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Impact of Climate Change on Fine Particulate Matter $(PM_{2.5})$ Air Quality

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Impact of climate change on fine particulate matter (PM_{2.5}) air quality

Abstract

This dissertation investigates the impact of 2000-2050 climate change on fine particulate matter (PM_{2.5}) air quality. We first applied a multiple linear regression model to study the correlations of total PM_{2.5} and its components with meteorological variables using the past decadal PM_{2.5} observations over the contiguous US. We find that daily variation in meteorology can explain up to 50% of PM_{2.5} variability.

Temperature is positively correlated with sulfate and organic carbon (OC) almost everywhere. The correlation of nitrate with temperature is negative in the Southeast but positive in California and the Great Plains. Relative humidity (RH) is positively correlated with sulfate and nitrate, but negatively with OC. Precipitation is strongly negatively correlated with all PM_{2.5} components.

We then compared the observed correlations of PM_{2.5} with meteorological variables with results from the GEOS-Chem chemical transport model. The results indicate that most of the correlations of PM_{2.5} with temperature and RH do not arise from direct dependence but from covariation with synoptic transport. We applied principal component analysis and regression to identify the dominant meteorological modes controlling PM_{2.5} variability, and showed that 20-40% of the observed PM_{2.5}

daily variability can be explained by a single dominant meteorological mode: cold frontal passages in the eastern US and maritime inflow in the West.

From 1999-2010 observations we further showed that interannual variability of annual mean $PM_{2.5}$ in most of the US is strongly correlated with the synoptic period T of the dominant meteorological mode as diagnosed from a spectral-autoregressive analysis. We then used the observed local $PM_{2.5}$ -to-period sensitivity to project $PM_{2.5}$ changes from the 2000-2050 changes in T simulated by fifteen IPCC AR4 GCMs following the SRES A1B scenario. We project a likely increase of $\sim 0.1 \mu\text{g m}^{-3}$ in annual mean $PM_{2.5}$ in the eastern US arising from less frequent frontal ventilation, and a likely decrease of $\sim 0.3 \mu\text{g m}^{-3}$ in the northwestern US due to more frequent maritime inflows. These circulation-driven changes are relatively small, representing only a minor climate penalty or benefit for $PM_{2.5}$ regulatory purpose.

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*Dedicated to my parents,
Laiha Wing and Hung Tai*

Chapter 1. Overview

1.1. Introduction

Air pollution is highly dependent on weather, and it follows that climate change could significantly impact air quality. The pollutants of the most public health concern are ozone and fine particulate matter with diameter less than 2.5 μm ($\text{PM}_{2.5}$). A number of studies reviewed by Jacob and Winner (2009) have used chemical transport models (CTMs) driven by general circulation models (GCMs) to diagnose the effects of 21st-century climate change on ozone and $\text{PM}_{2.5}$ air quality at northern mid-latitudes. They generally concur that 2000-2050 climate change will degrade ozone air quality in polluted regions by 1-10 ppb driven largely by temperature increase (Weaver et al., 2009). This finding is buttressed by observed correlations of ozone with temperature that are well reproduced by models (Jacob et al., 1993; Sillman and Samson, 1995; Rasmussen et al., 2012). By contrast, these GCM-CTM studies find potentially significant effect for $\text{PM}_{2.5}$ (± 0.1 - $1 \mu\text{g m}^{-3}$) but they show no consistency even in the sign of the effect (Jacob and Winner, 2009).

The uncertain sensitivity to climate change in the case of $\text{PM}_{2.5}$ reflects in part the complexity of the dependence of different $\text{PM}_{2.5}$ components on meteorological variables, and in part the coupling of aerosols to the hydrological cycle which is not well represented in GCMs (Racherla and Adams, 2006; Pye et al., 2009). Higher temperatures can lead to higher sulfate concentrations due to faster SO_2 oxidation, but

to lower nitrate and organic components due to volatility (Sheehan and Bowman, 2001; Aw and Kleeman, 2003; Dawson et al., 2007; Kleeman, 2008). Biogenic emissions of PM_{2.5} precursors including agricultural ammonia, soil NO_x, and volatile organic compounds (VOCs) increase with temperature and further complicate the PM_{2.5}-temperature relationship (Pinder et al., 2004; Bertram et al., 2005; Guenther et al., 2006). Higher relative humidity (RH) promotes aqueous-phase sulfate production and ammonium nitrate formation (Koch et al., 2003; Liao et al., 2006; Dawson et al., 2007), but inhibits fires, which are important contributors to organic aerosols in many regions (Park et al., 2007; Spracklen et al., 2009). An increase in precipitation causes a decrease in all PM_{2.5} components through scavenging (Liao et al., 2006; Dawson et al., 2007; Pye et al., 2009). Changes in precipitation and in planetary boundary layer (PBL) depth have a consistent effect on PM_{2.5} components but their projections in GCMs are highly uncertain (Jacob and Winner, 2009).

Synoptic-scale transport should also be an important factor driving the effect of climate change on PM_{2.5}. Previous studies have used principal component analysis (PCA) to identify important meteorological modes of variability for PM_{2.5} air quality (Cheng et al., 2007; Thishan Dharshana et al., 2010). Thishan Dharshana et al. (2010) found that as much as 30% of PM_{2.5} daily variability in the US Midwest is associated with passages of synoptic weather systems. Cold fronts associated with mid-latitude cyclone passages provide the dominant ventilation pathway for the eastern US (Cooper et al., 2001; Li et al., 2005). A general reduction in the frequency of these cyclones is expected as a result of greenhouse warming (Lambert and Fyfe, 2006; Christensen et

al., 2007; Pinto et al., 2007), potentially leading to more frequent and prolonged stagnation episodes (Mickley et al., 2004; Murazaki and Hess, 2006). Leibensperger et al. (2008) found a strong anticorrelation between summer cyclone frequency and ozone pollution in the eastern US for 1980-2006, and further showed evidence of a long-term decline in cyclone frequency over that period that significantly hindered attainment of ozone air quality standards. However, there is substantial uncertainty in regional projections of future cyclone frequency (Ulbrich et al., 2009; Lang and Waugh, 2011).

1.2. Motivation

The GCM-CTM projections for $PM_{2.5}$ are uncertain partly because the ability of these models to reproduce present-day relationships between $PM_{2.5}$ and meteorological variables has not been tested by observations. Only a few observational studies so far have examined such correlations of $PM_{2.5}$ with meteorological variables and then only for small regional domains and a limited suite of species and meteorological variables (Vukovich and Sherwell, 2002; Aw and Kleeman, 2003; Koch et al., 2003; Chu, 2004; Wise and Comrie, 2005). We need a better observational foundation for $PM_{2.5}$ -weather relationships. CTMs that can reasonably reproduce the present-day correlations can then be used to interpret and understand the sensitivity of $PM_{2.5}$ to various weather conditions and climate change.

Another difficulty in projecting the climate change effect on air quality is the underlying GCM uncertainty in simulating regional climate. This uncertainty arises both from model noise (climate chaos) and from model error (physics, parameters,

numerics). All GCM-CTM studies to date examining climate change effect on $PM_{2.5}$ have used a single climate change realization from a single GCM (Jacob and Winner, 2009), so it is no surprise that they would yield inconsistent results. The standard approach adopted by the Intergovernmental Panel on Climate Change (IPCC) to reduce uncertainties in GCM regional climate projections is to use multiple realizations from an ensemble of GCMs (Christensen et al., 2007). Such an ensemble analysis is not practical for GCM-CTM studies of air quality because of the computational expense associated with chemistry and aerosol microphysics. Therefore, an alternative is to focus on ensemble analysis of GCM projections of the major meteorological modes that determine air quality.

1.3. Research questions and approach

The objective of this dissertation was to determine the impact of 2000-2050 climate change on $PM_{2.5}$ air quality. We addressed the following major questions:

- What are the observed relationships of $PM_{2.5}$ and its major components with meteorological variables? How do we physically interpret these relationships?
- Can chemical transport models reproduce the observed correlations, and be used to understand the complex relationships between $PM_{2.5}$ and weather?
- Given the complex meteorological dependence of $PM_{2.5}$, can we still find a single, reliable metric that can capture most of the daily $PM_{2.5}$ variability and be used to predict $PM_{2.5}$ air quality?

- What climatic factors control the interannual variability of $PM_{2.5}$? Can we use such information to more robustly project future $PM_{2.5}$ air quality based on climate model projections?

To address these questions, we first applied a multiple linear regression (MLR) model to determine the correlations of $PM_{2.5}$ and its major components with meteorological variables using 1998-2008 daily observations (EPA-AQS and NCEP/NCAR Reanalysis 1) over the contiguous US. The data were deseasonalized and detrended to focus on synoptic-scale correlations. Our aim here was to uncover important correlations that can be used to gain insight into the sensitivity of $PM_{2.5}$ to climate change as well as to test the GCM-CTM representations of aerosol processes.

We then applied the GEOS-Chem global CTM to interpret the observed correlations between $PM_{2.5}$ components and meteorological variables in the contiguous US. A similar MLR model was used to correlate both observed and simulated daily mean concentrations of $PM_{2.5}$ for 2004-2008. As we will see, interpretation of these correlations is complicated by the covariation of meteorological variables with synoptic transport. To address this issue, we used PCA and regression (with validation from examination of weather charts) to determine the dominant meteorological modes of daily $PM_{2.5}$ variability in different US regions.

We further applied a spectral-autoregressive analysis to the dominant meteorological modes to determine their synoptic-scale frequencies. The corresponding synoptic periods in different US regions were correlated with detrended annual mean $PM_{2.5}$. A reduced-major-axis regression was used to determine the local $PM_{2.5}$ -to-period

sensitivity ($dPM_{2.5}/dT$), which would be used as a metric to infer the effect of climate change on $PM_{2.5}$.

To examine intra-model variability of future projections of synoptic periods due to climate chaos, we conducted an ensemble of five realizations of 2000-2050 climate change using the Goddard Institute for Space Studies (GISS) GCM III with A1B greenhouse and aerosol forcings. We diagnosed the 2000-2050 change in the period of cyclones in the US Midwest and from there inferred the impact on annual mean $PM_{2.5}$.

Finally, we examined the Coupled Model Intercomparison Project phase 3 (CMIP3) multi-model dataset of climate change simulations produced by the ensemble of GCMs contributing to the IPCC 4th Assessment Report (AR4). We used the CMIP3 archive of 15 GCMs under the A1B scenario to project the 2000-2050 trends of synoptic periods of the dominant meteorological modes, and from there deduced the corresponding regional trends in $PM_{2.5}$ across the continental US. These climate-driven $PM_{2.5}$ projections, independent of trends in anthropogenic emissions, would represent the “climate penalty” or “benefit” for $PM_{2.5}$, which will aid air quality managers to plan emission goals accordingly.

1.4. Major results

We found that daily variation in meteorology as described by the MLR including eight predictor variables (temperature, relative humidity, precipitation, 850-hPa geopotential height or sea level pressure, sea-level pressure tendency, wind speed, E-W and N-S wind direction) can explain up to 50% of the observed daily $PM_{2.5}$

variability in the US. Stagnation is a strong predictor; $\text{PM}_{2.5}$ concentrations in the US are on average $2.6 \mu\text{g m}^{-3}$ higher on a stagnant day vs. non-stagnant day.

We observed strong positive correlations of all $\text{PM}_{2.5}$ components with temperature in most of the US, except for nitrate in the Southeast where the correlation is negative. A temperature perturbation simulation with GEOS-Chem revealed that most of the correlations of $\text{PM}_{2.5}$ with temperature do not arise from direct dependence on temperature but from covariation with synoptic transport. Exceptions are nitrate and OC in the Southeast, where the direct dependence of ammonium nitrate thermodynamics and biogenic VOC emissions on temperature contributes significantly to the correlations. RH is generally positively correlated with sulfate and nitrate but negatively correlated with OC; the correlations also appear to be mainly driven by covariation of RH with synoptic transport. Total $\text{PM}_{2.5}$ is strongly negatively correlated everywhere with precipitation and wind speed. Correlation with vector winds shows that the industrial Midwest is a source of sulfate for much of the country, and that nitrate is generally highest under inflow from agricultural regions (reflecting NH_3 emissions). There is also some association of elevated OC with flow from regions of elevated biogenic and fire emissions in the Southeast and the West.

We found from the PCA and regression that 20-40% of the observed $\text{PM}_{2.5}$ day-to-day variability in different US regions can be explained by a single dominant synoptic meteorological mode: cold frontal passages in the eastern US and maritime inflow in the West. These and other transport modes are found to contribute to most of

the overall correlations of different PM_{2.5} components with temperature and RH except in the Southeast.

We showed that the interannual variability of annual mean PM_{2.5} in the Midwest for 1999-2010 is strongly correlated with cyclone frequency as diagnosed from the spectral-autoregressive analysis, with $dPM_{2.5}/dT = \sim 1 \mu\text{g m}^{-3} \text{ d}^{-1}$. Of the five realizations of 2000-2050 climate change using the GISS GCM III, three found a significant decrease in cyclone frequency over the US Midwest, one found no significant change and one found a significant increase. From this ensemble we derive a likely increase in annual mean PM_{2.5} of $\sim 0.1 \mu\text{g m}^{-3}$ in the Midwest in the 2050s climate. This is consistent with previous GCM-CTM studies using the same GCM and suggests that cyclone frequency may be a major driver of the effect of climate change on PM_{2.5} air quality. However, the variability of cyclone trends (including in sign) across multiple realizations of the same GCM with identical forcings demonstrates the importance of multiple climate change realizations in GCM-CTM studies because of climate chaos.

We further showed that, on a $4^\circ \times 5^\circ$ latitude-by-longitude grid scale, the observed 1999-2010 interannual variability of PM_{2.5} in most of the US is strongly correlated with the periods (T) of the dominant synoptic-scale meteorological modes, particularly in the eastern US where these modes correspond to frontal passages. We found that all GCMs have significant skill in reproducing T and its spatial distribution over the US, reflecting their ability to capture the baroclinicity of the atmosphere.

Finally, we examined the 2000-2050 trends in synoptic periods T across the US as simulated by the 15 GCMs from CMIP3. We found a general slowing down of

synoptic circulation in the eastern US, as measured by an increase in T. We inferred that changes in circulation driven by climate change will likely increase annual mean PM_{2.5} in the eastern US by ~0.1 μg m⁻³, reflecting a more stagnant mid-latitude troposphere and less frequent ventilation by frontal passages. We also projected a likely decrease by ~0.3 μg m⁻³ in the Northwest due to more frequent ventilation by maritime inflows. Potentially larger regional effects of climate change on PM_{2.5} air quality may arise from changes in temperature, biogenic emissions, wildfires, and vegetation. Overall, however, it is unlikely that 2000-2050 climate change will modify annual mean PM_{2.5} by more than 0.5 μg m⁻³. These climate change effects represent a relatively minor penalty or benefit for PM_{2.5} regulatory purpose.

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Chapter 2. Correlations between fine particulate matter (PM_{2.5}) and meteorological variables in the United States: implications for the sensitivity of PM_{2.5} to climate change

{ Tai, A.P.K., L.J. Mickley, and D.J. Jacob. 2010. Correlations between fine particulate matter (PM_{2.5}) and meteorological variables in the United States: implications for the sensitivity of PM_{2.5} to climate change. *Atmos. Environ.* 44, 3976-3984. }

Abstract

We applied a multiple linear regression (MLR) model to study the correlations of total PM_{2.5} and its components with meteorological variables using an 11-year (1998-2008) observational record over the contiguous US. The data were deseasonalized and detrended to focus on synoptic-scale correlations. We find that daily variation in meteorology as described by the MLR can explain up to 50% of PM_{2.5} variability with temperature, relative humidity (RH), precipitation, and circulation all being important predictors. Temperature is positively correlated with sulfate, organic carbon (OC) and elemental carbon (EC) almost everywhere. The correlation of nitrate with temperature is negative in the Southeast but positive in California and the Great Plains. RH is positively correlated with sulfate and nitrate, but negatively with OC and EC. Precipitation is strongly negatively correlated with all PM_{2.5} components. We find that PM_{2.5} concentrations are on average 2.6 $\mu\text{g m}^{-3}$ higher on stagnant vs. non-stagnant days. Our observed correlations provide a test for chemical transport models used to

simulate the sensitivity of PM_{2.5} to climate change. They point to the importance of adequately representing the temperature dependence of agricultural, biogenic and wildfire emissions in these models.

2.1. Introduction

Particulate matter with diameter of 2.5 µm or less (PM_{2.5}) is a major air quality concern because of its effects on human health. PM_{2.5} concentrations depend on meteorological conditions, suggesting that climate change could have significant effects on PM_{2.5} air quality. Several studies using chemical transport models (CTMs) driven by general circulation models (GCMs) have investigated the effects of 21st-century climate change on PM_{2.5} (Liao et al., 2006; Racherla and Adams, 2006; Tagaris et al., 2007; Heald et al., 2008; Avise et al., 2009; Pye et al., 2009). They find significant effects ($\pm 1 \mu\text{g m}^{-3}$) but there is no consistency across studies, including in the sign of effects, so that little can be concluded at present regarding the sensitivity of PM_{2.5} to climate change (Jacob and Winner, 2009).

The uncertain sensitivity to climate change in the case of PM_{2.5} reflects in part the complexity of the dependence of different PM_{2.5} components on meteorological variables, and in part the coupling of aerosols to the hydrological cycle which is not well represented in GCMs (Racherla and Adams, 2006; Pye et al., 2009). For example, sulfate concentrations are expected to increase with increasing temperature due to faster SO₂ oxidation, but semi-volatile components such as nitrate and organics are expected to decrease as they shift from the particle phase to the gas phase at higher temperature (Sheehan and Bowman, 2001; Aw and Kleeman, 2003; Dawson et al., 2007; Tsigaridis

and Kanakidou, 2007; Kleeman, 2008). Increasing cloud can increase sulfate due to in-cloud production, and higher relative humidity (RH) promotes the formation of ammonium nitrate, but an increase in precipitation causes a decrease in all PM_{2.5} components through scavenging (Koch et al., 2003; Liao et al., 2006; Dawson et al., 2007; Pye et al., 2009). Increased stagnation in the future climate may also worsen PM_{2.5} air quality (Liao et al., 2006; Leibensperger et al., 2008).

GCM-CTM studies of the effects of climate change on air quality can only be as good as the model descriptions of processes. Confidence is usually assessed by cross-model comparisons (Weaver et al., 2009) and comparisons with observed concentrations. However, biases common to all models may render consensus misleading, and comparisons with observed concentrations can only test the simulation of the present atmosphere, not the sensitivity to climate change. It would be far more relevant to test the ability of models to reproduce observed correlations of air quality with meteorological variables, as has been done for ozone through the observed correlation with temperature (Jacob and Winner, 2009). We need a better observational foundation to do the same with PM_{2.5}. Only a few observational studies so far have examined the correlations of PM with meteorological variables and then only for small regional domains and a limited suite of species and meteorological variables (Vukovich and Sherwell, 2002; Aw and Kleeman, 2003; Koch et al., 2003; Chu, 2004; Wise and Comrie, 2005).

To address this need, we present here a systematic statistical analysis to quantify the correlations of total PM_{2.5} and its different components with meteorological

variables on the scale of the contiguous US and for an 11-year record of observations (1998-2008). Our aim is to uncover important correlations that can be used to gain insight into the sensitivity of $PM_{2.5}$ to climate change as well as to test the GCM-CTM representations of aerosol processes.

2.2. Data and methods

2.2.1. Meteorological data

Daily mean meteorological data from 1998 to 2008 were obtained from the National Center for Environmental Prediction / National Center for Atmospheric Research (NCEP/NCAR) Reanalysis 1 (<http://www.cdc.noaa.gov/cdc/data.ncep.reanalysis.html>) (Kalnay et al., 1996; Kistler et al., 2001). Gridded U.S. daily precipitation observations were obtained from the National Oceanic and Atmospheric Administration (NOAA) Climate Prediction Center (<http://www.cpc.ncep.noaa.gov/products/precip/realtime/GIS/retro.shtml>). The meteorological parameters are listed in Table 2.1. They include surface temperature (x_1), three hydrometeorological parameters (x_2, x_3, x_4), two anticyclone parameters (x_5, x_6), wind speed (x_7), and wind direction (x_8, x_9). All except x_6, x_8 and x_9 were deseasonalized and detrended by subtracting the 30-day moving averages from the original data, allowing us to focus on the synoptic-scale variability.

