco and Sacramento compared to rural and mountainous areas (~ 0.2 – 0.8 °C).

The prediction of larger temperature increases over the ocean than over land is caused by large scale features of the PCM simulation that drive the WRF model during the downscaling exercise. Future PCM predictions for air temperature 2 m above the surface increase more over the Pacific Ocean than over the California land mass dur-

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ing the months of December, January, and February. Much of this change is driven by the sea-surface temperature inherited from the PCM simulations and by an increased frequency of precipitation events over land during the winter months. These PCM temperature and precipitation patterns are consistent with the results produced by the Community Climate System Model (CCSM) operated under the SRES A2 emissions scenario. Although some GCMs may predict greater future temperature increase over land than over the ocean during winter months, the results of the PCM simulations used in the current study are a legitimate example of a relatively modern simulation of climate that contributes to the ensemble of results that have been produced by the scientific community to date. In contrast to the winter trends, both PCM and CCSM predict that air temperatures over land increase more than air temperatures over water during the summer months. Hotter temperatures generally encourage lower PM_{2.5} concentrations in California through the evaporation of ammonium nitrate aerosol (Kleeman, 2008). A downward bias in future surface temperature over land would produce an upward bias in predicted future PM_{2.5} concentrations.

The predicted change in future (2047–2053 vs. 2000–2006) relative humidity (RH) at the surface is shown in Fig. 2b. RH is predicted to decrease by ~0.10–2.2% over the ocean as the air temperature warms because the amount of additional water vapor evaporated from the ocean surface does not keep pace with the additional capacity of the warmer atmosphere to hold water vapor. RH is predicted to increase by ~0.20–4.0% over land as the moist air cools by 0.5–0.8 °C reducing its capacity to hold water vapor. The SoCAB is predicted to experience the greatest increase in RH by as much as 4% compared to only ~0.2–0.7% in the SJV in the future. Increasing RH encourages the partitioning of ammonium nitrate to the particle phase (Kleeman, 2008). Any upward bias in predicted RH would once again bias PM_{2.5} concentrations upwards.

Wind speed and mixing height are two of the most critical meteorological parameters for air pollution calculations since they determine the ventilation of fresh emissions away from the surface. Air pollution episodes in California are generally characterized by a stagnation period (low ventilation) lasting more than 2-days. Figure 2c shows that

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the future wind speed (2047–2053) is predicted to increase by as much as $\sim 0.5 \text{ ms}^{-1}$ around Los Angeles in the SoCAB, and decrease by $\sim 0.1\text{--}0.2 \text{ ms}^{-1}$ in some parts of the SJV, and the Sacramento Valley air basins relative to current years (2000–2006). These changes are important since average wind speed in these air basins are approximately 2.7 ms^{-1} . The mixing height (planetary boundary layer) shown in Fig. 2d is predicted to increase by $\sim 40 \text{ m}$ in the coastal areas but decrease by $\sim 20 \text{ m}$ in the Sierra Nevada mountain range. The average future mixing height is predicted to increase by $\sim 6 \text{ m}$ in the SJV and by $\sim 20 \text{ m}$ in the lower part of the SoCAB. All of these changes are small compared to the base-case average mixing height of $\sim 600 \text{ m}$ and so they have little effect on pollutant concentrations. The major driver for changes to future air pollution concentrations associated with ventilation is the changes to wind speed illustrated in Fig. 2c.

Figure 2e shows that the average future (2047–2053) precipitation rate is predicted to increase by $\sim 0.3 \text{ mm hr}^{-1}$ in the Sierras and some areas in the SoCAB relative to current years (2000–2006). Precipitation in the SJV is predicted to increase by $\sim 0.01\text{--}0.04 \text{ mm hr}^{-1}$ in the future ($\sim 80\%$ increase over current levels). The increased rate reflects the increased humidity in the atmosphere resulting in greater precipitation rates during rain events and a general lengthening of the rainy season. Future SJV precipitation rates increased by $\sim 40\%$ in the current rainy season (DJF) but increased by $\sim 240\%$ in the months before and after the traditional rainy season (September, October, March, May). Increased precipitation enhances the wet deposition of pollutants leading to reduced atmospheric concentrations.

Figure 2f illustrates that future (2047–2053) UV radiation is predicted to decrease by $\sim 1.0\text{--}1.3\%$ in the Sierras, and by $\sim 0.5\text{--}0.6\%$ in the SJV and SoCAB. UV flux decreases as cloud cover increases and so the trend in UV radiation is consistent with the pattern of precipitation rates in the future, i.e. the UV radiation is likely to decrease as precipitation rates increase. UV radiation drives photochemical chemical reactions in the atmosphere leading to increased concentrations of secondary particulate matter. Reduced UV will reduce the production of secondary PM. Further details of the effects

of climate on air pollution meteorology in California are discussed by Zhan et al. (2010).

Figure 3a shows the present-day (2000–2006) annual average concentrations of $PM_{2.5}$ mass for the entire state of California. Higher $PM_{2.5}$ concentrations ($> \sim 20 \mu g m^{-3}$) are predicted around the urban centers such as Los Angeles, Bakersfield, Fresno, San Francisco and Sacramento because primary particulate emissions sources are concentrated in these regions. Elevated total $PM_{2.5}$ concentrations ($> \sim 10 \mu g m^{-3}$) are also predicted along the major transportation routes in the SJV and SoCAB.

Figures 3b–f show the contribution to $PM_{2.5}$ total mass from elemental carbon (EC), organic carbon (OC), nitrate (N(V)), ammonium (N(-III)) and sulfate (S(VI)) concentrations, respectively. Primary particulate matter components such as EC and OC account for approximately $\sim 60\%$ of the total $PM_{2.5}$ mass concentrations in California during the present-day (2000–2006) simulations. Maximum annual average EC concentrations of $\sim 0.5\text{--}1.0 \mu g m^{-3}$ are found in the major urban areas and along the major transportation routes as shown in Fig. 3b. The highest EC concentration of $\sim 1.9 \mu g m^{-3}$ was predicted around the San Francisco Bay Area under the present-day climate.

Predicted annual average OC concentrations are also higher around the major sources of primary emissions in the domain (Fig. 3c). Maximum annual average OC concentrations in the range of $\sim 4.0\text{--}10.0 \mu g m^{-3}$ are predicted in the San Joaquin and Sacramento Valleys near major urban areas, while predicted OC concentrations in the SoCAB are in the range of $\sim 3.0\text{--}6.0 \mu g m^{-3}$.

Figure 3d shows the predicted annual average concentrations of $PM_{2.5}$ nitrate in California. Nitrate accounts for $\sim 14\%$ of the annual average $PM_{2.5}$ mass over broad segments of the state. Predicted nitrate concentrations range from $\sim 1.2\text{--}2.6 \mu g m^{-3}$ in the SJV to $\sim 0.7\text{--}1.9 \mu g m^{-3}$ in the SoCAB. The majority of the particulate nitrate is neutralized by ammonium ion and so it is not surprising that the pattern of annual-average ammonium ion concentration (Fig. 3e) appears similar to the pattern of nitrate concentration. The highest annual average nitrate concentration ($\sim 2.6 \mu g m^{-3}$) and ammonium ion concentration ($\sim 1.6 \mu g m^{-3}$) were both predicted in the region between Fresno and

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Bakersfield where gas-phase ammonia emissions from agricultural sources are highest. Predicted ammonium nitrate concentrations had a strong seasonal dependence with concentrations in the winter season (DJF) ~ 3 times larger than the summer (JJA).

Predicted annual average concentrations of sulfate (S(VI)) were $< 1.0 \mu\text{g m}^{-3}$ in the SJV but as high as $1.0\text{--}3.9 \mu\text{g m}^{-3}$ in the SoCAB. Higher annual average concentrations of sulfate ($\sim 2.0 \mu\text{g m}^{-3}$) were also found along the coastal areas of the SoCAB. This spatial pattern gives strong clues that shipping emissions are the dominant source of sulfate aerosol in the present day simulations.

Climate-air quality predictions are often presented with little verification that the base-case modeling system is able to reproduce present-day air quality concentrations over multi-year periods. Climate-air quality modeling systems that use GCM inputs down-scaled to regional scales cannot resolve individual air pollution events but the multi-year average concentrations predicted by these systems should ideally match the multi-year average measurements in each region of interest. Previous studies have evaluated the performance of the UCD/CIT 3-D photochemical airshed model driven by observed meteorological fields (Ying et al., 2008). The present study is the first to evaluate model performance over a climatologically relevant time-frame (7-year averages) using prognostic meteorology produced by a GCM.

Figure 4 shows comparisons between the predicted and observed concentrations of primary and secondary $\text{PM}_{2.5}$ components averaged between the years 2000–2006 at representative locations in California’s most heavily populated air basins. The Central Los Angeles (CELA) site is located in the SoCAB. The Modesto (M14), Visalia (VCS), Fresno (FSF) and San Jose (SJ4) sites are situated in the greater SJV, and the Sacramento (S13) site is in the Sacramento Valley air basin. For each site, 24-h average measured concentrations of the $\text{PM}_{2.5}$ total mass and the major chemical components that contribute to $\text{PM}_{2.5}$ mass were obtained from CARB. The corresponding predicted concentrations were then extracted for the present-day (2000–2006) air quality simulations. Daily average concentrations were used to calculate mean values and standard deviations over the 7-year period. Panels (a–c) in Fig. 4 show the comparisons

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between the predicted and observed concentrations of $PM_{2.5}$ total mass, $PM_{2.5}$ elemental carbon (EC), and $PM_{2.5}$ organic carbon (OC). Annual average $PM_{2.5}$ total mass concentrations of $>20.0 \pm 2.0 \mu\text{g m}^{-3}$ were measured in both the SoCAB and the SJV. Predicted maximum $PM_{2.5}$ total mass concentrations ($\sim 12\text{--}13 \mu\text{g m}^{-3}$) were 35–40% lower than measured values, with consistent under-predictions at all sites. Model performance was relatively better at San Jose (SJ4) and Sacramento (S13) compared to other sites. In general, the variation between measured $PM_{2.5}$ concentrations at different sites is greater than the variation between predicted $PM_{2.5}$ concentrations at those same locations (compare the height of the average bars between locations in Fig. 4a). In contrast, the time variation of the predicted $PM_{2.5}$ concentrations at the same site can be greater than, equivalent to, or less than the time variation of measured $PM_{2.5}$ concentrations at that same site (compare the size of the uncertainty bars at each location in Fig. 4a).