Table 2.1. Meteorological parameters considered in the statistical analysis ^a.

Independent variable	Meteorological parameter
x_1	Surface air temperature (K) ^b
x_2	Surface air relative humidity (%) ^b
x_3	Daily total precipitation (cm d ⁻¹) ^c
x_4	Total column cloud cover (%) ^d
x_5	Geopotential height at 850 hPa (km)
x_6	Local rate of change of sea level pressure $dSLP/dt$ (hPa d ⁻¹)
x_7	Surface wind speed (m s ⁻¹) ^{b, e}
x_8	East-west wind direction indicator $\cos\theta$ (dimensionless) ^f
x_9	North-south wind direction indicator $\sin\theta$ (dimensionless) ^f

a. All meteorological parameters are 24-hour averages. Except for daily total precipitation and cloud cover, all data are from NCEP/NCAR Reanalysis 1 with spatial resolution of $2.5^\circ \times 2.5^\circ$.

b. “Surface” data are from the 0.995 sigma level.

c. Obtained from the NOAA Climate Prediction Center, regridded from original spatial resolution of $0.25^\circ \times 0.25^\circ$ to $2.5^\circ \times 2.5^\circ$.

d. Obtained from NCEP/NCAR Reanalysis 1, regridded from original spatial resolution in T62 Gaussian grid with 192×94 points to $2.5^\circ \times 2.5^\circ$.

e. Calculated from the horizontal wind vectors (u, v).

f. θ is the angle of the horizontal surface wind vector counterclockwise from the east.

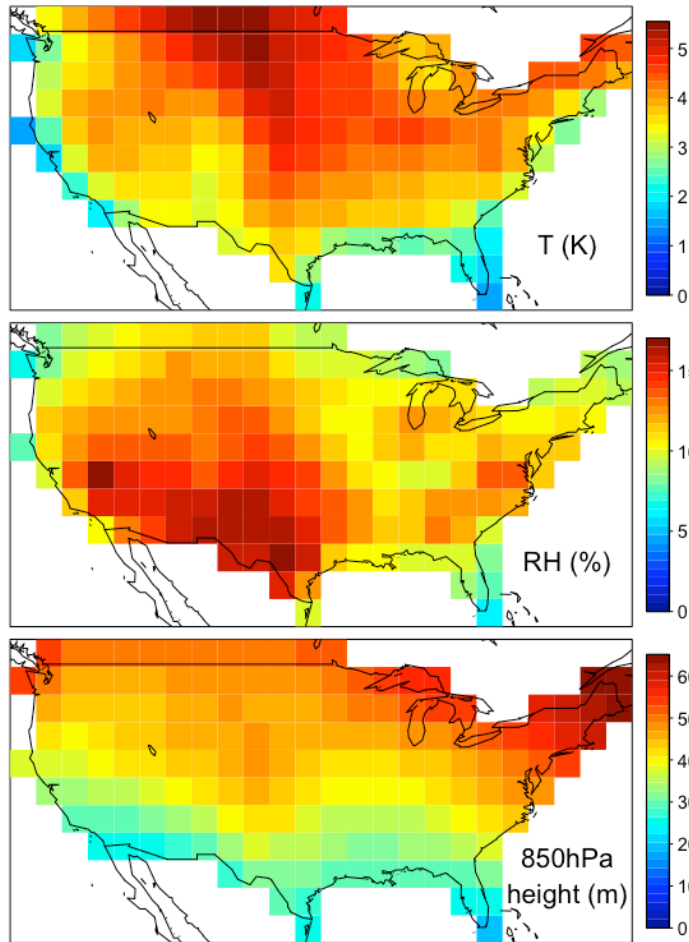


Figure 2.1. Daily variability of surface air temperature, relative humidity and 850-hPa geopotential height in the US. Figure shows standard deviations for deseasonalized and detrended observations from 1998-2008.

Figure 2.1 shows the standard deviation of a few deseasonalized and detrended meteorological variables. Temperature has greater variability in the North than in the South, inland than on the coasts. RH variability is largest in the Southwest and South-central. The 850-hPa geopotential height is more variable in the North, particularly in the Northeast and Midwest, reflecting frontal passages that drive ventilation of these regions.

2.2.2. $PM_{2.5}$ data

Daily mean surface concentrations of total $PM_{2.5}$ from 1998 to 2008 measured with the Federal Reference Method (FRM) were obtained from the EPA Air Quality System (EPA-AQS) (<http://www.epa.gov/ttn/airs/airsaqs/>), which covers a network of ~1000 sites in the contiguous US. Speciation data from 2000 to 2008 including sulfate, nitrate, ammonium, organic carbon (OC), and elemental carbon (EC) were obtained from EPA-AQS for State and Local Air Monitoring Stations (SLAMS) and the Speciation Trends Network (STN), a total of ~200 sites. All PM data were collected either every day, every 3rd day (most common for total $PM_{2.5}$) or every 6th day (most common for speciation data). Figure 2.2 shows the site locations in 2005 and the regional division used in this work.

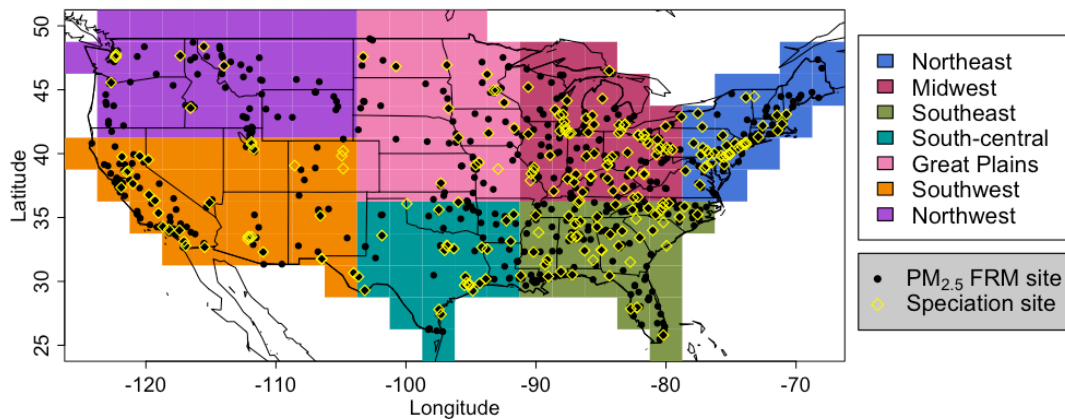


Figure 2.2. Locations of EPA Air Quality System $PM_{2.5}$ -monitoring sites in 2005. Black dots denote total $PM_{2.5}$ monitors where data are collected with Federal Reference Method (FRM); yellow diamonds denote monitors in chemical speciation network (SLAMS + STN). US regional divisions used in our analysis are also shown.

Interpolated $2.5^{\circ} \times 2.5^{\circ}$ 24-h average $PM_{2.5}$ fields were constructed from site measurements to produce an 11-year time series of $PM_{2.5}$ concentrations for each grid square. We used inverse distance weighting, in which all n sampled values (z_i) within a specified search distance (d_{\max}) are inversely weighted by their distances (d_{ij}) from the grid centroid to produce an average (z_j) for each grid square j :

$$z_j = \frac{\sum_{i=1}^n (1/d_{ij})^k z_i}{\sum_{i=1}^n (1/d_{ij})^k} \quad (2.1)$$

where k is the power parameter. We chose $k = 2$ and $d_{\max} = 500$ km. Results are not overly sensitive to the choice of interpolation method; an alternate method with simple spatial averaging of data in individual grid squares produced similar correlation results. Kriging has been used in the past for spatial interpolation of air quality data (Lefohn et al., 1988; Jerrett et al., 2005), but we did not use it here because the $PM_{2.5}$ data are too unevenly distributed (Wong et al., 2004).

Figure 2.3 shows the annual mean concentrations of total $PM_{2.5}$ and the five major $PM_{2.5}$ components, interpolated on the $2.5^{\circ} \times 2.5^{\circ}$ grid and averaged over the 11-year (total $PM_{2.5}$) and 9-year (speciation) periods. We do not consider dust and sea salt as they are generally small contributors to $PM_{2.5}$. Spatial interpolation is more robust in the East, where site density is higher and urban-rural contrast is lower than in the West (Malm et al., 2004; Tang et al., 2004). $PM_{2.5}$ concentrations have generally decreased over the 1998-2008 period and this long-term trend is removed from our analysis as described below.

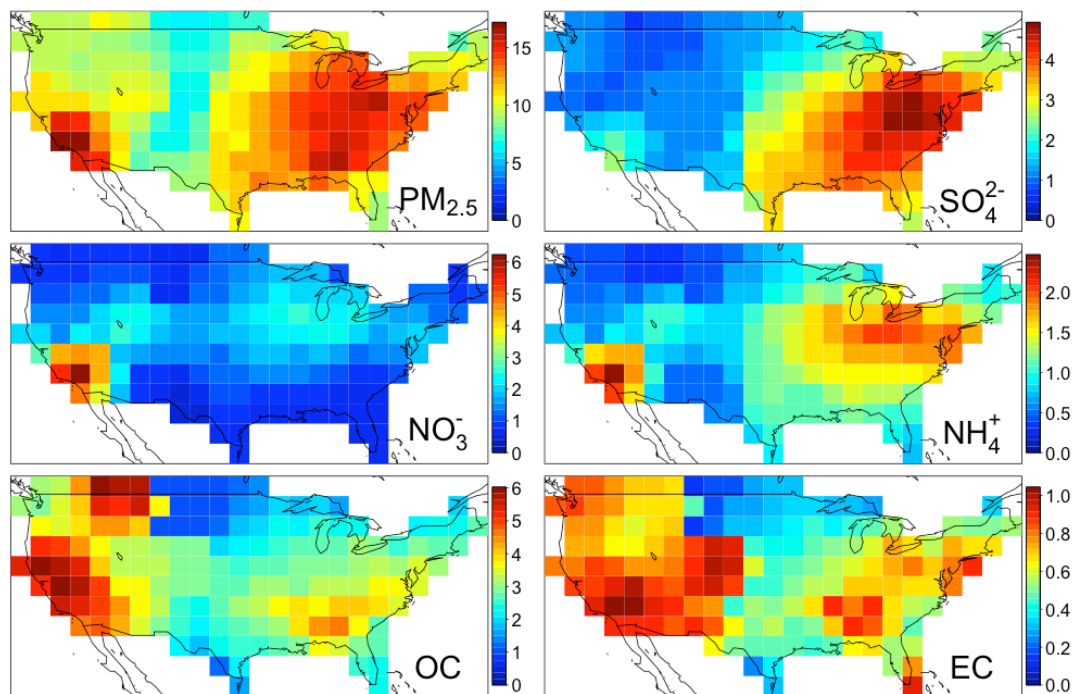


Figure 2.3. Annual mean concentrations of total $PM_{2.5}$ and its five major components, interpolated on a $2.5^\circ \times 2.5^\circ$ grid as described in text. Concentrations are in units of $\mu\text{g m}^{-3}$ and averaged over 1998-2008 for total $PM_{2.5}$ and 2000-2008 for individual species. OC concentrations were adjusted to account for background filter contamination by subtracting the 2005 mean field blank measurements. Note differences in scales between panels.

2.2.3. Multiple linear regression

We used a multiple linear regression (MLR) model to correlate $PM_{2.5}$ and its components to the meteorological variables in Table 2.1. All $PM_{2.5}$ data were deseasonalized and detrended in the same way as with the meteorological variables. This focuses the correlations on synoptic time scales, avoiding aliasing from common seasonal variations or long-term trends. The model is of the form

$$y = \beta_0 + \sum_{k=1}^9 \beta_k x_k + \text{interaction terms} \quad (2.2)$$

where y is the deseasonalized and detrended concentration of total $\text{PM}_{2.5}$ or its components for each grid square, (x_1, \dots, x_9) is the ensemble of meteorological variables in Table 2.1, and β_k are the regression coefficients. The interaction terms are up to third-order $(x_k x_l x_m)$. For each grid square, the regression was done stepwise to add and delete terms based on Akaike Information Criterion (AIC) statistics to obtain the best model fit (Venables and Ripley, 2003). The number of explanatory terms x_k in the MLR is on average 21. The Cook's distances (Cook, 1979) show that the regression results reflect the broad population rather than a small number of influential outliers. The variance inflation factor (Velleman and Welsch, 1981) ranges between 1.0 and 2.7, indicating that the problem of multicollinearity among meteorological variables is generally unimportant. The coefficient of determination (R^2) quantifies the fraction of variance of $\text{PM}_{2.5}$ that can be accounted for with the MLR model (Kutner, 2004).

In addition to full-year regressions, we also conducted regressions for seasonal subsets of data (DJF, MAM, JJA, SON). These generally showed results similar to the full-year correlations but we will highlight some prominent differences.

2.3. Correlations of PM_{2.5} and components with meteorological variables

2.3.1. Total PM_{2.5}

Figure 2.4 shows the relationships of total PM_{2.5} with meteorological variables, as measured by the MLR coefficients β_k in Eq. (2.2) associated with each meteorological variable. Interaction terms are relatively small and not shown. Individual PM_{2.5} components show similar correlations as total PM_{2.5} for all meteorological variables except temperature, RH, and wind direction. Component-specific correlations for these variables are discussed in the following subsections.

Temperature is positively correlated with PM_{2.5} concentrations throughout the US. This contrasts with the CTM sensitivity study of Dawson et al. (2007), which found an average negative temperature effect in the East of -0.016 and -0.17 $\mu\text{g m}^{-3} \text{K}^{-1}$ in summer and in winter, respectively, primarily due to volatilization of ammonium nitrate at higher temperature. Dawson et al. (2007) perturbed temperature in their CTM while holding all other variables constant. The positive temperature relationship that we find here reflects meteorological cofactors as discussed in Section 2.3.2.

Precipitation is negatively correlated with PM_{2.5} concentrations throughout the US, as would be expected from the scavenging sink. The correlation of PM_{2.5} with RH is positive in the Northeast and Midwest but negative in the Southeast and the West. The correlation with column cloud cover is generally weak. We find surface RH a better indicator than column cloud cover for liquid water content within the surface boundary layer.

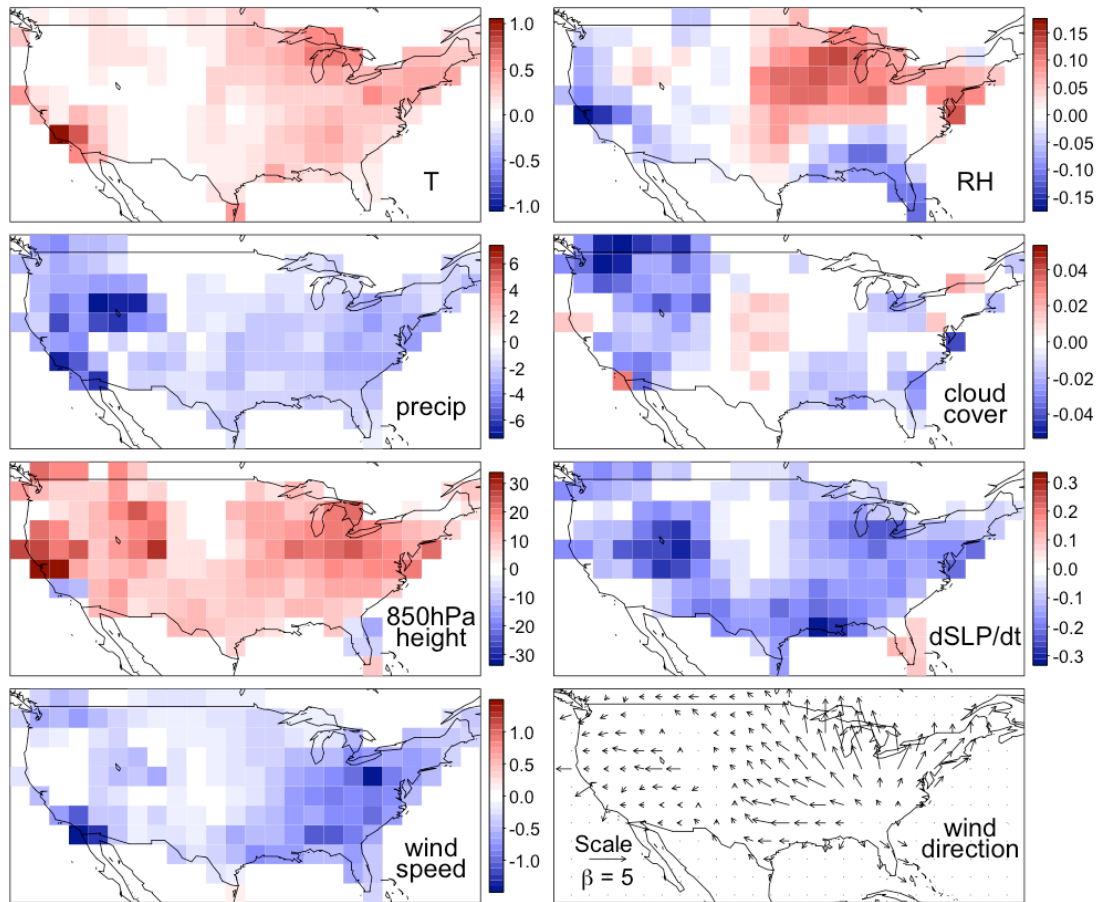


Figure 2.4. Correlations of total $\text{PM}_{2.5}$ with meteorological variables. Figure shows multiple linear regression coefficients, β_k , in units of $\mu\text{g m}^{-3} \text{D}^{-1}$, where D is dimension of each meteorological variable listed in Table 1. Wind direction panel shows vector sums of regression coefficients β_8 and β_9 . Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05).

Figure 2.4 also shows that high $\text{PM}_{2.5}$ concentrations are correlated with high 850-hPa geopotential height (anticyclonic conditions), decreasing sea-level pressure ($d\text{SLP}/dt < 0$), low wind speed, and (in the East) southerly flow. The positive association with anticyclonic conditions can be simply explained by dry weather and

subsidence inversions. The negative association with $dSLP/dt$ reflects PM accumulation on the tail end (west side) of anticyclones and PM removal by cold fronts.

Figure 2.5 shows the coefficients of determination (R^2) for the MLR model fit to observations, with values adjusted to account for different number of explanatory terms in the MLR at each location (Kutner, 2004). They range from 0.1 to 0.5 depending on grid square. Wise and Comrie (2005) similarly found R^2 values of 0.1-0.5 for correlations of PM to meteorological variables at sites in the Southwest. We find the largest R^2 in the Northeast, Midwest and Pacific Northwest, where meteorological variables can explain up to 50% of daily $PM_{2.5}$ variability. Values are lowest in the west-central US but this could reflect the paucity of sites to define mean concentrations in $2.5^\circ \times 2.5^\circ$ grid squares (Fig. 2.2).