Figure 4b illustrates that present-day (2000–2006) predictions for EC concentrations are 20–70% lower than measured values at all sites except for Sacramento, where the predictions are 40% greater than the measurements. EC is a primary pollutant and so wide-spread under-predictions suggest a systematic bias in emission rates, removal rates, and/or ventilation (wind speed \times mixing height). The fundamental equations and input data files used to represent emissions and removal rates have been validated during numerous retrospective modeling studies that used diagnostic wind fields (Held et al., 2005; Ying et al., 2008) and so it seems likely that the ventilation rates predicted by the prognostic PCM-WRF meteorological fields are slightly over-predicted. Recent studies confirm that prognostic meteorological fields provide greater ventilation than diagnostic meteorological fields in central California (Hu et al., 2009). A zero-order approximation of the air pollution system would predict that concentration (C) ($\mu\text{g m}^{-3}$) = emissions rate (E) ($\mu\text{g hr}^{-1}$) / ventilation rate (V) ($\text{m}^3 \text{hr}^{-1}$). From this simplified relationship we can derive that the relative change in concentration is inversely proportional to the relative change in ventilation ($\Delta C/C = -\Delta V/V$). The concentration under-prediction at each location could therefore be corrected with a change in aver-

age surface wind speed of $\sim 0.15 \text{ m s}^{-1}$ at the Sacramento (S13) site to $\sim 1.98 \text{ m s}^{-1}$ at the Modesto (M13) site.

Panel (c) shows the measured and predicted annual average concentrations of OC at the six representative sites chosen for model evaluation. Measured annual average OC concentrations ranged from $5.5\text{--}8 \mu\text{g m}^{-3}$ while predicted OC concentrations range from $3\text{--}5.4 \mu\text{g m}^{-3}$. In short, OC concentrations were under-estimated by $\sim 2\text{--}60\%$ at all sites except for San Jose (SJ4) where predictions match observations within $\sim 2\%$. OC can either be emitted directly from sources (primary) or it can be produced in the atmosphere through precursor reactions that form products with low saturation vapor pressure (SOA). In the present study, $\sim 99\%$ of the annual average OC concentrations was attributed to primary organic compounds with the remaining $\sim 1\%$ attributed to SOA formation. Recent studies suggest that primary organic aerosol is semi-volatile (Huffman et al., 2009) implying that SOA formation dominates total OC formation but major questions remain unanswered concerning the exact composition and formation mechanism of this material. Mechanisms that depend on high concentrations of intermediate-volatility precursor compounds have been proposed (Presto et al., 2009), and even implemented in regional calculations (Robinson et al., 2007), but the intermediate volatility framework characterizes all compounds solely by their saturation vapor pressure. It therefore requires numerous parameterizations for critical elements (rate at which compounds transition between volatility bins, temperature dependence of partitioning, etc.) that (to date) are based on simplistic laboratory experiments. Much work remains to be done using additional laboratory experiments and retrospective field studies supported by comprehensive measurements to constrain these critical parameters before the intermediate volatility framework can be used confidently for climate – air quality predictions. More chemically complete models for SOA formation that include calculations for activity coefficients in aqueous and organic phases (Kleeman et al., 2007) have been shown to form roughly the same amount of SOA as more simplistic two product absorption models (Ying et al., 2007). The more computationally efficient two-product model was used in the current study, with the caveat that it does not completely

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describe all aspects of SOA formation.

Panels (d–f) show the measured and predicted annual average concentrations of $\text{PM}_{2.5}$ nitrate, ammonium, and sulfate, respectively. The plots show that the model under-predicts the concentrations of secondary species at all the chosen monitoring sites by $\sim 42\text{--}71\%$. The majority of this under-prediction is likely caused by the excess ventilation produced by the PCM-WRF meteorological predictions (see discussion for Fig. 4b) exacerbated by the non-linear chemical system for the formation of secondary PM (Aw et al. 2003). The pattern of variation between sites appears to be similar for the measured and predicted concentrations of secondary PM since model predictions are able to identify the sites with the highest and lowest concentrations of each secondary pollutant.

Natural annual and intra-annual variability makes it extremely difficult to quantify climate impacts on airborne particulate matter concentrations in California. Panels in Fig. 5 show the differences between predicted annual average $\text{PM}_{2.5}$ concentrations and their 7-year averages at Riverside, central Los Angeles, Bakersfield, and Fresno over the periods 2000–2006 and 2047–2053. The relative standard deviations (standard deviation divided by the mean) are 8%, 6%, 11% and 11% for the present (2000–2006), and 10%, 8%, 13% and 7% for the future (2047–2053) periods at Riverside, central Los Angeles, Bakersfield, and Fresno, respectively. This annual variability in $\text{PM}_{2.5}$ mass concentrations is mainly caused by normal variation in yearly meteorology combined with ENSO effects operating on a $\sim 3\text{--}8$ year cycle. One of the key objectives in this study was to capture this annual variability both in the present (2000–2006) and future (2047–2053) air quality simulations so that the effects of global change can be understood over a climatologically relevant period.

The differences between the future (2047–2053) and present-day (2000–2006) annual average concentrations of $\text{PM}_{2.5}$ total mass, EC and OC are illustrated in Fig. 6. Panels (a, c, e) display the difference between the 7-year averages while panels (b, d, f) display the significance of this result expressed as the p-value. The p-value quantifies the likelihood that present day (2000–2006) concentrations and future (2047–2053)

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concentrations are equal. The p-value is calculated using information about the mean and variance within each 7-year window. Small p-values are produced when large differences exist between present and future concentrations while the annual variability of concentrations within the present and future analysis windows is small. The 7-year analysis windows used in the current study are long by atmospheric chemical modeling standards but relatively short by statistical standards and so p-values were calculated using the student T distribution with 6 degrees of freedom rather than the Normal distribution.