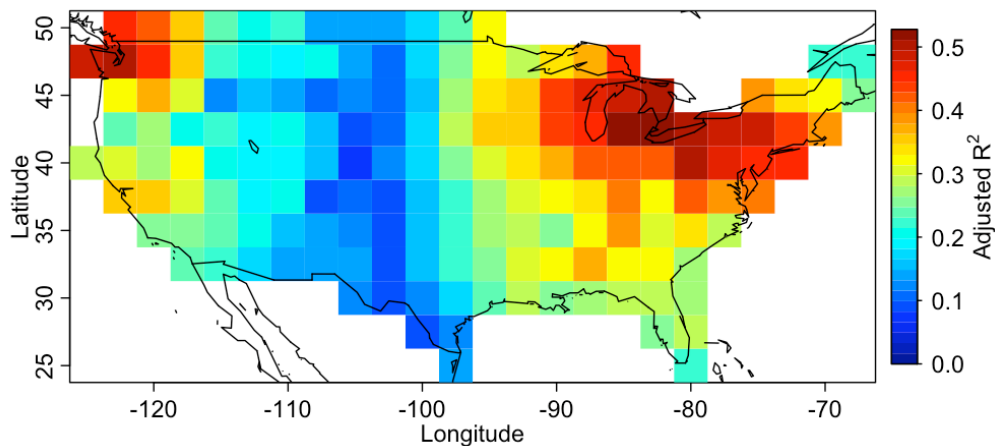


Figure 2.5. Coefficients of determination (R^2) for multiple linear regression of deseasonalized and detrended 1998-2008 total $PM_{2.5}$ concentrations on meteorological variables of Table 2.1. Values are adjusted to account for different number of explanatory terms at each location.

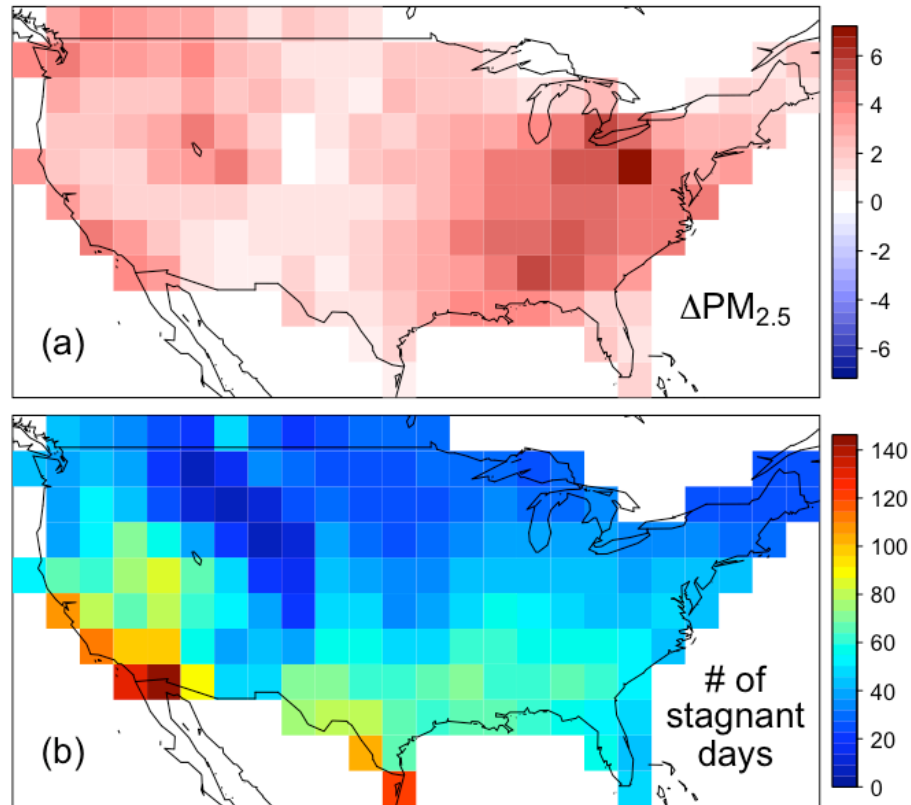


Figure 2.6. (a) Average differences in deseasonalized total PM_{2.5} concentrations on stagnant vs. non-stagnant days, based on deseasonalized and detrended 1998-2008 observations. Stagnation is defined following Wang and Angell (1999). Only differences with 95% confidence (p -value < 0.05) are shown. (b) Number of stagnant days per year averaged over 1998-2008.

Stagnation is characterized by anticyclonic condition, weak wind, no precipitation, and usually high temperature. Taken together, the results above illustrate strong association of high PM_{2.5} levels with stagnation. A simple linear regression of deseasonalized and detrended total PM_{2.5} concentrations on a categorical variable for stagnation (one for a stagnant day, zero otherwise) was conducted to estimate the average differences in total PM_{2.5} between a stagnant vs. non-stagnant day. A stagnant

day is defined in our study as having daily mean SLP geostrophic wind $< 8 \text{ m s}^{-1}$, daily mean 500 hPa wind $< 13 \text{ m s}^{-1}$, and daily total precipitation $< 0.01 \text{ cm d}^{-1}$ (Wang and Angell, 1999). The result is shown in Fig. 2.6. Total $\text{PM}_{2.5}$ is on average $2.6 \mu\text{g m}^{-3}$ higher on a stagnant day. Fig. 2.6 also shows the average number of stagnant days per year, highlighting the severity of stagnation in the Southwest.

2.3.2. $\text{PM}_{2.5}$ components vs. temperature

Figure 2.7 shows the deseasonalized relationships of the major $\text{PM}_{2.5}$ components with surface air temperature, as measured by the MLR coefficient β_1 in Eq. (2.2). We do not show ammonium as it is mainly the counter-ion for sulfate and nitrate. The relationships in Fig. 2.7 are positive almost everywhere for all components except nitrate. The relationship for nitrate is negative in the South but positive in the North and California. We elaborate on each component below.

The MLR coefficients for sulfate in the East are on average 530 and $25 \text{ ng m}^{-3} \text{ K}^{-1}$ in summer and winter, respectively. CTM sensitivity simulations also find an increase of sulfate with temperature due to higher SO_2 oxidation rates (Aw and Kleeman, 2003; Dawson et al., 2007; Kleeman, 2008) but the dependence is much weaker. Dawson et al. (2007) found for the same region an average sulfate response of 34 and $1.6 \text{ ng m}^{-3} \text{ K}^{-1}$ in summer and in winter, respectively, an order of magnitude smaller than our coefficients. This suggests that the observed correlation of sulfate with temperature is mainly determined by joint association with southerly flow, stagnation, and ventilation of pollution by cold fronts.

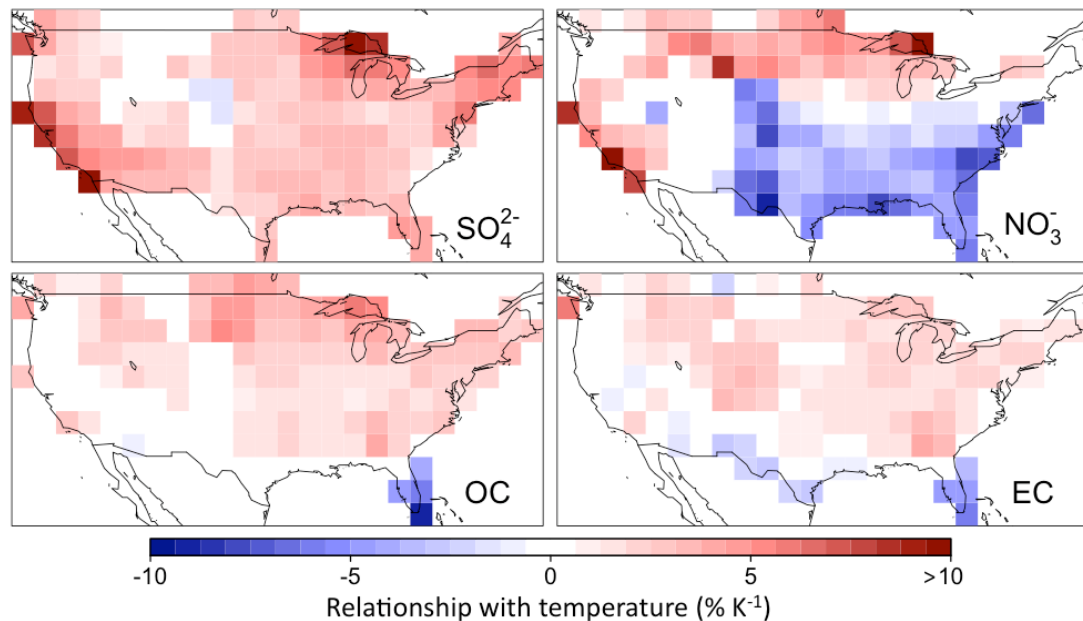


Figure 2.7. Correlations of PM_{2.5} components with surface air temperature. Figure shows multiple linear regression coefficients β_1 , normalized to annual mean concentrations of Fig. 2.3. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05).

The strong positive correlation that we find for nitrate in the North and California contrasts with CTM sensitivity studies indicating a strong negative dependence of nitrate on temperature due to increased volatilization of ammonium nitrate (Aw and Kleeman, 2003; Dawson et al., 2007; Kleeman, 2008). Part of the explanation could be the joint association with stagnation and cold fronts. Also, these CTM sensitivity studies did not account for the increase in agricultural NH_3 and NO_x emissions with increasing temperature (Bouwman et al., 2002; Pinder et al., 2004; Aneja et al., 2008). Nitrate formation in most of the US is limited by the supply of NH_3 (Park et al., 2004). In the Great Plains where nitrate formation is limited by the supply

of nitric acid (Park et al., 2004), the positive correlation may reflect the temperature dependence of soil NO_x emissions (Bertram et al., 2005).

OC and EC increase with temperature nearly everywhere, although generally more weakly than sulfate. The weaker correlation of inert EC vs. sulfate might suggest a chemical influence on the sulfate correlation, but the EC measurements are also subject to larger errors (Chow et al., 2004; Flanagan et al., 2006). The OC correlation is mostly driven by the summer months (170 ng m⁻³ K⁻¹), which may reflect biogenic volatile organic compound (VOC) emissions and wildfires. The Dawson et al. (2007) CTM sensitivity study found an average OC response of -14 ng m⁻³/K in summer and -13 ng m⁻³/K in winter driven by volatility, but they did not account for variability of biogenic VOC emissions or wildfires.

2.3.3. PM_{2.5} components vs. relative humidity

Figure 2.8 shows the deseasonalized relationships of the major PM_{2.5} components with RH, as measured by the MLR coefficient β_2 in Eq. (2.2). The coefficients for sulfate and nitrate are generally positive. For sulfate this likely reflects the dominant source from in-cloud SO₂ oxidation and the association with moist southerly flow shown by the wind patterns in Fig. 2.4. The stronger positive association of nitrate with RH likely reflects the RH dependence of the ammonium nitrate formation equilibrium (Stelson and Seinfeld, 1982). In the agricultural Midwest and Great Plains where ammonia is in excess, production of nitrate can be largely determined by RH (Kleeman, 2008), possibly explaining the particularly strong nitrate-RH correlation there.

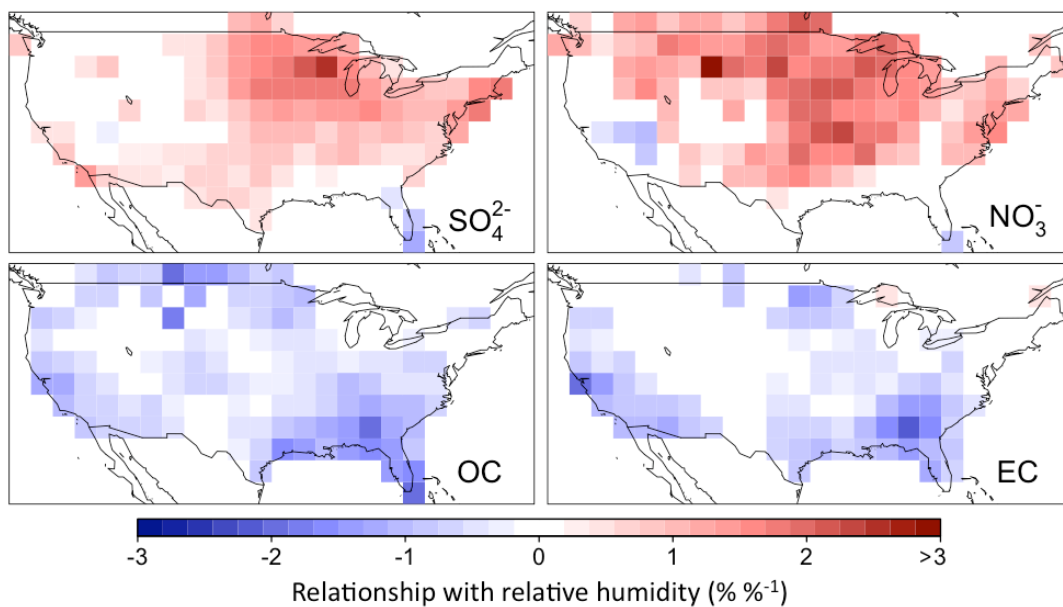


Figure 2.8. Same as Fig. 2.7 but for correlations of $PM_{2.5}$ components with surface air relative humidity.

We find that OC and EC have a negative association with RH, most strongly in the Southeast and the West. This explains the negative association of total $PM_{2.5}$ with RH in these regions (Fig. 2.4). It may reflect the association of low RH with fires, which are major contributors to carbonaceous aerosols in both regions (Park et al., 2007), and also the association of high RH with clean marine air. These factors apparently dominate over any enhanced formation of OC aerosol in aqueous-phase particles at high RH (Volkamer et al., 2007; Fu et al., 2009).

2.3.4. $PM_{2.5}$ components vs. wind direction

Figure 2.9 shows the normalized vector sums of MLR coefficients β_8 and β_9 in Eq. (2.2), which indicate the wind direction most strongly associated with high

concentrations of PM components. This dramatically illustrates the role of SO₂ emissions in the Ohio Valley as a source of sulfate for much of the country. By contrast, nitrate shows a major influence from the agricultural areas in the Midwest and Great Plains with large NH₃ emissions. OC has more distributed sources with some exported influence from the Southeast and the West, likely reflecting biogenic and fire sources (Liao et al., 2007; Park et al., 2007). EC shows little correlation with wind direction except in the Northeast where southwesterly flow carries polluted air.

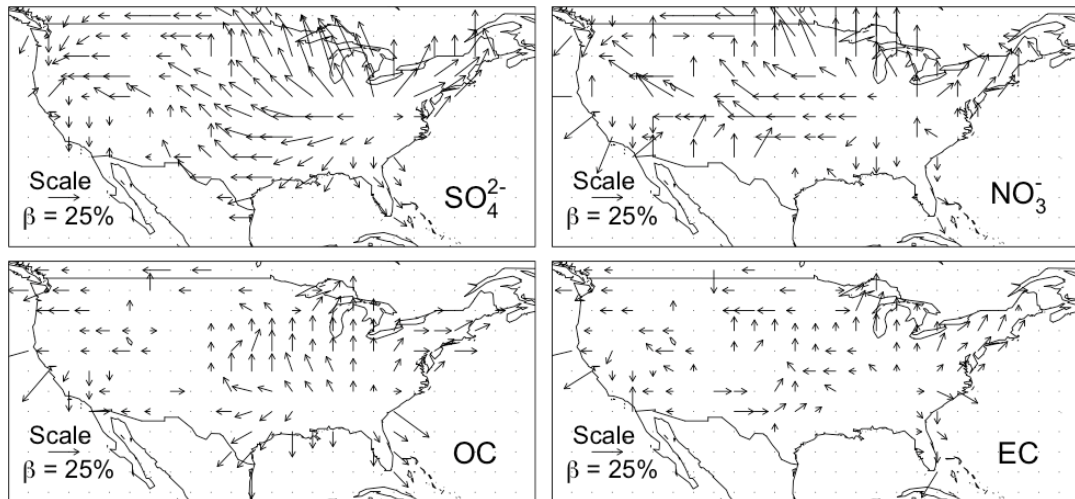


Figure 2.9. Correlations of PM_{2.5} components with wind direction. Figure shows vector sums of multiple linear regression coefficients β_8 and β_9 , normalized to annual mean concentrations of Fig. 2.3. Length of arrows (in units of % per unit sine or cosine) indicates magnitude of correlation. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05).

2.4. Implications for the effects of climate change on air quality

The observed relationships between $PM_{2.5}$ and meteorological variables presented here offer a test of the reliability of GCM-CTM simulations in describing the response of $PM_{2.5}$ to climate change. Our results point to some potential effects of climate change and also to some processes that need to be better represented in CTMs.

The most robust projection for 21st-century climate change is a warming of the surface (Christensen et al., 2007). We find a strong positive correlation of observed $PM_{2.5}$ with temperature driven mainly by sulfate and OC, in contrast to previous CTM sensitivity studies that perturbed temperature only and found a negative response (Aw and Kleeman, 2003; Dawson et al., 2007; Kleeman, 2008). These studies did not account for the correlation of temperature with stagnation or other meteorological conditions, which could play an important role in the observed correlations. But our results also suggest that the temperature dependence of fires and biogenic (including agricultural) emissions of NH_3 , NO_x , and VOCs may play an important role in driving the correlation of $PM_{2.5}$ with temperature and need to be resolved in GCM-CTM studies. Changes in precipitation patterns can obviously affect $PM_{2.5}$ concentrations, as reflected in the negative observed correlation. GCM simulations for the 21st-century climate find a consistent increase in annual mean precipitation in the Northeast and a decrease in the Southwest, but predictions for the rest of the US are less consistent (Christensen et al., 2007). Pye et al. (2009) pointed out that the association of deeper boundary layer mixing with reduced precipitation might represent a compensating effect on $PM_{2.5}$. Models in general find a great sensitivity of $PM_{2.5}$ to mixing depth due to

dilution (Dawson et al., 2007; Kleeman, 2008). Projections of changes in mixing depth for the 21st-century climate are inconsistent across different GCMs (Jacob and Winner, 2009). Mixing depth could either increase or decrease, depending in particular on the changes in soil moisture (Wu et al., 2008).

Increased stagnation in the future climate would cause a corresponding increase in PM_{2.5} levels, as shown in Fig. 2.6. GCMs consistently find more frequent and prolonged stagnation episodes at northern mid-latitudes in the future climate (Mickley et al., 2004; Murazaki and Hess, 2006; Wu et al., 2008). Leibensperger et al. (2008) found for the East in summer a strong anticorrelation between the number of stagnant days and the frequency of mid-latitudes cyclones. They pointed out that mid-latitude cyclone frequency has been decreasing over the 1980-2006 period and attributed this trend to greenhouse warming. Extrapolating their 1980-2006 trend in summer cyclone frequency (-0.15 a^{-1}) to 2050, and using their observed anticorrelation between cyclone frequency and stagnant days, would imply 4.5 more stagnant days per summer in the East by 2050. From our results in Fig. 2.6, this translates to an average increase of $0.24 \mu\text{g m}^{-3}$ in summer mean PM_{2.5} concentrations with a maximum increase of $0.93 \mu\text{g m}^{-3}$ in the Midwest.

2.5. Conclusions

We applied a multiple linear regression (MLR) model to determine the correlations of total fine particulate matter (PM_{2.5}) and its major components with meteorological variables using 1998-2008 daily observations over the contiguous US.

The data were deseasonalized and detrended to focus on synoptic-scale correlations. Our goals were to improve the understanding of the sensitivity of PM_{2.5} to meteorology, and to develop an observational resource that can test the ability of chemical transport models (CTMs) to project the sensitivity of PM_{2.5} to future climate change as simulated by general circulation models (GCMs).

We found that daily variation in meteorology as described by the MLR including nine predictor variables (temperature, relative humidity, precipitation, cloud cover, 850-hPa geopotential height, sea-level pressure tendency, wind speed, E-W and N-S wind direction) can explain up to 50% of daily PM_{2.5} variability in the US. Stagnation is a strong predictor; PM_{2.5} concentrations in the US are on average 2.6 $\mu\text{g m}^{-3}$ higher on a stagnant day vs. non-stagnant day.

Correlations with temperature, RH, and wind direction differ for individual PM_{2.5} components, leading to regional differences in the correlations for total PM_{2.5} depending on the relative abundance of each component. In the case of temperature, correlations of sulfate, organic carbon (OC), and elemental carbon (EC) are predominantly positive, reflecting the joint association with stagnation and cold front ventilation, and with biogenic and fire emissions. Nitrate is negatively correlated with temperature in the South, as expected from the volatility of ammonium nitrate, but positively correlated in California and the Great Plains, which may reflect the temperature dependence of agricultural NH₃ and NO_x emissions.