Figure 6a shows that the future annual average $PM_{2.5}$ mass concentration is predicted to increase by $\sim 0.2\text{--}0.5\ \mu\text{g m}^{-3}$ in the Sacramento Valley, and by $\sim 0.7\ \mu\text{g m}^{-3}$ in some areas in the SJV. The corresponding p-values in panel (b) are ≥ 0.5 indicating that these changes are small relative to the annual variability in the predicted signal. In fact, the model calculations do not predict significant changes (at the 95% confidence level) in the $PM_{2.5}$ concentrations anywhere in California except in a small region around coastal and central Los Angeles where $PM_{2.5}$ concentrations are predicted to decrease by $\sim 0.6\text{--}1.9\ \mu\text{g m}^{-3}$. The corresponding p-value plot (panel b) indicates that these changes are large relative to the annual variability. As discussed previously, the majority of this decreased concentration stems from increased wind speeds in the SoCAB providing greater ventilation of primary emissions. The increased wind speed is predicted largely because of the synoptic features of the GCM inputs that were used in the analysis. GCM calculations have a certain degree of inherent uncertainty and so ensembles of GCM calculations are usually employed to build confidence in the results of climate calculations. The present study provides a first data-point for a GCM-WRF-air quality model calculation of climate impacts on air quality that should be followed up with a full ensemble of downscaling runs using multiple GCMs as their starting point.

In general, the major features of the $PM_{2.5}$ mass analysis illustrated in Fig. 6a, b are echoed for $PM_{2.5}$ EC in Fig. 6c, d and for $PM_{2.5}$ OC in Fig. 6e, f. The only significant trends at the 95% confidence level are localized in a sub-region of Los Angeles where concentrations generally decrease due to the effects of increased ventilation. Future

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(2047–2053) annual average EC concentrations are predicted to decrease by $\sim 0.02\text{--}0.09\ \mu\text{g m}^{-3}$ while future annual average OC concentrations are predicted to decrease by $\sim 0.5\text{--}0.6\ \mu\text{g m}^{-3}$ in coastal and central Los Angeles relative to current conditions (2000–2006).

5 Figure 7 shows the predicted change in annual average concentrations for the secondary $\text{PM}_{2.5}$ species and their corresponding p-values using the same format as Fig. 6. Patterns for nitrate and ammonium ion (panels a–d) are similar with 95% confidence achieved in the SoCAB and in the band surrounding the southern portion of the SJV and extending along the Pacific coast between LA and San Francisco. $\text{PM}_{2.5}$ nitrate concentrations are predicted to decrease in both of these regions by up to $0.4\ \mu\text{g m}^{-3}$ with a corresponding decrease of $0.2\ \mu\text{g m}^{-3}$ in ammonium ion concentrations. The only significant change in predicted $\text{PM}_{2.5}$ sulfate concentrations at the 10 95% confidence level is a decrease of $0.1\ \mu\text{g m}^{-3}$ around the port of Los Angeles.

4 Summary and conclusions

15 The effect of global climate change on annual average $\text{PM}_{2.5}$ mass concentrations in California has been studied using seven year analysis periods that allowed for meaningful statistical analysis of the results. A regional climate-air quality modeling system was employed that downscaled PCM output using the WRF model followed by emissions generation and air quality simulations using the UCD/CIT photochemical model 20 to predict $\text{PM}_{2.5}$ concentrations. Simulations were carried out for the present-day climate (2000–2006) and the future (2047–2053) under the “business as usual” global emissions scenario. Air pollutant emissions in California were held at their nominal year 2000 levels to directly examine the effect of climate on regional air quality.

25 The present-day results from the climate-air quality modeling system successfully predicted the spatial pattern of high $\text{PM}_{2.5}$ concentrations in California but the absolute magnitude of the annual average $\text{PM}_{2.5}$ concentrations were under-predicted by $\sim 35\text{--}40\%$ at different sites in the major air basins. Of the total $\text{PM}_{2.5}$ mass, primary PM concentrations were only under-estimated by 2–70% while secondary PM con-

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centrations were under-estimated by ~42–71%. The majority of this under-prediction is likely caused by the excess ventilation produced by the PCM-WRF meteorological predictions that should be consistent in both present and future climate so that a minimum amount of net bias is introduced into the comparison. The present study is the first to evaluate climate – air quality model performance over a climatologically relevant time-frame (7-year averages) using prognostic meteorology produced by a GCM. It is likely that other modeling systems would have similar performance features if they were checked against observations.

Future predictions indicate that changes to global climate lead to changes in key regional meteorological parameters that affect airborne particulate matter concentrations in California. An analysis of the major meteorological variables shows that the future surface temperature, relative humidity (RH), rain rate, and wind speed are predicted to increase while the UV radiation is predicted to decrease in major urban areas in the SJV and SoCAB. These changes lead to a predicted $\sim 0.6\text{--}1.9\ \mu\text{g m}^{-3}$ decrease in $\text{PM}_{2.5}$ concentrations in coastal and central Los Angeles, but no other statistically significant changes to $\text{PM}_{2.5}$ concentrations were predicted in other parts of California (at the 95% confidence level). The natural annual variability in the $\text{PM}_{2.5}$ predictions were larger than the changes induced by climate. This result contradicts previous studies that have predicted decreased $\text{PM}_{2.5}$ concentrations throughout California in response to climate change. Those previous studies used coarser spatial resolution and shorter averaging times that made it difficult to fully analyze geographical variation and annual variability in California. The 8 km spatial resolution combined with the 7 year analysis periods used in the current study provide the most comprehensive picture to date about climate effects on $\text{PM}_{2.5}$ concentrations in California.