Relative humidity (RH) is positively correlated with sulfate and nitrate, which may reflect in-cloud sulfate formation and the RH dependence of ammonium nitrate

formation. In contrast, RH is negatively correlated with OC and elemental carbon (EC), possibly reflecting sources from fires.

Correlation with vector winds shows that the industrial Midwest is a source of sulfate for much of the country, and that nitrate is generally highest under inflow from agricultural regions (reflecting NH₃ emissions). There is also some association of elevated OC with flow from regions of elevated biogenic and fire emissions in the Southeast and the West. Perturbations to wind patterns from climate change would thus have a major effect on the distribution and composition of PM_{2.5}.

Our results point to some potential effects of climate change (including changes in temperature, precipitation patterns and stagnation) on future PM air quality, and stress the importance of adequately representing the temperature dependence of agricultural, biogenic and wildfire emissions in GCM-CTM studies.

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Chapter 3. Meteorological modes of variability for fine particulate matter (PM_{2.5}) air quality in the United States: implications for PM_{2.5} sensitivity to climate change

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Abstract

We applied a multiple linear regression model to understand the relationships of PM_{2.5} with meteorological variables in the contiguous US and from there to infer the sensitivity of PM_{2.5} to climate change. We used 2004-2008 PM_{2.5} observations from ~1000 sites (~200 sites for PM_{2.5} components) and compared to results from the GEOS-Chem chemical transport model (CTM). All data were deseasonalized to focus on synoptic-scale correlations. We find strong positive correlations of PM_{2.5} components with temperature in most of the US, except for nitrate in the Southeast where the correlation is negative. Relative humidity (RH) is generally positively correlated with sulfate and nitrate but negatively correlated with organic carbon. GEOS-Chem results indicate that most of the correlations of PM_{2.5} with temperature and RH do not arise from direct dependence but from covariation with synoptic transport. We applied principal component analysis and regression to identify the dominant meteorological

modes controlling $PM_{2.5}$ variability, and show that 20-40% of the observed $PM_{2.5}$ day-to-day variability can be explained by a single dominant meteorological mode: cold frontal passages in the eastern US and maritime inflow in the West. These and other synoptic transport modes drive most of the overall correlations of $PM_{2.5}$ with temperature and RH except in the Southeast. We show that interannual variability of $PM_{2.5}$ in the US Midwest is strongly correlated with cyclone frequency as diagnosed from a spectral-autoregressive analysis of the dominant meteorological mode. An ensemble of five realizations of 1996-2050 climate change with the GISS general circulation model (GCM) using the same climate forcings shows inconsistent trends in cyclone frequency over the Midwest (including in sign), with a likely decrease in cyclone frequency implying an increase in $PM_{2.5}$. Our results demonstrate the need for multiple GCM realizations (because of climate chaos) when diagnosing the effect of climate change on $PM_{2.5}$, and suggest that analysis of meteorological modes of variability provides a computationally more affordable approach for this purpose than coupled GCM-CTM studies.

3.1. Introduction

Air pollution is highly dependent on weather, and it follows that climate change could significantly impact air quality. The pollutants of most public health concern are ozone and fine particulate matter with diameter less than $2.5 \mu m$ ($PM_{2.5}$). Studies using chemical transport models (CTMs) driven by general circulation models (GCMs) consistently project a worsening of ozone air quality in a warming climate (Weaver et

al., 2009). This finding is buttressed by observed correlations of ozone with temperature that are well reproduced by models (Jacob et al., 1993; Sillman and Samson, 1995; Rasmussen et al., 2012). By contrast, GCM-CTM studies of the effect of climate change on PM_{2.5} show no consistency even in the sign of effect (Jacob and Winner, 2009). In previous work (Tai et al., 2010), we examined the observed correlations of PM_{2.5} and its components in the US with meteorological variables as a means to understand PM_{2.5} response to climate change. Here we develop this approach further to define meteorological modes of variability for PM_{2.5} and interpret the observed correlations and modes using the GEOS-Chem CTM. We apply the Goddard Institute for Space Studies (GISS) GCM to illustrate how the modes enable effective diagnosis of the effect of climate change on PM_{2.5}.

The uncertainty in assessing climatic effects on PM_{2.5} reflects the complex dependence of different PM_{2.5} components on meteorological variables. Higher temperatures can lead to higher sulfate concentrations due to faster SO₂ oxidation, but to lower nitrate and organic components due to volatility (Sheehan and Bowman, 2001; Aw and Kleeman, 2003; Dawson et al., 2007; Kleeman, 2008). Biogenic emissions of PM_{2.5} precursors including agricultural ammonia, soil NO_x, and volatile organic compounds (VOCs) increase with temperature and further complicate the PM_{2.5}-temperature relationship (Pinder et al., 2004; Bertram et al., 2005; Guenther et al., 2006). Higher relative humidity (RH) promotes aqueous-phase sulfate production and ammonium nitrate formation (Koch et al., 2003; Liao et al., 2006; Dawson et al., 2007), but inhibits fires, which are important contributors to organic aerosols in many regions

(Park et al., 2007; Spracklen et al., 2009). Changes in precipitation and in planetary boundary layer (PBL) depth have a consistent effect on PM_{2.5} components but their projections in GCMs are highly uncertain (Jacob and Winner, 2009).

Synoptic-scale transport should be an important factor driving the effect of climate change on PM_{2.5}. Previous studies have used principal component analysis (PCA) to identify important meteorological modes of variability for PM_{2.5} air quality (Cheng et al., 2007; Thishan Dharshana et al., 2010). Thishan Dharshana et al. (2010) found that as much as 30% of PM_{2.5} daily variability in the US Midwest is associated with passages of synoptic weather systems. Cold fronts associated with mid-latitude cyclone passages provide the dominant ventilation pathway for the eastern US (Cooper et al., 2001; Li et al., 2005). A general reduction in the frequency of these cyclones is expected as a result of greenhouse warming (Lambert and Fyfe, 2006; Christensen et al., 2007; Pinto et al., 2007), potentially leading to more frequent and prolonged stagnation episodes (Mickley et al., 2004; Murazaki and Hess, 2006). Leibensperger et al. (2008) found a strong anticorrelation between summer cyclone frequency and ozone pollution in the eastern US for 1980-2006, and further showed evidence of a long-term decline in cyclone frequency over that period that significantly hindered attainment of ozone air quality standards. Tai et al. (2010) projected a PM_{2.5} enhancement of up to 1 $\mu\text{g m}^{-3}$ in the Midwest from 2000-2050 climate change due to more frequent stagnation.

In this study, we first apply the GEOS-Chem global CTM to interpret the observed correlations between PM_{2.5} components and meteorological variables in the contiguous US. As we will see, interpretation is complicated by the covariation of

meteorological variables with synoptic transport. To address this issue, we use PCA and regression to determine the dominant meteorological modes of observed daily $PM_{2.5}$ variability in different US regions, and show how spectral analysis of these modes enables a robust estimate of the effect of climate change on $PM_{2.5}$ air quality.

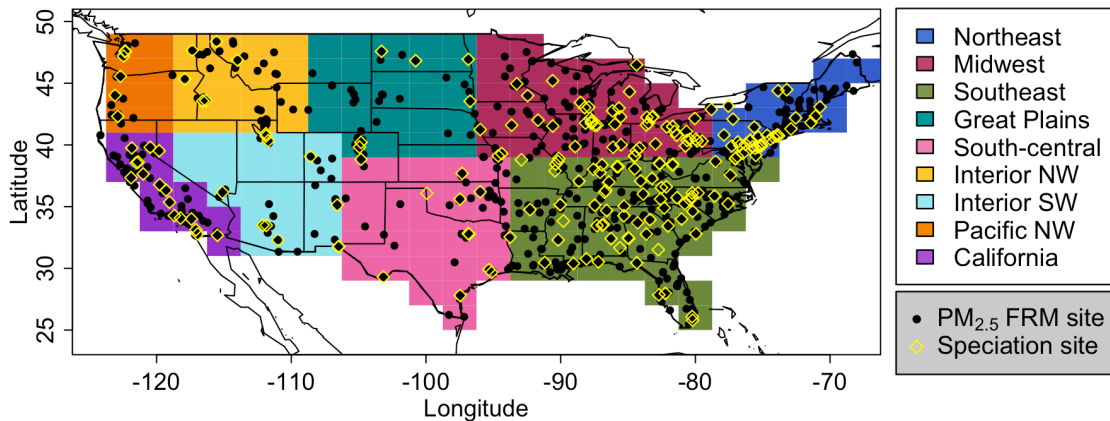


Figure 3.1. US regions used to study the correlations of $PM_{2.5}$ with meteorological modes of variability. Also shown are the EPA Air Quality System (AQS) $PM_{2.5}$ monitoring sites in 2006, including total $PM_{2.5}$ monitors using the Federal Reference Method (FRM) and chemical speciation monitors from the SLAMS + STN networks.

3.2. Data and models

3.2.1. $PM_{2.5}$ observations

Daily mean surface concentrations of total $PM_{2.5}$ and speciated components including sulfate, nitrate, and organic carbon (OC) for 2004-2008 were obtained from the ensemble of sites of the EPA Air Quality System (EPA-AQS)

(<http://www.epa.gov/ttn/airs/airsaqs/>), shown in Fig. 3.1. Total $PM_{2.5}$ data are from the

Federal Reference Method (FRM) network of about 1000 sites in the contiguous US. Speciation data are from the State and Local Air Monitoring Stations (SLAMS) and Speciation Trends Network (STN) of about 200 sites. These sites measure every one, three or six days. Tai et al. (2010) show maps of the annual mean data for total PM_{2.5} (1998-2008) and individual components (2000-2008). We do not discuss ammonium and elemental carbon (EC) here because ammonium is mainly the counter-ion for sulfate and nitrate, and the correlation patterns of EC with meteorological variables generally follow those of OC (Tai et al., 2010).

3.2.2. GEOS-Chem simulations

We used the GEOS-Chem global CTM to conduct full-year simulations of coupled gas-phase and aerosol chemistry. GEOS-Chem (<http://geos-chem.org>) uses assimilated meteorological data from the NASA Global Earth Observing System (GEOS-5) with 6-h temporal resolution (3-h for surface variables and PBL depth), 0.5° latitude by 0.667° longitude (0.5°×0.667°) horizontal resolution, and 47 hybrid pressure-sigma vertical levels. We conducted GEOS-Chem simulations at three different horizontal resolutions: native 0.5°×0.667°, 2°×2.5°, and 4°×5°. The coarser resolutions have been used previously with meteorological fields from the GISS GCM to investigate effects of climate change on air quality (Wu et al., 2008; Pye et al., 2009; Leibensperger et al., 2011a). For the native resolution simulation we used a nested continental version of GEOS-Chem over North America (140-40°W, 10-70°N) with 2°×2.5° resolution for the rest of the world (Chen et al., 2009; Zhang et al., 2011). The

native simulation was conducted for one year (2006) and the $2^{\circ} \times 2.5^{\circ}$ and $4^{\circ} \times 5^{\circ}$ simulations for three years (2005-2007) using GEOS-Chem version 8-3-2. We included a non-local PBL mixing scheme formulated by Holtslag and Boville (1993) and implemented in GEOS-Chem by Lin and McElroy (2010).

GEOS-Chem includes a fully coupled treatment of tropospheric ozone- NO_x -VOC-aerosol chemistry (Park et al., 2004; Liao et al., 2007). Gas-aerosol phase partitioning of the sulfate-nitrate-ammonium-water system is calculated using the ISORROPIA II thermodynamic equilibrium model (Fountoukis and Nenes, 2007). In-cloud SO_2 oxidation uses liquid water content information from the GEOS-5 archive (Fisher et al., 2011). Secondary organic aerosol (SOA) formation is computed with a standard mechanism based on reversible gas-aerosol partitioning of semi-volatile VOC oxidation products (Chung and Seinfeld, 2002). SOA precursors include isoprene, terpenes, and aromatic hydrocarbons (Henze et al., 2008).

Anthropogenic emissions of sulfur, ammonia and NO_x emissions in the US are from the EPA 2005 National Emissions Inventory (<http://www.epa.gov/ttn/chief/net/2005inventory.html>), and primary anthropogenic OC and EC emissions are from Cooke et al. (1999). Non-US anthropogenic emissions are described by Park et al. (2006). Biomass burning emissions of OC and EC are from the Global Fire Emissions Database (GFED v2) (Giglio et al., 2006). These emissions are included in the model as monthly averages and do not contribute to day-to-day variability of $\text{PM}_{2.5}$. In contrast, soil NO_x emissions (Yienger and Levy, 1995) and biogenic emissions of isoprene, terpenes, and methylbutenol (Guenther et al., 2006) are

updated locally every three hours as a function of temperature, solar radiation, and precipitation. Scavenging of PM_{2.5} by precipitation follows the scheme of Liu et al. (2001). Dry deposition follows a standard resistance-in-series scheme (Wesely, 1989) as implemented by Wang et al. (1998).

Total PM_{2.5} in GEOS-Chem is taken to be the sum of sulfate, nitrate, ammonium, OC and EC. Detailed evaluations of the GEOS-Chem simulation of PM_{2.5} and its components over the US have been presented in a number of publications using observations from surface sites, aircraft, and satellites (Heald et al., 2006; Park et al., 2006; van Donkelaar et al., 2006; Heald et al., 2008; van Donkelaar et al., 2008; Fu et al., 2009; Drury et al., 2010; Leibensperger et al., 2011a; Zhang et al., 2012). These evaluations mainly focused on seasonal concentrations and showed no prominent biases. Here we will focus on the ability of the model to reproduce observed correlations of PM_{2.5} with meteorological variables.

3.2.3. Multiple linear regression

We examined the correlations of PM_{2.5} and its components with meteorological variables for 2004-2008 (EPA-AQS) and 2005-2007 (GEOS-Chem) by applying a standardized multiple linear regression (MLR) model:

$$\frac{y(t) - \bar{y}}{s_y} = \sum_{k=1}^8 \beta_k \frac{x_k(t) - \bar{x}_k}{s_k} \quad (3.1)$$

where y represents the deseasonalized daily PM_{2.5} concentration (total PM_{2.5} or individual component), x_k represents the eight deseasonalized meteorological variables from GEOS-5 listed in Table 3.1, \bar{x}_k and \bar{y} are the temporal means of x_k and y , s_k and

s_y are their standard deviations, β_k is the dimensionless, normalized regression coefficient, and t is time. To compare observed with simulated correlations, we interpolate the EPA-AQS data onto the GEOS-Chem grid (Tai et al., 2010) and use the interpolated PM_{2.5} fields for regression.

The MLR model is applied to each individual grid cell for both the observed and simulated PM_{2.5} fields. All data (x_k and y) are deseasonalized and detrended by subtracting the 30-day moving averages from the original data so that $\bar{x}_k = \bar{y} = 0$. This allows us to focus on synoptic-scale variability and avoid aliasing from common seasonal or interannual variations. The standardized regression coefficients β_k allow direct comparisons between the correlations of different PM_{2.5} components with different meteorological variables (Kutner et al., 2004). The original regression coefficients β_k^* in units of $\mu\text{g m}^{-3} \text{D}^{-1}$, where D is the dimension of meteorological variable x_k in Table 3.1, can be recovered by

$$\beta_k^* = \frac{s_y}{s_k} \beta_k \quad (3.2)$$

The observed coefficients of determination (R^2) for the MLR model have values ranging from 0.1 (in the west-central US where data are sparse) to 0.5 (in the Midwest and Northeast), agreeing with previous studies (Wise and Comrie, 2005; Tai et al., 2010). In addition to the standardized MLR analysis, we also conducted a stepwise MLR analysis with interaction terms as described by Tai et al. (2010). The interaction terms were generally found to be insignificant.

Table 3.1. Meteorological variables used for PM_{2.5} correlation analysis. ^a

Variable	Meteorological parameter
x_1	Surface air temperature (K) ^b
x_2	Surface air relative humidity (%) ^b
x_3	Surface precipitation (mm d ⁻¹)
x_4	Geopotential height at 850 hPa (km)
x_5	Sea level pressure tendency $dSLP/dt$ (hPa d ⁻¹)
x_6	Surface wind speed (m s ⁻¹) ^{b, c}
x_7	East-west wind direction indicator $\cos\theta$ (dimensionless) ^d
x_8	North-south wind direction indicator $\sin\theta$ (dimensionless) ^d

a. Assimilated meteorological data with $0.5^\circ \times 0.667^\circ$ horizontal resolution from the NASA Goddard Earth Observing System (GEOS-5). All data used are 24-h averages, and are deseasonalized and detrended as described in the text.

b. At 6 m above the surface (0.994 sigma level).

c. Calculated from the horizontal wind vectors (u , v).

d. θ is the angle of the horizontal wind vector counterclockwise from the east. Positive values of x_7 and x_8 indicate westerly and southerly winds, respectively.

We conducted the MLR analysis for the model at all three resolutions ($0.5^\circ \times 0.667^\circ$, $2^\circ \times 2.5^\circ$, $4^\circ \times 5^\circ$) and found the patterns of correlations to be similar. Figure 3.2 shows as an example (to be discussed later) the simulated and observed relationships of nitrate with temperature as measured by the recovered regression coefficient β_1^* in Eq. (3.2). In general, $2^\circ \times 2.5^\circ$ and $4^\circ \times 5^\circ$ regression results agree well with each other for all meteorological variables and all components. The native-

resolution regression does not show as extensive and significant correlations. A likely explanation is that averaging over larger grid cells smooths out local effects, yielding more robust correlation statistics. We will use $2^\circ \times 2.5^\circ$ resolution for model-observation comparisons in what follows.

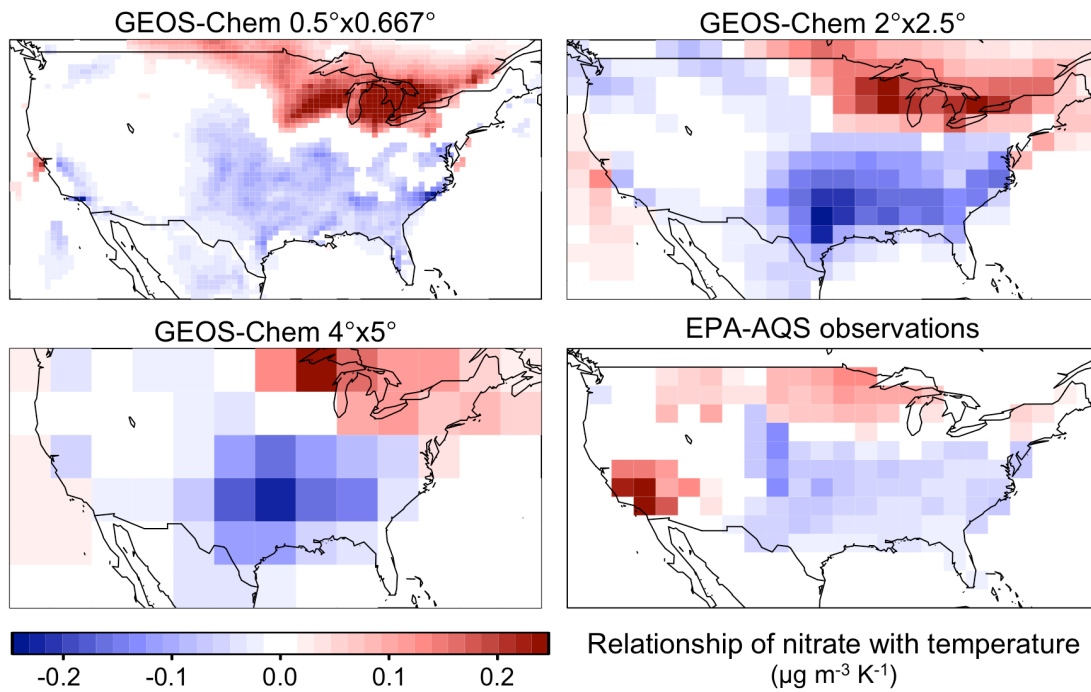


Figure 3.2. Simulated (2005-2007) and observed (2004-2008) relationships of nitrate $\text{PM}_{2.5}$ with surface air temperature, as measured by the multiple linear regression coefficient β_1^* in Eq. (3.2) with units of $\mu\text{g m}^{-3} \text{K}^{-1}$. Simulated relationships are shown for three different GEOS-Chem model resolutions: $0.5^\circ \times 0.667^\circ$, $2^\circ \times 2.5^\circ$ and $4^\circ \times 5^\circ$. Observations are averaged over the $2^\circ \times 2.5^\circ$ grid. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05).