GCM calculations have a certain degree of inherent uncertainty and so ensembles of GCM calculations are usually employed to build confidence in the results of climate calculations. The present study provides a first data-point for a GCM-WRF-air quality model calculation of climate impacts on air quality in California that should be followed up with a full ensemble of downscaling runs using multiple GCMs as their starting point.

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Table 1. Domain-averaged initial concentrations of major model gas and particle species at the surface.

Species	Unit	Domain average concentration			
		Winter (DJF)	Spring (MAM)	Summer (JJA)	Fall (SON)
Carbon monoxide (CO)	ppm	0.2823	0.2823	0.2823	0.2823
Carbon dioxide (CO ₂)	ppm	332.00	332.00	332.00	332.00
Sulfur dioxide (SO ₂)	ppm	0.0020	0.0020	0.0020	0.0020
Nitrogen dioxide (NO ₂)	ppm	0.0046	0.0046	0.0046	0.0046
Nitric oxide (NO)	ppm	0.0033	0.0033	0.0033	0.0033
Reactive hydro carbon (RHC)	ppm	0.0513	0.0513	0.0513	0.0513
Ozone (O ₃)	ppm	0.0401	0.0401	0.0401	0.0401
Nitric acid (HNO ₃)	ppm	0.0010	0.0010	0.0010	0.0010
Ammonia (NH ₃)	ppm	0.0029	0.0029	0.0029	0.0029
Hydrochloric acid (HCl)	ppm	0.0010	0.0010	0.0010	0.0010
Formaldehyde (HCHO)	ppm	0.0000	0.0000	0.0000	0.0000
Acetaldehyde (CH ₃ CHO)	ppm	0.0000	0.0000	0.0000	0.0000
Aldehydes (RCHO)	ppm	0.0000	0.0000	0.0000	0.0000
Acetone (ACET)	ppm	0.0006	0.0006	0.0006	0.0006
Methyl ethyl ketone (MEK)	ppm	0.0000	0.0000	0.0000	0.0000
Methyl glyoxile (MGLY)	ppm	0.0000	0.0000	0.0000	0.0000
Glyoxile (GLY)	ppm	0.0000	0.0000	0.0000	0.0000
Peroxi acetyl nitrate (PAN)	ppm	0.0003	0.0008	0.0008	0.0005
Hydrogen peroxide (H ₂ O ₂)	ppm	0.005	0.005	0.005	0.005
Isoprene (ISO)	ppm	0.0003	0.0003	0.0003	0.0003
Elemental carbon (EC)	µg m ⁻³	0.18	0.18	0.18	0.18
Organic carbon (OC)	µg m ⁻³	0.66	0.66	0.66	0.66
Nitrate (N(V))	µg m ⁻³	0.36	0.36	0.36	0.36
Sulfate (S(VI))	µg m ⁻³	0.45	0.45	0.45	0.45
Chloride (Cl(-1))	µg m ⁻³	0.19	0.19	0.19	0.19
Sodium (NA(1))	µg m ⁻³	0.21	0.21	0.21	0.21
Ammonium (N(-III))	µg m ⁻³	0.24	0.24	0.24	0.24
OTHER	µg m ⁻³	1.62	1.6	1.6	1.60
H2OPART	µg m ⁻³	0.891	0.89	0.89	0.89

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Table 2. Average boundary concentrations of various model gas and particle species concentrations along the western boundary of the modeling domain.

Species	Unit	Domain average concentration			
		Winter (DJF)	Spring (MAM)	Summer (JJA)	Fall (SON)
Carbon monoxide (CO)	ppm	0.4000	0.4000	0.4000	0.4000
Carbon dioxide (CO ₂)	ppm	332.0000	332.0000	332.0000	332.0000
Sulfur dioxide (SO ₂)	ppm	0.0010	0.0010	0.0010	0.0010
Nitrogen dioxide (NO ₂)	ppm	0.0010	0.0004	0.0003	0.0004
Nitric oxide (NO)	ppm	0.0010	0.0004	0.0003	0.0004
Reactive hydro carbon (RHC)	ppm	0.0069	0.0069	0.0069	0.0069
Ozone (O ₃)	ppm	0.0350	0.0400	0.0400	0.0350
Nitric acid (HNO ₃)	ppm	0.0007	0.0007	0.0007	0.0004
Ammonia (NH ₃)	ppm	0.0025	0.0025	0.0025	0.0025
Formaldehyde (HCHO)	ppm	0.0015	0.0015	0.0015	0.0015
Acetaldehyde (CH ₃ CHO)	ppm	0.0020	0.0020	0.0020	0.0020
Aldehydes (RCHO)	ppm	0.0014	0.0014	0.0014	0.0014
Acetone (ACET)	ppm	0.0015	0.0015	0.0015	0.0015
Glyoxile (GLY)	ppm	0.0002	0.0002	0.0002	0.0002
Peroxi acetyl nitrate (PAN)	ppm	0.0005	0.0005	0.0005	0.0003
Isoprene (ISO)	ppm	0.0000	0.0000	0.0000	0.000
Elemental carbon (EC)	µg m ⁻³	0.01	0.01	0.01	0.01
Organic carbon (OC)	µg m ⁻³	0.02	0.02	0.02	0.02
Nitrate (N(V))	µg m ⁻³	0.04	0.03	0.03	0.03
Sulfate (S(VI))	µg m ⁻³	0.05	0.05	0.05	0.05
Chloride (Cl(-1))	µg m ⁻³	0.06	0.06	0.06	0.06
Sodium (NA(1))	µg m ⁻³	0.06	0.06	0.06	0.06
Ammonium (N(-III))	µg m ⁻³	0.01	0.01	0.01	0.01
OTHER	µg m ⁻³	0.03	0.03	0.03	0.03
H2OPART	µg m ⁻³	1.47	0.35	1.09	0.65

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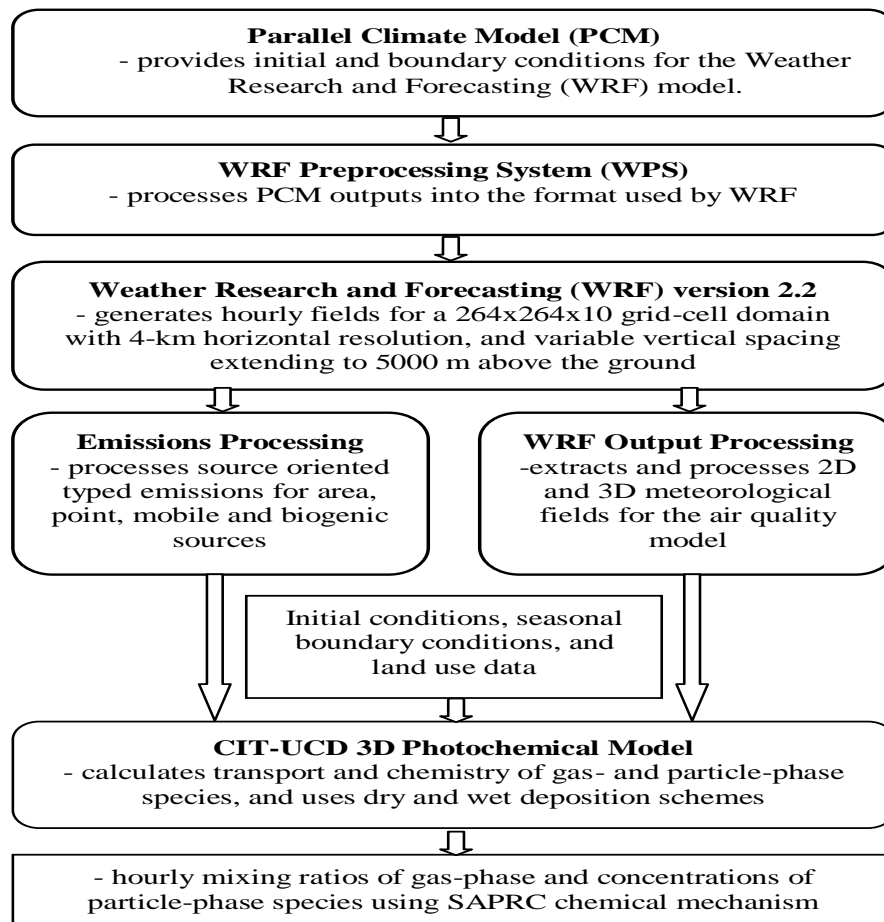
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**Fig. 1.** Flow Chart of the climate downscaling and air quality modeling systems.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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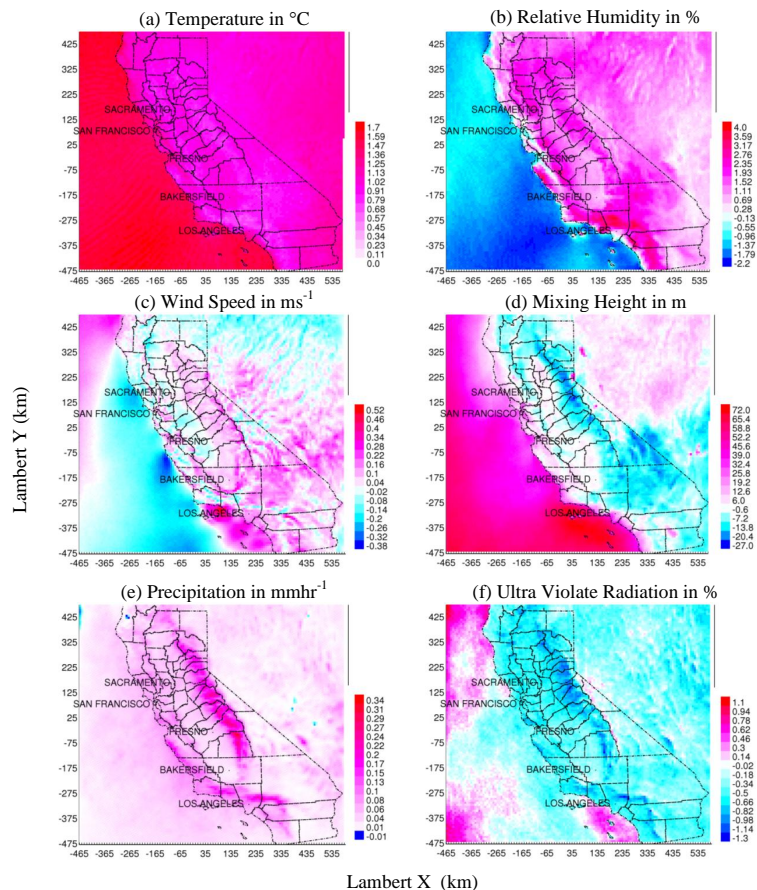


Fig. 2. (a–f) show changes in annual average values in meteorological parameters likely to occur due to climate change in the future (2047–2053) from the present-day (2000–2006).