3.3. Correlations of PM_{2.5} with meteorological variables

3.3.1. Correlations with temperature

Figure 3.3 (left and middle panels) shows the observed and simulated relationships of sulfate, nitrate, and OC with temperature as measured by the standardized regression coefficient β_1 in Eq. (3.1). The relationships may reflect both a direct dependence of PM_{2.5} on temperature and a covariation of temperature with other meteorological variables affecting PM_{2.5}. To separate the two effects, we conducted a direct sensitivity analysis with GEOS-Chem by increasing temperatures by 1 K throughout the troposphere while keeping all other meteorological variables constant. The resulting sensitivities are shown in the right panels of Fig. 3.3, normalized to the standard deviations of deseasonalized concentrations and temperature to make them directly comparable to the standardized regression coefficients β_1 in the left and middle panels.

Sulfate in the observations shows a positive relationship with temperature over most of the US. The model is generally consistent with the observations but does not capture the Southwest maximum. Results from the direct sensitivity analysis, however, show a generally negative dependence of sulfate on temperature particularly in the West. This contrasts with a previous CTM sensitivity analysis by Dawson et al. (2007) that found a positive dependence of sulfate on temperature, though much weaker than the observed relationship (Tai et al., 2010). Dawson et al. (2007) attributed their result to faster SO₂ oxidation kinetics at higher temperature, but we find in GEOS-Chem that this is more than offset by the increased volatility of H₂O₂ and SO₂, slowing down the

in-cloud aqueous-phase production of sulfate. In any case, it is clear from the model that the observed positive relationship of sulfate with temperature must reflect covariation of temperature with meteorological variables rather than a direct dependence. We elaborate on this in Section 3.4.

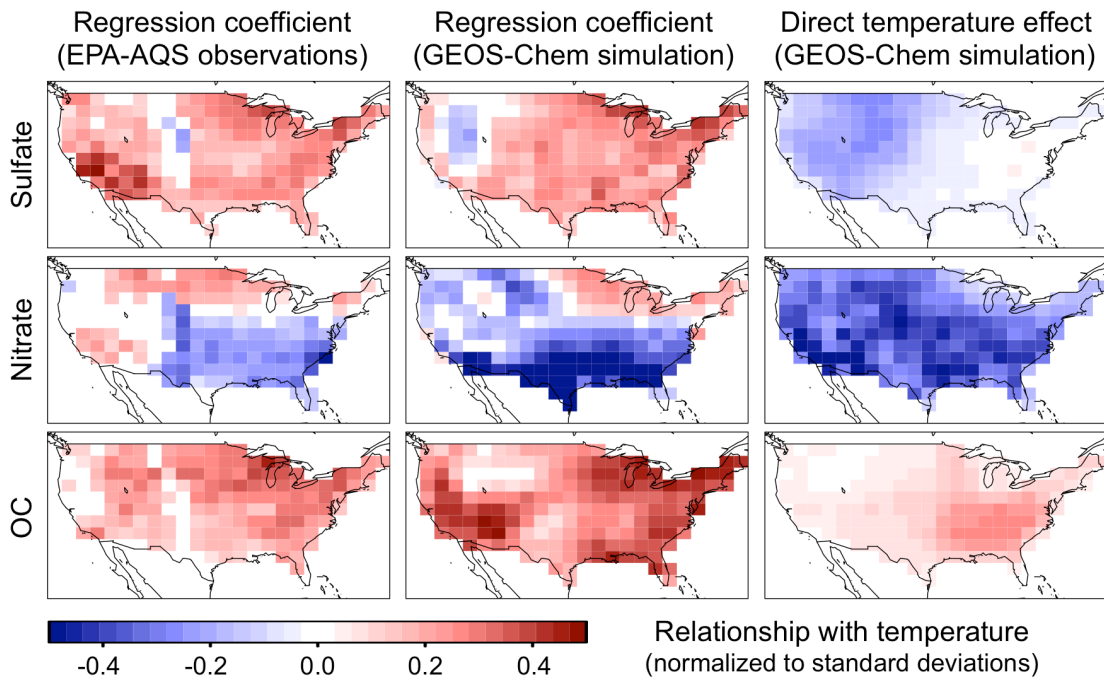


Figure 3.3. Relationships of sulfate, nitrate, and organic carbon (OC) $PM_{2.5}$ concentrations with surface air temperature. The left and middle panels show the observed (2004-2008) and simulated (2005-2007) standardized regression coefficients β_1 in Eq. (3.1). Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05). The right panels show the direct effects of temperature on sulfate, nitrate and OC as determined by applying a global +1 K temperature perturbation in the GEOS-Chem simulation, and normalizing the results to the standard deviations of deseasonalized concentrations and temperatures to allow direct comparison to β_1 .

Nitrate in the observations shows a negative relationship with temperature in the Southeast but a positive relationship in the North and the Southwest. The model reproduces these results except for the positive relationship in the Southwest. The negative relationship in the model is too strong in the South but the higher-resolution $0.5^\circ \times 0.667^\circ$ simulation does not show such a bias (Fig. 3.2). The direct sensitivity of nitrate to temperature in the model is negative everywhere, with magnitude comparable to that found by Dawson et al. (2007), and reflecting the volatility of ammonium nitrate (Stelson and Seinfeld, 1982). We see from Fig. 3.3 that this direct dependence could account for most of the observed negative relationship of nitrate with temperature in the Southeast, but it is more than offset in the North by the positive association of temperature with southerly flow importing polluted air. The observed positive relationship of nitrate with temperature in the Southwest may reflect the temperature dependence of ammonia and fire emissions; in the model these emissions are specified as monthly means.

OC in the observations shows a positive relationship with temperature throughout the US, and the same is found in the model although the relationship is steeper. The direct sensitivity study in the model also shows a positive dependence of OC on temperature. Dawson et al. (2007) previously found a negative dependence due to OC volatility but did not consider the temperature dependence of biogenic VOC emissions, which is included in our analysis and more than offsets the volatility effect. Day and Pandis (2011) similarly found an increase in OC at higher temperatures mainly due to increased VOC emissions. We see from Fig. 3.3 that the direct temperature

dependence may be a significant contributor to the positive relationship between OC and temperature in the Southeast, where biogenic emissions are particularly high, but it has little effect elsewhere.

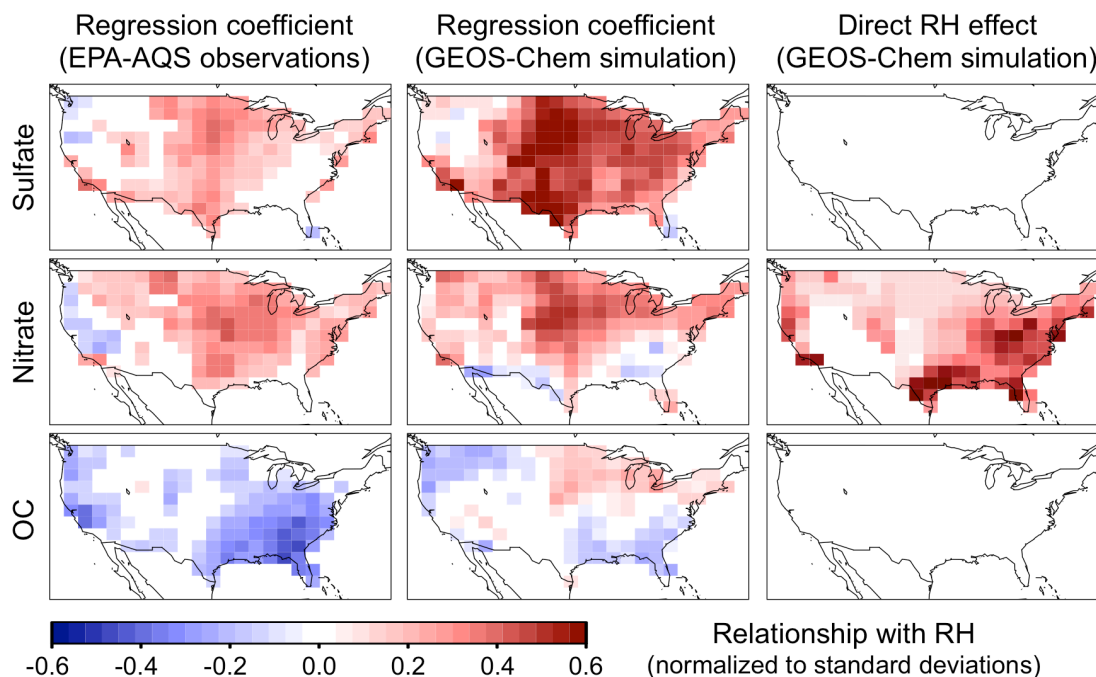


Figure 3.4. Same as Fig. 3.3 but for relative humidity (RH). The right panels show the direct effects of RH as determined by applying a global -1 % RH perturbation in the GEOS-Chem simulation.

3.3.2. Correlations with relative humidity

Figure 3.4 shows the observed and simulated correlations of sulfate, nitrate, and OC with RH, expressed as the standardized regression coefficient β_2 in Eq. (3.1). The relationships are generally positive for sulfate and nitrate both in the observations and

the model. The OC-RH relationship is generally negative with some model biases in the Great Plains and Midwest. Results from a model perturbation simulation similar to that for temperature are also shown in Fig. 3.4, indicating negligible direct dependence of sulfate and OC on RH, but a significant positive relationship for nitrate due to more favorable ammonium nitrate formation at higher RH (Stelson and Seinfeld, 1982). The direct positive sensitivity of nitrate in the southeastern coast is offset by the negative influence from the association of high RH with clean marine air, leading to the weak overall correlation there.

3.3.3. Correlations with precipitation and wind speed

Figure 3.5 shows the observed and simulated relationships of total $PM_{2.5}$ with precipitation and wind speed as measured by β_3 and β_6 in Eq. (3.1). Similar effects are found for all individual $PM_{2.5}$ components (Tai et al., 2010). The observations show strong negative relationships reflecting aerosol scavenging and ventilation. These are generally well captured by the model. The precipitation effect appears to be primarily driven by large-scale rather than convective precipitation in the US. Fang et al. (2011) similarly illustrated the dominance of large-scale precipitation in wet scavenging of soluble pollutants.

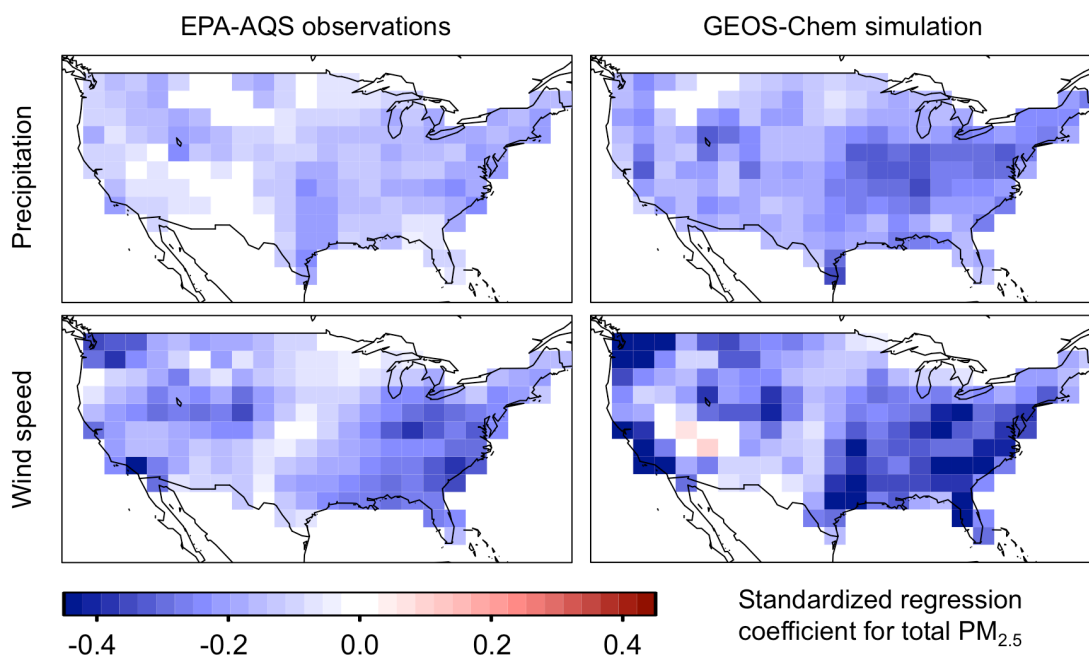


Figure 3.5. Relationships of total $\text{PM}_{2.5}$ concentrations with precipitation and wind speed, expressed as the standardized regression coefficients β_3 and β_6 , respectively. The left panels show observations (2004-2008) and the right panels model values (2005-2007). Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p -value < 0.05).

3.4. Major meteorological modes controlling $\text{PM}_{2.5}$ variability

Results from the previous section show that much of the correlation of $\text{PM}_{2.5}$ with individual meteorological variables is driven by covariance between meteorological variables, with an apparent major contribution from synoptic transport. To resolve this covariance we turn to principal component analysis (PCA) of the meteorological variables to identify the meteorological modes controlling $\text{PM}_{2.5}$ variability.

3.4.1. Principal component analysis and regression

We conducted a PCA for the 2004-2008 GEOS-5 data by averaging spatially over each region of Fig. 3.1 the eight deseasonalized meteorological variables of Table 3.1. The resulting time series for each region were decomposed to produce time series of eight orthogonal principal components (PCs) (U_1, \dots, U_8):

$$U_j(t) = \sum_{k=1}^8 \alpha_{kj} \frac{X_k(t) - \bar{X}_k}{s_k} \quad (3.3)$$

where X_k represents the regionally averaged GEOS-5 variable, \bar{X}_k and s_k the temporal mean and standard deviation of X_k , and α_{kj} the elements of the orthogonal transformation matrix. Each PC represents a distinct meteorological regime or mode. We identified the nature of meteorological mode by examining the values of α_{kj} in Eq. (3.3). PCs with high $|\alpha_{kj}|$ values (e.g., greater than 0.3 and topping the other $|\alpha_{kj}|$ values) for geopotential height, pressure tendency, and wind direction are presumably associated with synoptic-scale weather systems, and can be referred to as synoptic transport modes. We then followed $U_j(t)$ day by day and visually examined the corresponding weather maps for multiple months during 2004-2008. From this we assigned a generalized meteorological feature for a given PC when the same feature could be associated with the majority of peaks and troughs of $U_j(t)$. The PCs are ranked by their variances, usually with the leading three or four PCs capturing most of the meteorological variability. For instance, in the eastern US, a single mode representing

cyclone and cold frontal passages (discussed further in Section 3.4.2) typically accounts for ~20% of total meteorological variability.

We then applied a principal component regression (PCR) model to correlate observed and simulated PM_{2.5} concentrations with the eight PCs for each region

$$\frac{Y(t) - \bar{Y}}{s_Y} = \sum_{j=1}^8 \gamma_j U_j(t) \quad (3.4)$$

where Y represents the regionally averaged PM_{2.5} concentration, γ_j the PC regression coefficients, and \bar{Y} and s_Y the temporal mean and standard deviation of Y . The ratio of regression to total sum of squares (SSR _{j} /SST) for each PC is calculated by

$$\frac{\text{SSR}_j}{\text{SST}} = \frac{\sum_t [\gamma_j U_j(t)]^2}{\sum_t \left\{ \left[\frac{Y(t) - \bar{Y}}{s_Y} \right]^2 \right\}} \quad (3.5)$$

where the summation is over the entire time series $Y(t)$ and $U_j(t)$. This ratio quantifies the fraction of variance of PM_{2.5} that can be explained by a single PC. From Eq. (3.3) and (3.4), the fraction (f_k) of the overall correlation of PM_{2.5} with a given meteorological variable X_k (e.g., in Fig. 3.4 through 3.6) that is associated with a particular PC can be estimated by

$$f_k = \frac{\alpha_{kj} \gamma_j}{\sum_m \alpha_{km} \gamma_m} \quad (3.6)$$

where the summation is over the m PCs that have a significant effect on PM_{2.5} (p -value < 0.01). Here the denominator represents the total effect of X_k on PM_{2.5} that is equivalent to a regionally averaged version of β_k in Eq. (3.1). The PCR model was applied to both the full-year data and to seasonal subsets.

3.4.2. Dominant meteorological modes of $PM_{2.5}$ variability

Figure 3.6 shows as an example the dominant meteorological mode contributing to total $PM_{2.5}$ variability in the Midwest as determined by the highest SSR_j/SST ratio in Eq. (3.5). Based on the PCR model this mode alone explains 29% of the observed $PM_{2.5}$ variability with a regression coefficient $\gamma_j = -0.41$. The top panel of Fig. 6 shows the time series of this mode for January 2006 together with the deseasonalized observed total $PM_{2.5}$ concentrations, illustrating strong anticorrelation ($r = -0.54$). The bottom left panel shows the meteorological composition of this dominant mode as measured by PC coefficients α_{kj} in Eq. (3.3), consisting of low temperature, high precipitation, low and rising pressure, and strong northwesterly winds. From weather maps we can verify that high positive values of this PC represent the center of an eastward-propagating mid-latitude cyclone with a precipitating cold front at the southwest tail end. High negative values indicate the “opposite” regime – warm and dry stagnant condition at the tail end of an anticyclone. Figure 3.6 (top and bottom right) shows, for instance, that as $U_j(t)$ rose from a minimum to maximum between 28 and 30 January 2006 in the Midwest, a mid-latitude cyclone was approaching and the associated cold front swept over the region bringing down total $PM_{2.5}$ by $9 \mu\text{g m}^{-3}$.

Figure 3.7 shows as another example the dominant meteorological mode of $PM_{2.5}$ variability in California, demonstrating again a strong anticorrelation between the time series of this mode and $PM_{2.5}$ concentrations ($r = -0.80$). This mode has similar meteorological composition to that in Fig. 3.6 except for wind direction. Positive phases of this mode represent ventilation by cold maritime inflows associated with synoptic

disturbances, whereas negative phases represent warm, stagnant conditions associated with high-pressure systems. The bottom panel shows, for instance, that between 6 and 8 January 2005, a precipitating maritime inflow reduced $PM_{2.5}$ by $16 \mu\text{g m}^{-3}$.

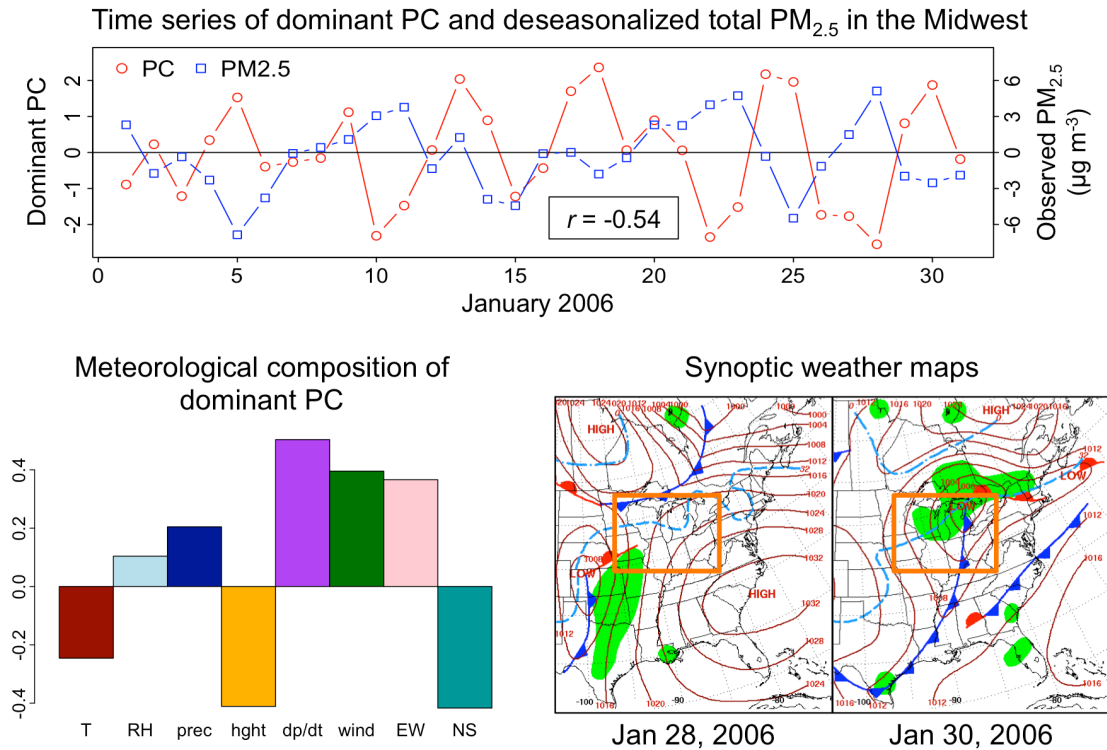


Figure 3.6. Dominant meteorological mode for observed $PM_{2.5}$ variability in the Midwest inferred from the principal component analysis. Top panel: time series of deseasonalized observed total $PM_{2.5}$ concentrations and the dominant meteorological mode or principal component (PC) in January 2006. Bottom left: composition of this dominant mode as measured by the coefficients α_{ki} in Eq. (3.3). Meteorological variables (x_k) are listed in Table 3.1. Bottom right: synoptic weather maps from NCEP (<http://www.hpc.ncep.noaa.gov/dailywxmap/>) for 28 and 30 January, corresponding to maximum negative and positive influences from the principal component. The Midwest is delineated in orange.

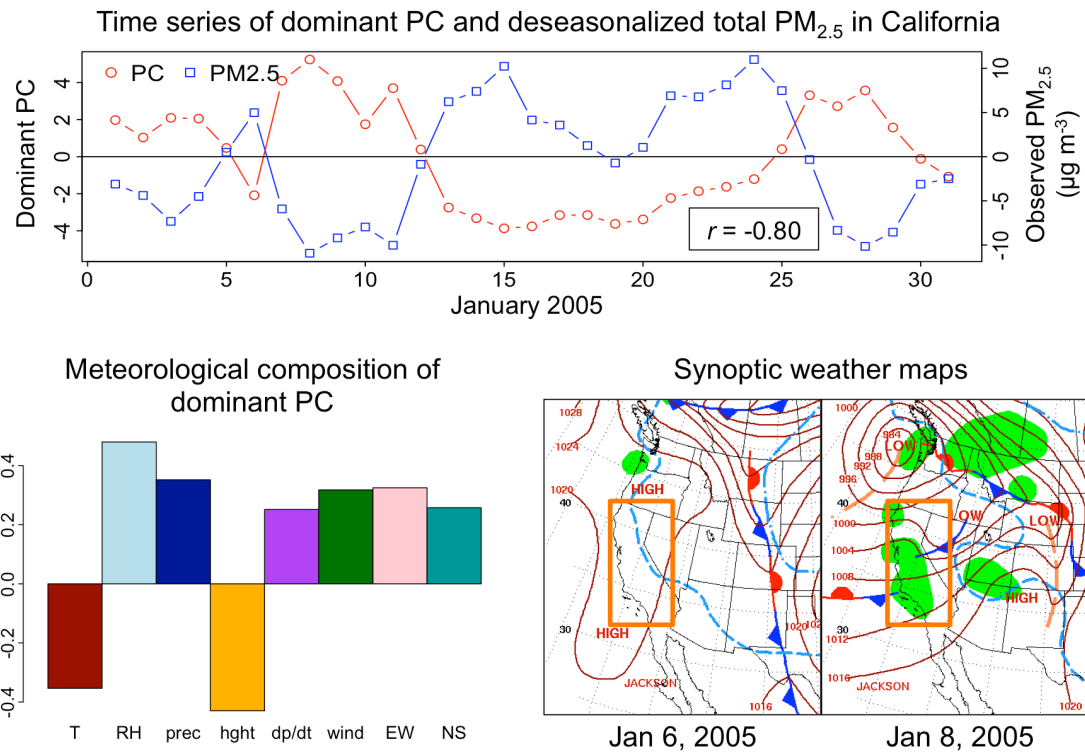


Figure 3.7. Same as Fig. 3.6 but for California.

The analysis above was conducted for all regions of Fig. 3.1. Table 3.2 summarizes the characteristics of the dominant PC controlling PM_{2.5} variability for five selected regions. In the eastern US (Northeast, Midwest and Southeast), the observed dominant modes resemble that for the Midwest described above (Fig. 3.6). In the Northeast, another mode representing southwesterlies associated with high pressure over the western North Atlantic is equally important. In the Pacific Northwest, the dominant mode resembles that for California (Fig. 3.7). In general, the PCR results illustrate the importance of synoptic-scale transport in controlling the observed daily variability of PM_{2.5}. As shown in Table 3.2, this control appears to be well represented

in GEOS-Chem, supporting the ability of the model to describe the variability in $PM_{2.5}$ associated with this transport.

Table 3.2. Dominant meteorological modes for regional $PM_{2.5}$ variability.

US Region	PM _{2.5} variability explained ^a		PC regression coefficient γ_j ^b		Description ^c
	EPA-AQS	GEOS-Chem	EPA-AQS	GEOS-Chem	
Northeast	17%	21%	-0.31	-0.33	Cold front associated with mid-latitude cyclone
Midwest	29%	25%	-0.41	-0.38	
Southeast	31%	15%	-0.42	-0.29	
Pacific Northwest	36%	45%	-0.35	-0.39	Synoptic-scale maritime
California	26%	13%	-0.28	-0.21	inflow

a. From Eq. (3.5).

b. From Eq. (3.4).

c. For positive phases of the dominant PC.

Using Eq. (3.6), we find overall that the synoptic transport modes account for more than 70% of the observed correlations of $PM_{2.5}$ components with temperature in the Northeast and Midwest. This reflects the association of elevated temperature with southerly flow and stagnation. In the Southeast, however, we find that more than 60% of the observed correlations of nitrate and OC with temperature and RH arise from a single non-transport mode consisting of low temperature and high RH. Nitrate has a positive dependence on that mode because of ammonium nitrate thermodynamics, while

OC has a negative dependence reflecting biogenic VOC emissions and the occurrence of fires. The weaker importance of transport in driving the nitrate-temperature relationship in the Southeast likely reflects the lower frequency of cold fronts. In California, the transport and non-transport modes are comparably important in shaping the observed correlations of PM_{2.5} components with temperature and RH.

3.5. Cyclone frequency as a metric for climate change effect on PM_{2.5}

Mid-latitude cyclones and their associated cold fronts are known to provide the dominant year-round mechanism for ventilating the US Midwest and Northeast (Cooper et al., 2001; Li et al., 2005), and they emerge in our analysis of Section 3.4 as the dominant meteorological mode of PM_{2.5} variability. Previous studies diagnosing cyclone frequency have relied on identifying local pressure minima (Mickley et al., 2004; Lambert and Fyfe, 2006; Lang and Waugh, 2011) or used storm tracking algorithms (Geng and Sugi, 2001; Bauer and Del Genio, 2006; Bengtsson et al., 2006). Here we diagnose cyclone frequency by applying a fast Fourier transform (FFT) to the time series of the dominant Midwest PC representing cyclone and frontal passages as shown in Fig. 3.6. We use 1999-2010 meteorological data from the NCEP/NCAR Reanalysis 1 (Kalnay et al., 1996; Kistler et al., 2001), which provides a longer record than GEOS-5. PCA of the NCEP/NCAR data yields essentially the same meteorological modes as GEOS-5. Figure 3.8 (gray thin line) shows the FFT spectrum for the dominant cyclone mode in the Midwest for 1999-2010. The low-frequency structure (with periods > 20 d) is an artifact of the 30-day moving average applied to the meteorological data to

remove seasonality. We smooth the time series with a second-order autoregressive (AR2) filter (Wilks, 2006), indicating a median spectral frequency of 52 a^{-1} (cyclone period of about 7 days).

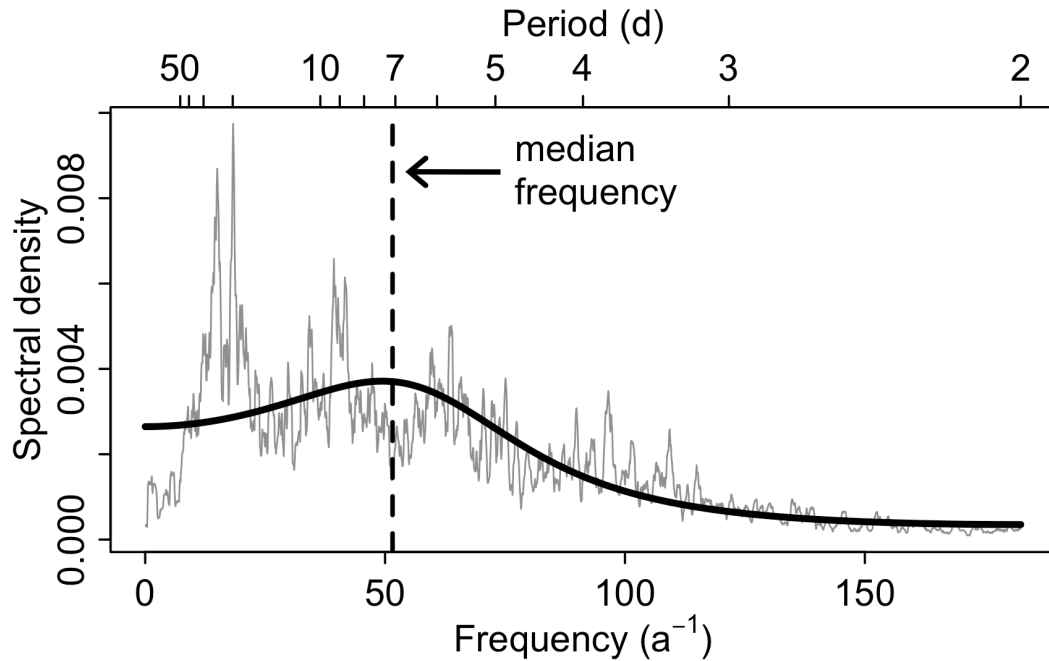


Figure 3.8. Frequency spectrum of the daily time series of the dominant meteorological mode (cyclone/frontal passages) in the US Midwest (Fig. 3.1) for 1999-2010 using NCEP/NCAR Reanalysis 1 data. The thin line shows the fast Fourier transform (FFT) spectrum and the thick line shows the smoothed spectrum from a second-order autoregressive (AR2) model. The vertical dashed line indicates the median AR2 spectral frequency used as a metric of cyclone frequency.

We applied the spectral-autoregressive method above to find the median cyclone frequencies and periods for individual years of the 1999-2010 record. Figure 3.9 shows the time series of annual mean anomalies in total PM_{2.5} concentrations and cyclone periods for the Midwest, where the correlation is strongest ($r = 0.76$) corresponding to a PM_{2.5}-to-cyclone period sensitivity of $0.94 \pm 0.43 \mu\text{g m}^{-3} \text{d}^{-1}$ (95% confidence interval). Leibensperger et al. (2008) previously found a strong interannual correlation of summer ozone with cyclone frequency in the Northeast using the 1980-2006 record of NCEP/NCAR data. Our analysis does not show the same for PM_{2.5} in this region, possibly because of the short record (12 years) available for PM_{2.5}. Cyclone frequencies found by Leibensperger et al. (2008) are generally lower, possibly because their storm-tracking algorithm may neglect weaker cyclones and fronts.

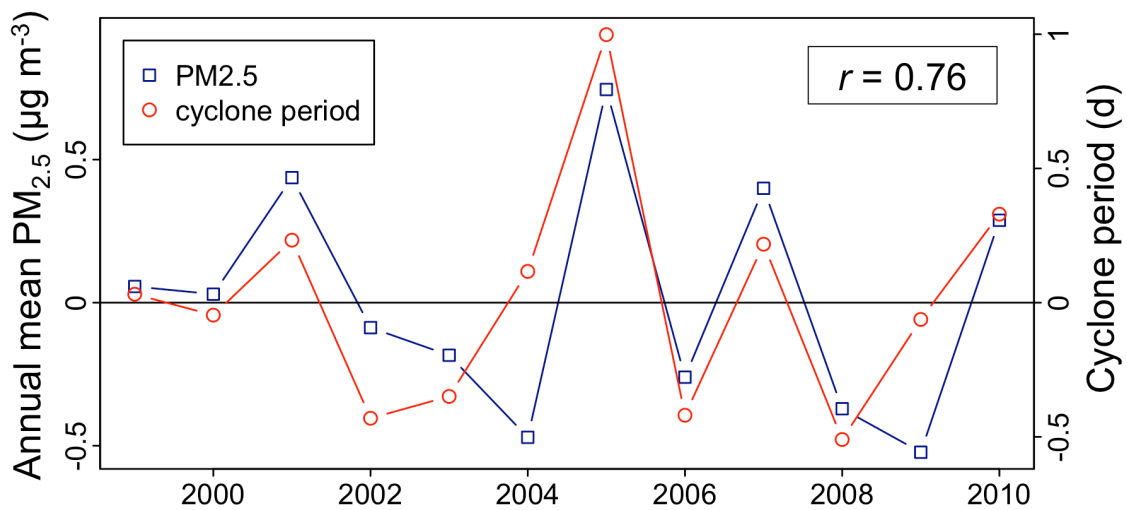


Figure 3.9. Anomalies of annual mean PM_{2.5} concentrations and median cyclone periods for the US Midwest (Fig. 3.1).

The strong interannual correlation of $PM_{2.5}$ with cyclone frequency, at least in the Midwest, encourages the use of cyclone frequency as a metric to diagnose the effect of climate change on $PM_{2.5}$. We used for this purpose an ensemble of five realizations of 1950-2050 climate change generated by (Leibensperger et al., 2011b) with the GISS GCM III (Rind et al., 2007) applied to the IPCC A1B scenario (Nakicenovic and Swart, 2000) and including time-dependent aerosol radiative forcings. For each realization we examined the change in median cyclone frequency between the present-day (1996-2010) and the future (2036-2050), by applying the spectral-autoregressive method to the dominant cyclone PC for each 15-year time series, and using a Monte Carlo method to diagnose the probability distribution and significance of the change based on variability of the AR2 parameters. Three out of the five realizations indicated statistically significant decreases in cyclone frequencies between 1996-2010 and 2036-2050 of -3.2, -3.4 and -1.5 a^{-1} (p -value < 0.05). One realization showed a significant increase of 2.7 a^{-1} and another showed no significant change. Figure 3.10 shows the combined probability distribution of cyclone frequency change in the Midwest from all five realizations and the corresponding responses of annual mean $PM_{2.5}$ based on the $PM_{2.5}$ -to-cyclone period sensitivity reported above, indicating a roughly 70% probability of reduced cyclone frequency and elevated $PM_{2.5}$ in the Midwest by 2050. This corresponds to a mean decrease in cyclone frequency of $-1.1 \pm 4.8 \text{ a}^{-1}$ and a resulting increase in annual mean $PM_{2.5}$ of $0.13 \pm 0.60 \mu\text{g m}^{-3}$.

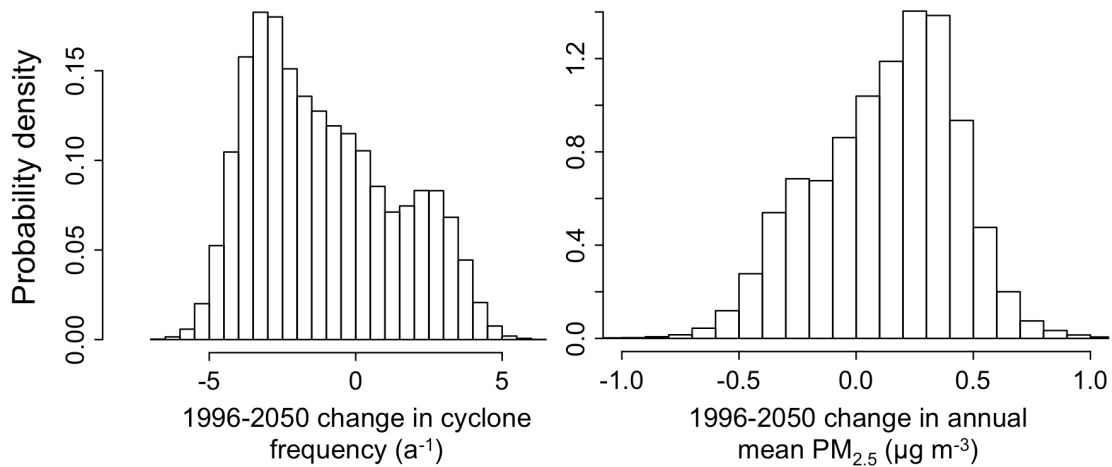


Figure 3.10. Probability distribution for the change in median cyclone frequency in the US Midwest between 1996-2010 and 2036-2050, and the corresponding change in annual mean PM_{2.5} concentrations. Results are from five realizations of the NASA Goddard Institute for Space Studies (GISS) GCM III applied to the IPCC A1B scenario of greenhouse gas and aerosol forcings.

Previous GISS-GEOS-Chem GCM-CTM studies of the effects of 2000-2050 climate change on PM_{2.5} air quality projected a mean increase of 0.1-0.5 μg m⁻³ in the Midwest in the 2050 climate based on one GCM realization (Pye et al., 2009; Lam et al., 2011). Their estimates are within the range of our projection from the cyclone frequency trend alone. However, the large variability of the cyclone trends (including in sign) across five realizations of the same GCM underscores the imperative need for multiple realizations in diagnosing the effect of climate change on PM_{2.5} air quality. All GCM-CTM studies in the literature reviewed by Jacob and Winner (2009) have used single climate realizations and this may partly explain the inconsistency in their results.

Other climatic factors than cyclone and frontal frequency may also affect future PM_{2.5} air quality in the US. Mean temperature increases may be particularly important

for the Southeast as discussed previously. Changes in precipitation and PBL depth are obviously important. As scavenging within a precipitating column is highly efficient (Balkanski et al., 1993), precipitation frequency, often modulated by synoptic weather, may be more relevant as a predictor than climatological mean precipitation.

3.6. Conclusions

Projecting the effects of climate change on $PM_{2.5}$ air quality requires an understanding of the dependence of $PM_{2.5}$ on meteorological variables. We used here a multiple linear regression model to correlate both observed (EPA-AQS) and simulated (GEOS-Chem) daily mean concentrations of total $PM_{2.5}$ and its major components with a suite of meteorological variables in the contiguous US for 2004-2008. All data were deseasonalized to focus on synoptic correlations. We applied principal component analysis (PCA) and regression to identify the dominant meteorological modes controlling $PM_{2.5}$ variability, and showed how trend analysis for these modes can be used to estimate the effects of climate change on $PM_{2.5}$.