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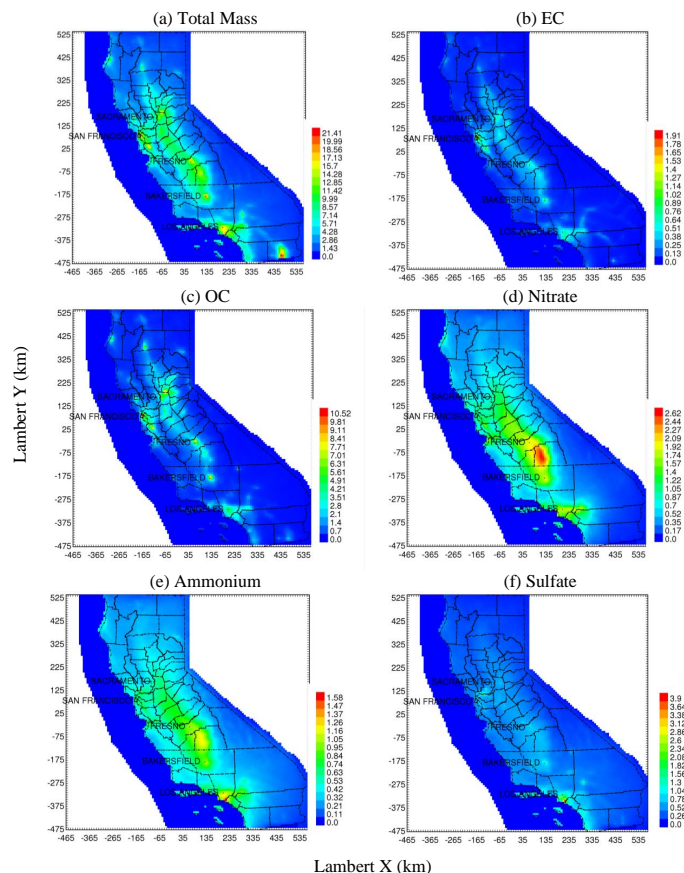


Fig. 3. Annual average Particulate Matter 2.5 (PM_{2.5}) concentrations in CA for the present-day (2000–2006) **(a)** total mass, **(b)** elemental carbon, **(c)** organic carbon, **(d)** nitrate, **(e)** ammonium, and **(f)** sulfate.

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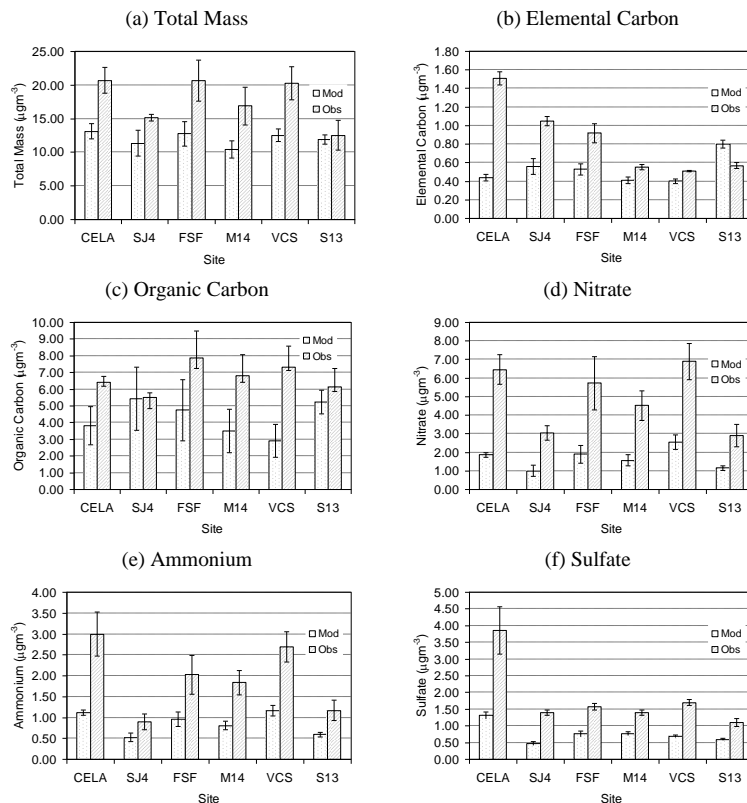


Fig. 4. Comparison between modeled and observed 7-year average PM_{2.5} (a) total mass, (b) elemental carbon, (c) organic carbon, (d) nitrate, (e) ammonium, and (f) sulfate concentrations at different sites in California: (1) CELA-central Los Angeles North Main St, (2) SJ4-San Jose 4th St, (3) FSF-Fresno 1st St, (4) M14-Modesto 14th St, (5) VCS-Visalia Church St, and (6) S13-Sacramento T St.

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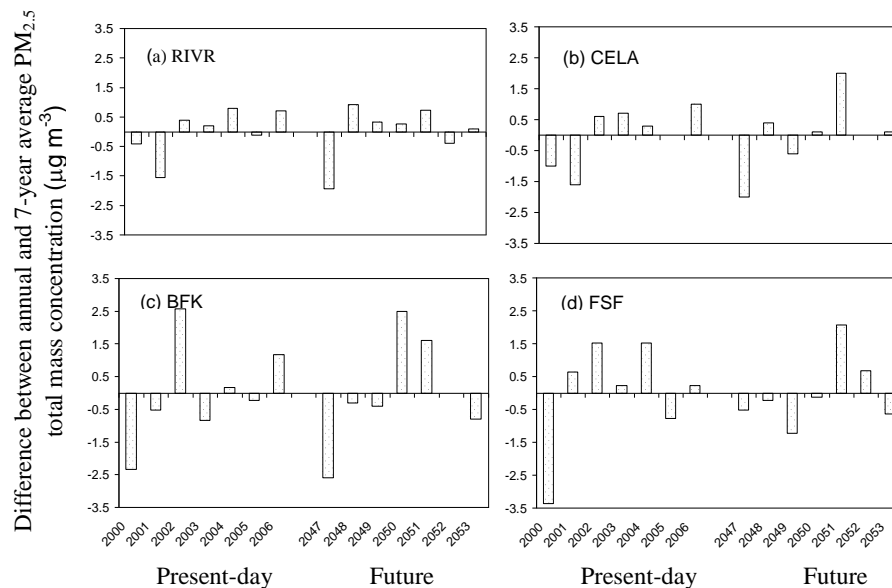