We observe strong positive correlations of all $PM_{2.5}$ components with temperature in most of the US, except for nitrate in the Southeast where the correlation is negative. A temperature perturbation simulation with GEOS-Chem reveals that most of the correlations of $PM_{2.5}$ with temperature do not arise from direct dependence on temperature but from covariation with synoptic transport. Exceptions are nitrate and OC in the Southeast, where the direct dependence of ammonium nitrate thermodynamics and biogenic VOC emissions on temperature contributes significantly to the

correlations. RH is generally positively correlated with sulfate and nitrate but negatively correlated with OC; the correlations also appear to be mainly driven by covariation of RH with synoptic transport. Total PM_{2.5} is strongly negatively correlated everywhere with precipitation and wind speed.

We find from the PCA and regression that 20-40% of the observed PM_{2.5} day-to-day variability in different US regions can be explained by a single dominant synoptic meteorological mode: cold frontal passages in the eastern US and maritime inflow in the West. These and other transport modes are found to contribute to most of the overall correlations of different PM_{2.5} components with temperature and RH except in the Southeast.

We show that the interannual variability of annual mean PM_{2.5} in the Midwest for 1999-2010 is strongly correlated with cyclone frequency as diagnosed from a spectral-autoregressive analysis of the dominant meteorological mode of variability, with a PM_{2.5}-to-cyclone period sensitivity of $0.9 \pm 0.4 \mu\text{g m}^{-3} \text{ d}^{-1}$. We conducted an ensemble of five realizations of 1996-2050 climate change using the GISS GCM III with A1B greenhouse and aerosol forcings. Three of these found a significant decrease in cyclone frequency over the US Midwest, one found no significant change and one found a significant increase. From this ensemble we derive a likely increase in annual mean PM_{2.5} of $0.13 \pm 0.60 \mu\text{g m}^{-3}$ in the Midwest in the 2050s climate. This is consistent with previous GCM-CTM studies using the same GCM and suggests that cyclone frequency may be a major driver of the effect of climate change on PM_{2.5} air quality. However, the variability of cyclone trends (including in sign) across multiple

realizations of the same GCM with identical forcings demonstrates the importance of multiple climate change realizations in GCM-CTM studies because of climate chaos. All GCM-CTM studies to date have used single realizations because of computational expense, and this may partly explain the wide inconsistencies in their projections of PM_{2.5} response to climate change. The climate trend analysis in this study, using the Midwest as an illustration, is preliminary. A comprehensive analysis using outputs from various GCMs will be the topic of a future paper.

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Chapter 4. Impact of 2000-2050 climate change on fine particulate matter (PM_{2.5}) air quality inferred from a multi-model analysis of meteorological modes

{ Tai, A.P.K., L.J. Mickley, and D.J. Jacob. 2012. Impact of 2000-2050 climate change on fine particulate matter (PM_{2.5}) air quality inferred from a multi-model analysis of meteorological modes. *Atmos. Chem. Phys. Discuss.*, 12, 18107-18131. }

Abstract

Studies of the effect of climate change on fine particulate matter (PM_{2.5}) air quality using general circulation models (GCMs) have yielded inconsistent results including in the sign of the effect. This reflects uncertainty in the GCM simulations of the regional meteorological variables affecting PM_{2.5}. Here we use the CMIP3 archive of data from fifteen different IPCC AR4 GCMs to obtain improved statistics of 21st-century trends in the meteorological modes driving PM_{2.5} variability over the contiguous US. We analyze 1999-2010 observations to identify the dominant meteorological modes driving interannual PM_{2.5} variability and their synoptic periods T. We find robust correlations ($r > 0.5$) of annual mean PM_{2.5} with T, especially in the eastern US where the dominant modes represent frontal passages. The GCMs all have significant skill in reproducing present-day statistics for T and we show that this reflects their ability to simulate atmospheric baroclinicity. We then use the local PM_{2.5}-to-period sensitivity ($dPM_{2.5}/dT$) from the 1999-2010 observations to project PM_{2.5} changes from the 2000-2050 changes in T simulated by the 15 GCMs following the SRES A1B

greenhouse warming scenario. By weighted-average statistics of GCM results we project a likely 2000-2050 increase of $\sim 0.1 \mu\text{g m}^{-3}$ in annual mean $\text{PM}_{2.5}$ in the eastern US arising from less frequent frontal ventilation, and a likely decrease of $\sim 0.3 \mu\text{g m}^{-3}$ in the northwestern US due to more frequent maritime inflows. These circulation-driven changes are relatively small. Potentially larger regional effects of 2000-2050 climate change on $\text{PM}_{2.5}$ may arise from changes in temperature, biogenic emissions, wildfires, and vegetation, but are still unlikely to affect annual $\text{PM}_{2.5}$ by more than $0.5 \mu\text{g m}^{-3}$.

4.1. Introduction

Air pollution is strongly sensitive to weather conditions and is therefore affected by climate change. A number of studies reviewed by Jacob and Winner (2009) have used chemical transport models (CTMs) driven by general circulation models (GCMs) to diagnose the effects of 21st-century climate change on air quality at northern mid-latitudes. These GCM-CTM studies generally concur that 2000-2050 climate change will degrade ozone air quality in polluted regions by 1-10 ppb, but they do not agree on even the sign of the effect for fine particulate matter ($\text{PM}_{2.5}$). Change in ozone is largely driven by change in temperature, but for $\text{PM}_{2.5}$ the dependence on meteorological variables is far more complex, including different sensitivities for different $\text{PM}_{2.5}$ components (Liao et al., 2006; Dawson et al., 2007; Heald et al., 2008; Kleeman, 2008; Pye et al., 2009; Tai et al., 2010).

Tai et al. (2012) proposed an alternate approach for diagnosing the effect of climate change on $\text{PM}_{2.5}$ through identification of the principal meteorological modes

driving observed PM_{2.5} variability. For example, it is well known that cold fronts associated with mid-latitude cyclones drive pollutant ventilation in the eastern US (Cooper et al., 2001; Li et al., 2005). Tai et al. (2012) found that the frequency of cold fronts was a major predictor of the observed interannual variability of PM_{2.5} in the Midwest. GCMs project a general 21st-century decrease in mid-latitude cyclone frequency as a result of greenhouse warming (Bengtsson et al., 2006; Lambert and Fyfe, 2006; Christensen et al., 2007; Pinto et al., 2007; Ulbrich et al., 2008), from which one could deduce a general degradation of air quality. This cause-to-effect relationship has been found in a few GCM-CTM studies (Mickley et al., 2004; Murazaki and Hess, 2006).

However, there is substantial uncertainty in regional projections of future cyclone frequency (Ulbrich et al., 2009; Lang and Waugh, 2011). Indeed, a general difficulty in projecting the effect of climate change on air quality is the underlying GCM uncertainty in simulating regional climate change. This uncertainty arises both from model noise (climate chaos) and from model error (physics, parameters, numerics). Model noise can be important. Tai et al. (2012) conducted five realizations of 2000-2050 climate change in the GISS GCM 3 (Rind et al., 2007) under the same radiative forcing scenario and found that the frequency of cyclones ventilating the US Midwest decreased in three of the realizations, increased in one, and had no trend in one. All GCM-CTM studies to date examining the effect of climate change on PM_{2.5} have used a single climate change realization from a single GCM (Jacob and Winner, 2009), so it is no surprise that they would yield inconsistent results. This is less of an

issue for GCM-CTM projections of ozone air quality because ozone responds most strongly to changes in temperature (Jacob and Winner, 2009), and all GCMs show consistent warming for the 21st-century climate even on regional scales (Christensen et al., 2007).

The standard approach adopted by the Intergovernmental Panel on Climate Change (IPCC) to reduce uncertainties in GCM projections of regional climate change is to use multiple realizations from an ensemble of GCMs, assuming that model diversity provides some measure of model error (Christensen et al., 2007). Such an ensemble analysis is not practical for GCM-CTM studies of air quality because of the computational expense associated with chemistry and aerosol microphysics. An alternative is to focus on GCM projections of the meteorological modes determining air quality. A resource for this purpose is the World Climate Research Programme's (WCRP's) Coupled Model Intercomparison Project phase 3 (CMIP3) multi-model dataset of 2000-2100 climate change simulations produced by the ensemble of GCMs contributing to the IPCC 4th Assessment Report (AR4).

Here we use this multi-model ensemble to project the responses of PM_{2.5} air quality in different US regions to 2000-2050 climate change. We focus on annual mean PM_{2.5}, which is of primary policy interest (EPA, 2012). We first examine the observed sensitivity of annual mean PM_{2.5} to the frequencies of the dominant meteorological modes in different US regions. We then use the CMIP3 archive of 15 GCMs to project the trends of these frequencies in the future climate, and from there we deduce the corresponding regional trends in PM_{2.5}. These climate-driven PM_{2.5} projections,

independent of trends in anthropogenic emissions, will represent the “climate penalty” or “benefit” for $PM_{2.5}$, which will aid air quality managers to plan emission goals accordingly.

4.2. Observed sensitivity of $PM_{2.5}$ to meteorological modes

Previous studies have demonstrated the importance of synoptic weather in controlling $PM_{2.5}$ variability (Thishan Dharshana et al., 2010; Tai et al., 2012). Tai et al. (2012) identified cyclone passage with associated cold front as the meteorological mode whose period T (length of one cycle, i.e., inverse of frequency) is most strongly correlated with interannual variability of $PM_{2.5}$ in the US Midwest. They proposed that the corresponding $PM_{2.5}$ -to-period sensitivity ($dPM_{2.5}/dT$) could be used to project the response of $PM_{2.5}$ to future climate change; a change ΔT in cyclone period would cause a change $\Delta PM_{2.5} = (dPM_{2.5}/dT)\Delta T$. The physical meaning of this $dPM_{2.5}/dT$ metric is clear when the meteorological mode acts as a pulse, either ventilating a source region (as in the case of a cold front) or polluting a remote region (as in the case of a warm front). We will attempt here to generalize it to the ensemble of conditions over the continental US.

Daily mean $PM_{2.5}$ data for 1999-2010 were obtained from the EPA Air Quality System (AQS) (<http://www.epa.gov/ttn/airs/airsaqs/>) Federal Reference Method (FRM) network of about 1000 sites in the contiguous US. The daily site measurements were interpolated following Tai et al. (2010) onto a $4^\circ \times 5^\circ$ latitude-by-longitude grid, and annual means for each of the 12 years were calculated for each grid cell. Figure 4.1

shows as an example the 1999-2010 time series of annual mean $PM_{2.5}$ for the $4^\circ \times 5^\circ$ grid cell centered over Chicago (asterisk in Fig. 4.2). Linear regression indicates a downward trend of $-0.34 \mu\text{g m}^{-3} \text{ a}^{-1}$, reflecting the improvement of air quality due to emission controls (EPA, 2012). Superimposed on this long-term trend is interannual variability that we assume to be meteorologically driven. The standard deviation of the detrended annual mean $PM_{2.5}$ is $0.79 \mu\text{g m}^{-3}$, or 5.3% of the 12-year mean. For the ensemble of $4^\circ \times 5^\circ$ grid cells in the US we find that the interannual standard deviation of the detrended data ranges from 3 to 19%. Relative interannual variability is largest in the western US but there it could be driven in part by forest fires (Park et al., 2007).

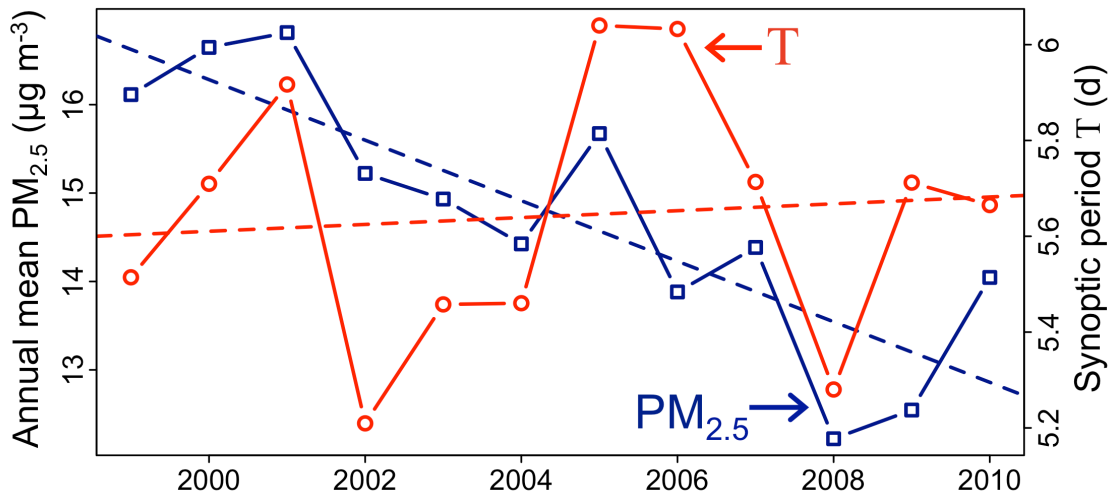


Figure 4.1. Observed 1999-2010 time series of annual mean $PM_{2.5}$ and synoptic period T of the dominant meteorological mode (cold frontal passage) for the $4^\circ \times 5^\circ$ grid square centered over Chicago at $N42^\circ W87.5^\circ$ (asterisk in Fig. 4.2). Linear regression lines are shown as dashed. The detrended variables have a correlation of $r = 0.62$.

Table 4.1. Variables used to define meteorological modes for PM_{2.5} variability. ^a

Variable	Description
x_1	Surface air temperature (K) ^b
x_2	Surface air relative humidity (%) ^b
x_3	Precipitation rate (mm d ⁻¹)
x_4	Sea level pressure (hPa)
x_5	Sea level pressure tendency $dSLP/dt$ (hPa d ⁻¹)
x_6	Surface wind speed (m s ⁻¹) ^{b, c}
x_7	East-west wind direction indicator $\cos\theta$ (dimensionless) ^d
x_8	North-south wind direction indicator $\sin\theta$ (dimensionless) ^d

a. From the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis 1 for 1981-2010. All data are 24-h averages and are deseasonalized as described in the text.

b. “Surface” data are from 0.995 sigma level.

c. Calculated from the horizontal wind vectors (u , v).

d. θ is the angle of the horizontal wind vector counterclockwise from the east. Positive values of x_7 and x_8 indicate westerly and southerly winds, respectively.

We follow the approach of Tai et al. (2012) to determine the dominant meteorological modes for interannual PM_{2.5} variability on the 4°×5° grid. Daily meteorological variables for 1981-2010 (Table 4.1) were obtained from the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis 1

(<http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html>) (Kalnay et al., 1996; Kistler et al., 2001). We regridded the original $2.5^\circ \times 2.5^\circ$ data onto the $4^\circ \times 5^\circ$ grid and deseasonalized them by subtracting the 30-day moving averages.

Following Tai et al. (2012), we decomposed the daily time series of the meteorological variables (Table 4.1) for each $4^\circ \times 5^\circ$ grid cell to produce time series of eight principal components (U_1, \dots, U_8):

$$U_j(t) = \sum_{k=1}^8 \alpha_{kj} \frac{x_k(t) - \bar{x}_k}{s_k} \quad (4.1)$$

where x_k is the deseasonalized meteorological variable, \bar{x}_k and s_k are the temporal mean and standard deviation of x_k , α_{kj} describes the elements of the orthogonal transformation matrix defining the meteorological modes (Tai et al., 2012), and t is time. Each $U_j(t)$ represents the principal component time series for a distinct meteorological mode. We then applied Fourier transform to $U_j(t)$ with a second-order autoregressive (AR2) filter to obtain a smoothed frequency spectrum for each year (Wilks, 2011), and extracted the median AR2 spectral frequency (f) to calculate the corresponding period of the meteorological mode ($T = 1/f$). See Tai et al. (2012) for further description and example application.

From there we applied reduced major axis regression to the 1999-2010 annual time series of detrended $PM_{2.5}$ and T in each $4^\circ \times 5^\circ$ grid cell to determine $dPM_{2.5}/dT$. The dominant meteorological mode for each grid cell was identified as that whose period is most strongly correlated with annual mean $PM_{2.5}$ and explains more than 25% of interannual $PM_{2.5}$ variability (p -value < 0.095). Figure 4.1 shows as an example the

time series of the period of the dominant meteorological mode in the Chicago grid cell (frontal passage). The detrended variables correlate with $r = 0.62$ and $dPM_{2.5}/dT = 2.9 \pm 1.4 \mu\text{g m}^{-3} \text{d}^{-1}$ (95% confidence interval), reflecting the importance of the frequency of frontal ventilation in controlling interannual $PM_{2.5}$ variability in the Midwest.

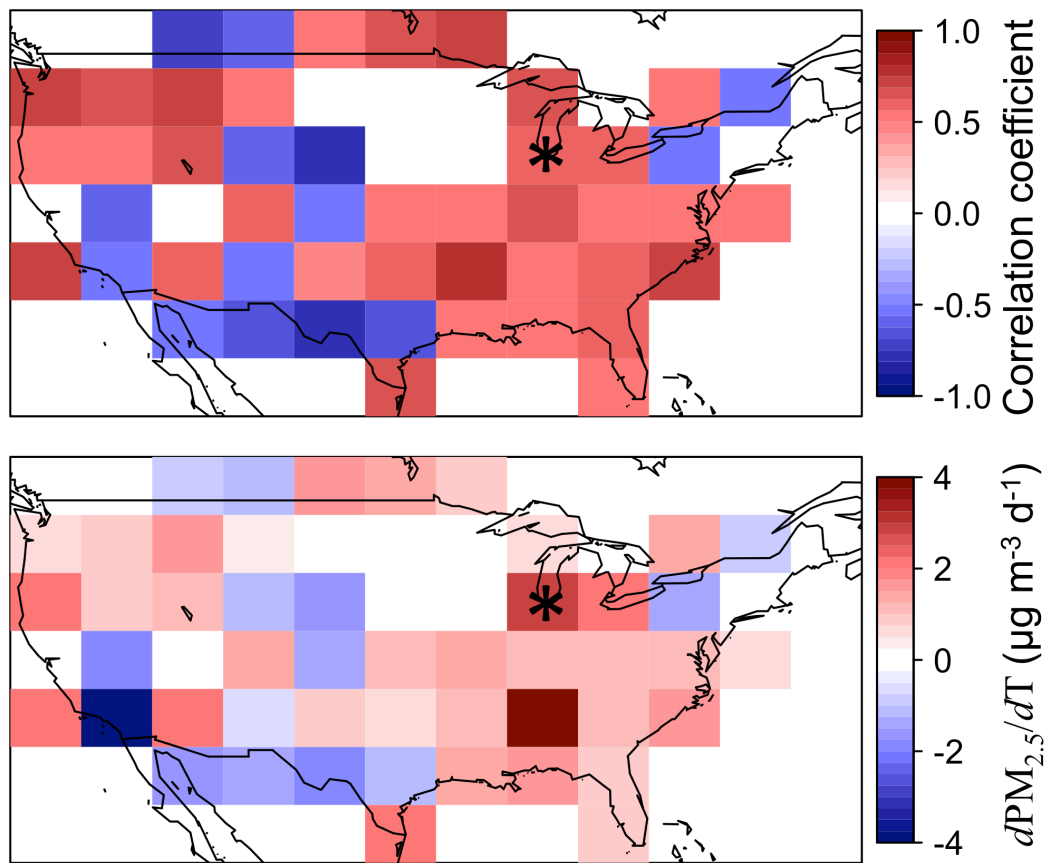


Figure 4.2. Interannual correlation of annual mean $PM_{2.5}$ with the period T of the dominant meteorological mode for 1999-2010 observations: correlation coefficients (top) and reduced-major-axis regression slopes $dPM_{2.5}/dT$ (bottom). Only values significant with 90% confidence (p -value < 0.1) are shown. The asterisk marks the Chicago grid cell for which the time series of $PM_{2.5}$ and T are shown in Fig. 4.1.