Fig. 5. Predicted variations around the 7-year average $PM_{2.5}$ total mass concentration at four sites in California: **(a)** Riverside-Rubidoux (RIVR), **(b)** Central Los Angeles (CELA), **(c)** Bakersfield (BFK), and **(d)** Fresno (FSF).

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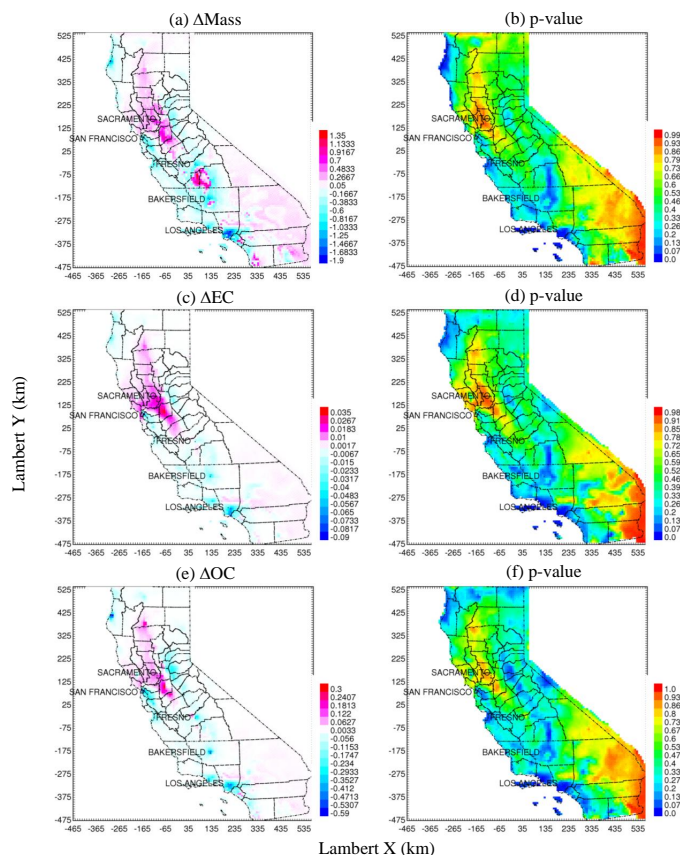


Fig. 6. Changes in annual average total mass and primary $\text{PM}_{2.5}$ concentrations, and their corresponding p-values in CA likely to occur in the future (2047–2053) due to climate change from the present-day (2000–2006). The p-value quantifies the likelihood that average future concentrations are equal to present day concentrations.

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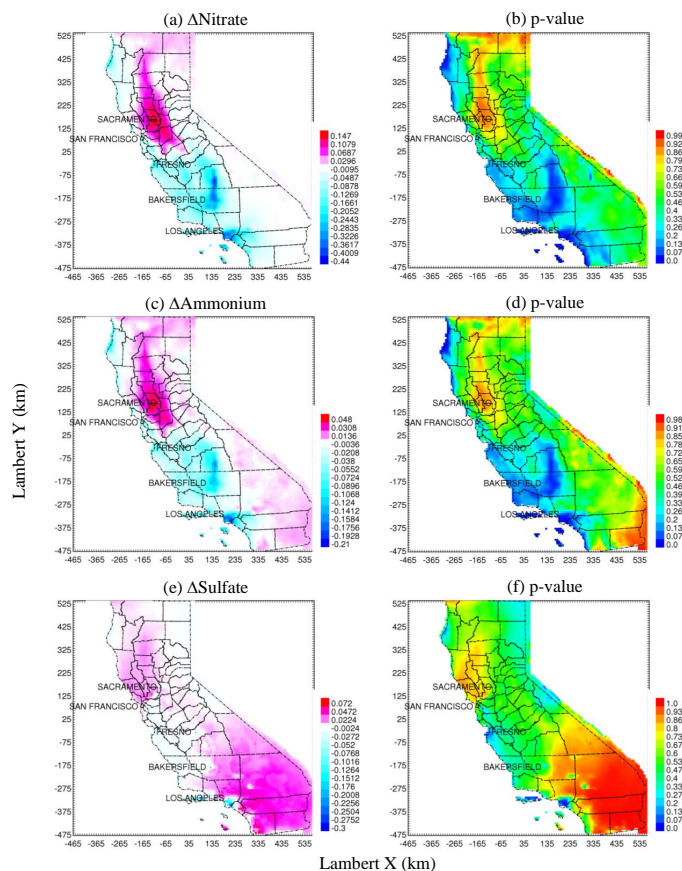


Fig. 7. Changes in annual average secondary $PM_{2.5}$ concentrations, and their corresponding p-values in CA likely to occur in the future (2047–2053) due to climate change from the present-day (2000–2006). The p-value quantifies the likelihood that average future concentrations are equal to present day concentrations.

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