Figure 4.2 shows the interannual correlations between $PM_{2.5}$ and T, and the corresponding slopes $dPM_{2.5}/dT$, for the dominant meteorological modes across the US. If two or more modes show similar correlation in a given grid cell, the leading principal component is shown. The mean values of T range from 5 to 9 days (Fig. 4.3), a typical synoptic time scale for frontal passages. There are two outlying grid cells in the interior Northwest where T exceeds 13 days and the physical meaning is not clear. The slopes $dPM_{2.5}/dT$ are usually positive in the eastern US, reflecting the ventilation associated with frontal passage. Negative $dPM_{2.5}/dT$ values in two Northeast grid cells may reflect transport of pollution in southwesterly flow behind warm fronts. Positive $dPM_{2.5}/dT$ in the Northwest can be understood to reflect periodic ventilation by maritime inflow and scavenging by the accompanying precipitation (Tai et al., 2012). In other parts of the western US the physical interpretation of $dPM_{2.5}/dT$ is less clear, and the $PM_{2.5}$ data may not be representative of the $4^\circ \times 5^\circ$ grid cell because of sparsity of observations, urban bias, and complex topography (Malm et al., 2004; Tai et al., 2010). Nevertheless, we often find significant $PM_{2.5}$ -T correlations.

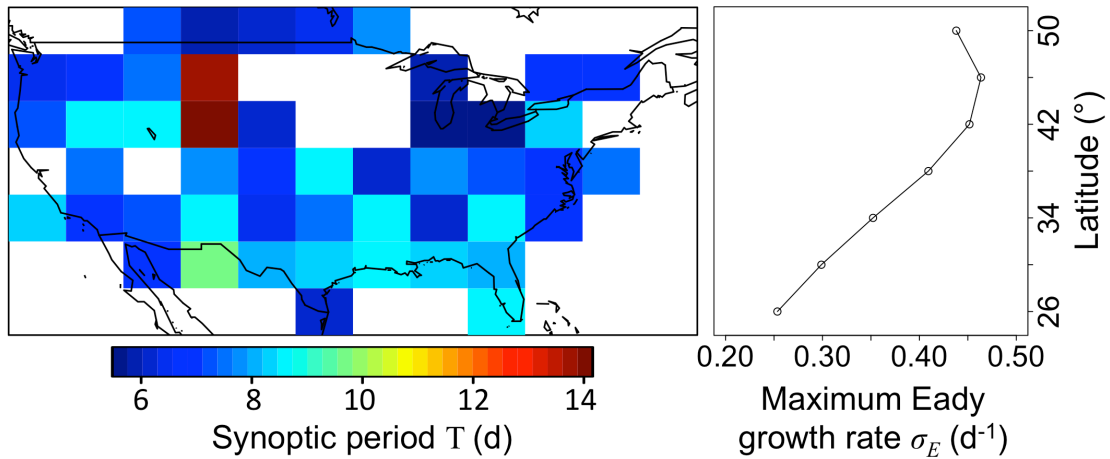


Figure 4.3. Mean synoptic periods T of the dominant meteorological modes for interannual $PM_{2.5}$ variability in NCEP/NCAR Reanalysis 1 observations for 1981-2000. Also shown is the latitudinal profile of maximum Eady growth rate σ_E as calculated by Eq. (4.2) for 0° - 180° W and 850-500 hPa.

4.3. GCM simulations of meteorological modes relevant to $PM_{2.5}$

We examined the ability of the IPCC AR4 GCMs to reproduce the present-day synoptic periods of the dominant meteorological modes for $PM_{2.5}$ interannual variability as prelude to applying these GCMs to diagnose future changes in these periods. We used the 15 IPCC AR4 GCMs from the CMIP3 multi-model dataset (<https://esg.llnl.gov:8443/index.jsp>) that had archived all the daily variables from Table 4.1 needed to project the GCM data onto the meteorological modes defined by the NCEP/NCAR observations. The GCM data have original horizontal resolution ranging from $1^\circ \times 1^\circ$ to $4^\circ \times 5^\circ$ and were all regridded here to $4^\circ \times 5^\circ$. We analyzed the 20th century simulations (20C3M) for 1981-2000, generated the principal component time series $U_j(t)$ for the meteorological modes defined by the NCEP/NCAR observations, and

obtained the median periods of these modes on the $4^\circ \times 5^\circ$ grid to compare to observations.

Figure 4.4 compares the GCM median periods T of the dominant meteorological modes with the NCEP/NCAR observations of Fig. 4.3. The models show strong skill in reproducing the spatial variability of T . We see from Fig. 4.3 that much of this variability is driven by a meridional gradient in synoptic periods, with shorter periods at higher latitudes. This gradient appears in turn to reflect the baroclinicity of the atmosphere. Mid-latitude synoptic weather is mostly driven by baroclinic instability that arises from strong meridional temperature gradients (Holton, 2004) and can be measured by the maximum Eady growth rate (σ_E) (Lindzen and Farrell, 1980):

$$\sigma_E = 0.31 \frac{g}{NT} \left| \frac{\partial T}{\partial y} \right| \quad (4.2)$$

where g is the gravitational acceleration, N is the Brunt-Väisälä frequency, T is the zonal mean temperature, and y is the meridional distance. As shown in Fig. 4.3, σ_E calculated from the NCEP/NCAR data at 850-500 hPa increases sharply between the tropics and 40°N , consistent with the decreasing trend of T . All models can reproduce this observed latitudinal trend in baroclinicity very well, with R^2 values ranging between 0.72-0.95 across the 15 GCMs. We further found that for a given $4^\circ \times 5^\circ$ grid cell, the inter-model variability across the 15 GCMs in the period T of the dominant meteorological mode is correlated with modeled baroclinicity as measured by σ_E . This is illustrated in Fig. 4.5 for the Chicago grid cell. Thus the ability of the GCMs to reproduce T and its variability reflects their ability to reproduce atmospheric baroclinicity.

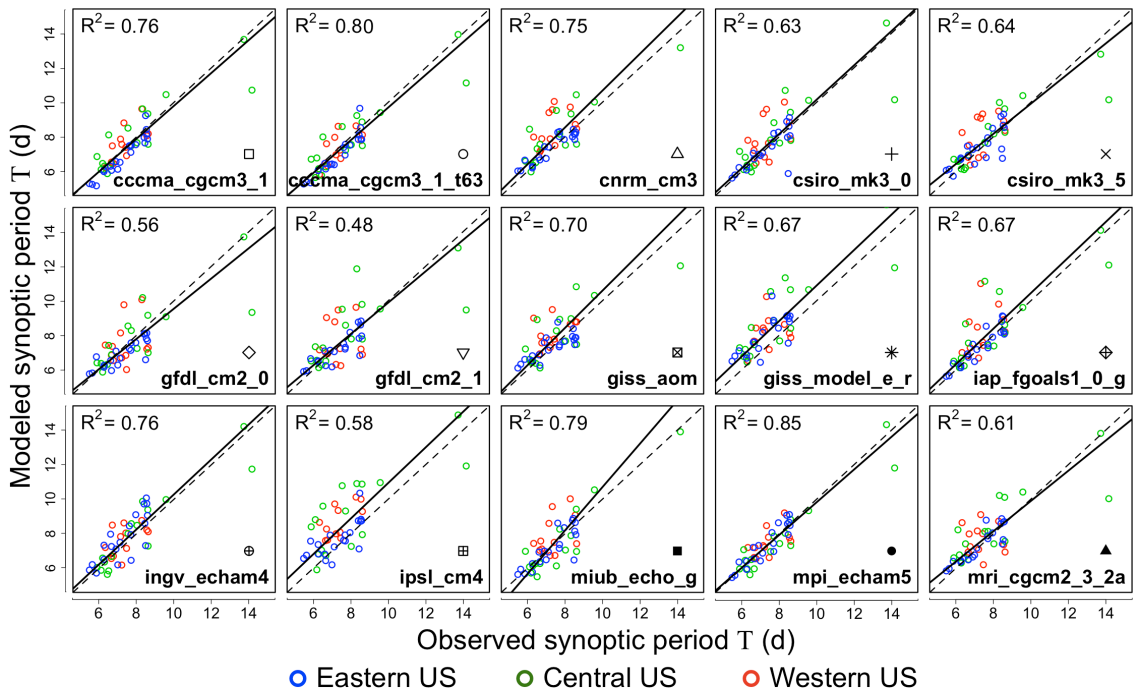


Figure 4.4. Scatterplots of modeled vs. observed synoptic periods T of dominant meteorological modes for interannual $PM_{2.5}$ variability in the US for 1981-2000. Observed values are from NCEP/NCAR Reanalysis 1, and modeled values from 15 IPCC AR4 GCMs. GCM names are given in each panel, and the symbol above each name is used to identify the model in Fig. 4.5 and 4.7. Each data point represents T for one $4^\circ \times 5^\circ$ grid cell, and the ensemble of points represents the continental US separated as eastern (east of $95^\circ W$), central ($110^\circ - 95^\circ W$), and western (west of $110^\circ W$). The solid black line is the reduced major-axis regression slope, with coefficient of variation (R^2) also given. The 1:1 line is shown as dashed.

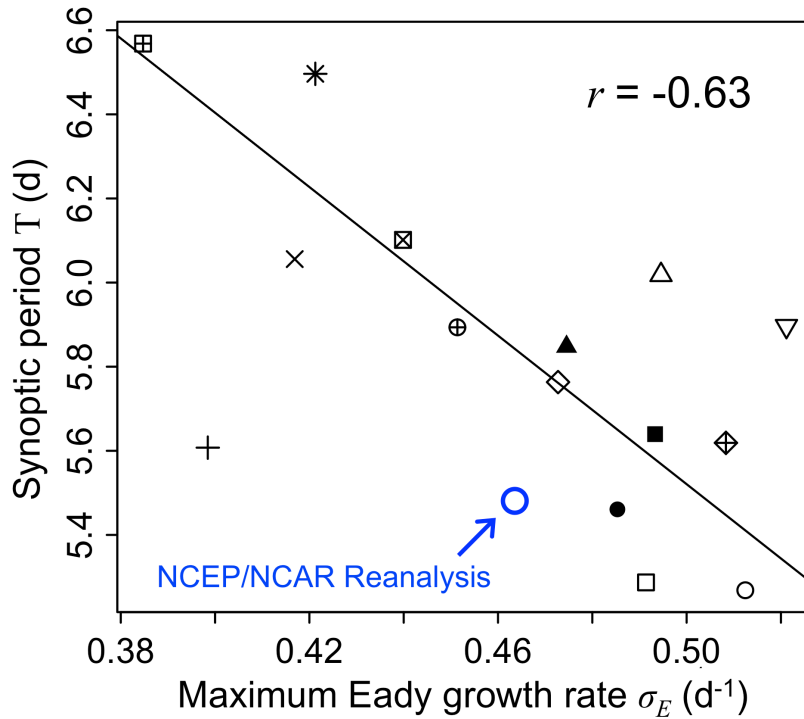


Figure 4.5. Relationship between atmospheric baroclinicity and synoptic period T of the dominant meteorological mode for $PM_{2.5}$ variability in the Chicago grid cell as simulated by 15 IPCC AR4 GCMs for 1981-2000. The observed value from the NCEP/NCAR Reanalysis 1 is also indicated. Baroclinicity is measured as the maximum Eady growth rate σ_E for 44° - 48° N and 850-500 hPa. Each symbol represents an individual GCM (see Fig. 4.4). Correlation coefficient and reduced-major-axis regression slope are also shown.

4.4. Effect of climate change on $PM_{2.5}$

The general skill of the IPCC AR4 GCMs to reproduce present-day synoptic periods relevant to $PM_{2.5}$ variability lends some confidence in their ability to project future changes in these periods. Following the general IPCC strategy, we can expect the ensemble of 15 GCMs to provide a better projection than any single GCM. However, as

Fig. 4.4 shows, some models perform better than others, and we should give less weight to poorly performing models. We use here the approach by Tebaldi et al. (2004, 2005), which combines Bayesian analysis with the reliability ensemble average (REA) method (Giorgi and Mearns, 2002) to discount models with large biases (with respect to observations) and outliers (with respect to future projections). This produces weighted averages and confidence intervals for future projections of synoptic periods.

We used the CMIP3 archive of GCM data for 2046-2065 following the SRES A1B greenhouse warming scenario, which assumes CO₂ to reach 522 ppm by 2050 (Nakicenovic and Swart, 2000). Comparison to the GCM data for 1981-2000 (*Sect. 4.3*) gives a measure of 2000-2050 climate change. The top panel of Fig. 4.6 shows the weighted-average changes in periods (ΔT) of the dominant meteorological modes for interannual PM_{2.5} variability, and the bottom panel shows the corresponding changes in annual PM_{2.5} concentrations ($\Delta \text{PM}_{2.5}$) obtained by $\Delta \text{PM}_{2.5} = (d\text{PM}_{2.5}/dT)\Delta T$ where $d\text{PM}_{2.5}/dT$ is the observed local relationship (Fig. 4.2). If two or more modes are similarly dominant in a given grid cell, we calculate an average effect from these modes. Figure 4.7 shows the aggregated results for nine regions in the US with the distribution across GCMs.

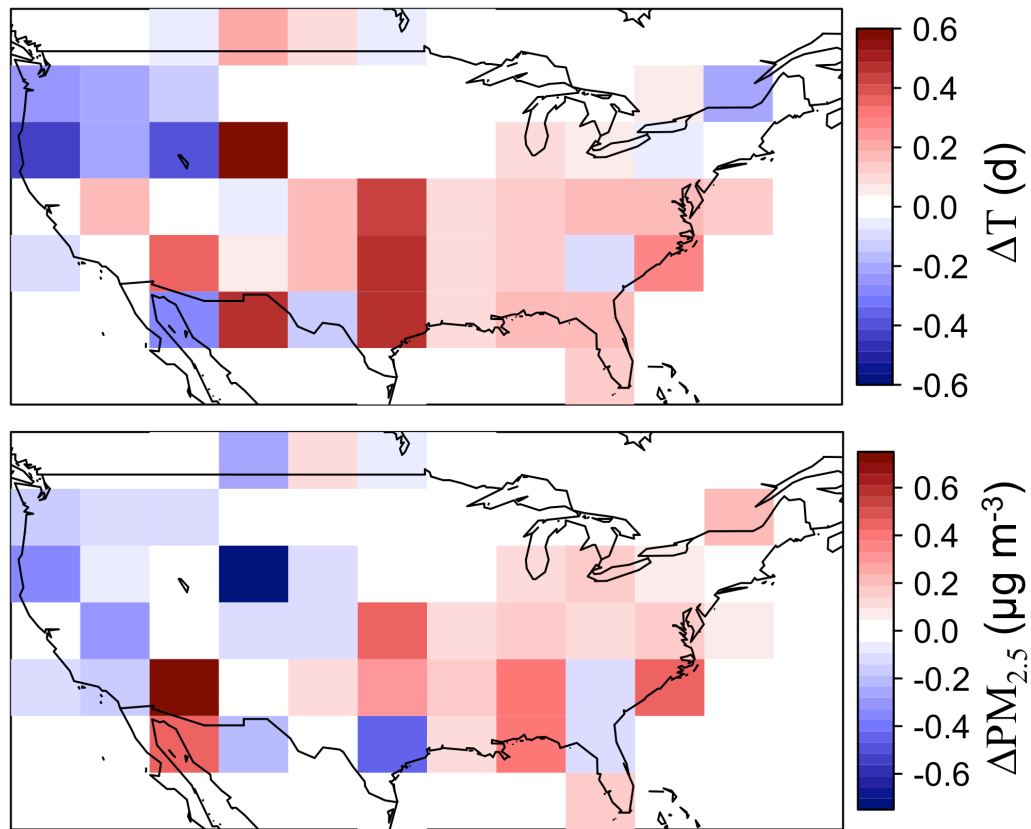


Figure 4.6. Projected 2000-2050 changes in the periods of the dominant meteorological modes for $PM_{2.5}$ variability (top), and implied changes in annual mean $PM_{2.5}$ (bottom). The changes in synoptic periods (ΔT) are weighted averages from the ensemble of IPCC AR4 GCMs calculated using the Bayesian-REA approach of Tebaldi et al. (2004, 2005). The implied changes in $PM_{2.5}$ ($\Delta PM_{2.5}$) are calculated as $\Delta PM_{2.5} = (dPM_{2.5}/dT)\Delta T$ where $dPM_{2.5}/dT$ is the local relationship from Fig. 4.2. When two or more meteorological modes have similar correlation with annual $PM_{2.5}$, an average effect from these modes is calculated.

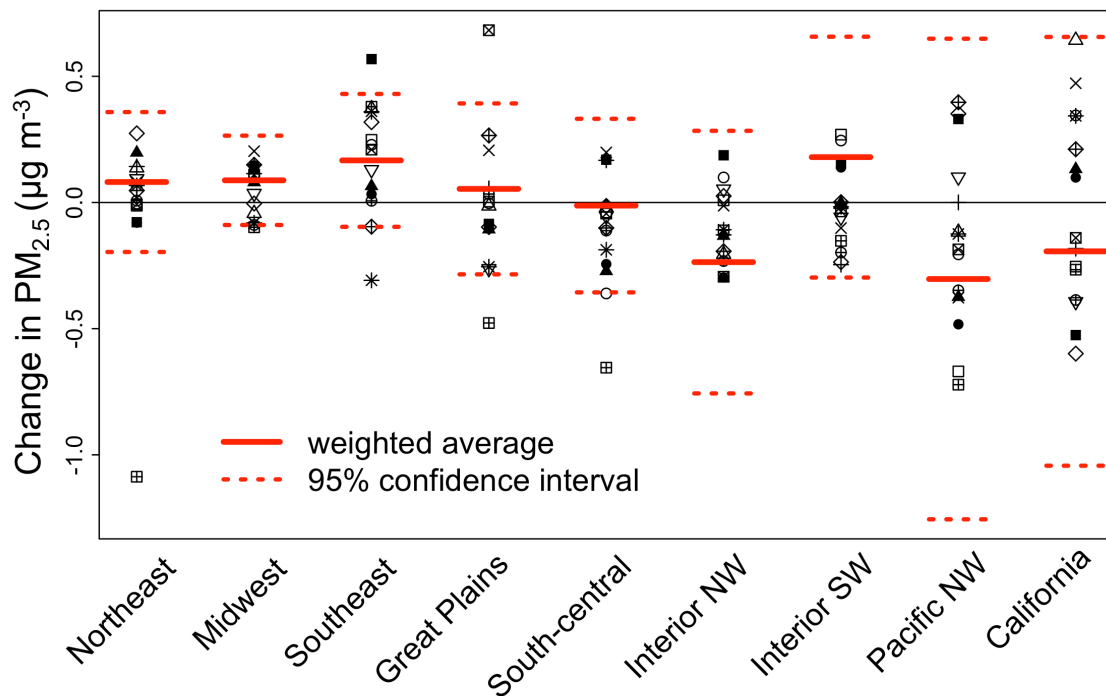


Figure 4.7. 2000-2050 regional changes in annual mean $PM_{2.5}$ concentrations due to changes in the periods of dominant meteorological modes for nine US regions. Regional division follows that of Tai et al. (2012). Symbols represent individual IPCC AR4 GCMs (see Fig. 4.4). Weighted averages and confidence intervals are calculated using the Bayesian-REA approach from Tebaldi et al. (2004, 2005).

We see from Fig. 4.6 and Fig. 4.7 that the future climate features a general increase in $PM_{2.5}$ -relevant synoptic periods in the eastern US, reflecting a more stagnant mid-latitude troposphere with reduced ventilation by frontal passages. This is a robust result which follows from reduced baroclinic instability and poleward shift of storm tracks associated with greenhouse warming (Geng and Sugi, 2003; Mickley et al., 2004; Yin, 2005; Lambert and Fyfe, 2006; Murazaki and Hess, 2006; Pinto et al., 2007; Ulbrich et al., 2008). This in turn leads to a likely (74-91% chance) increase in annual

mean PM_{2.5} with a weighted mean of about 0.1 $\mu\text{g m}^{-3}$ in the eastern US (Northeast, Midwest, and Southeast in Fig. 4.7). In the Northwest (Pacific and Interior NW in Fig. 4.7), we find a likely (71-83% chance) decrease in PM_{2.5} with a weighted mean of about -0.3 $\mu\text{g m}^{-3}$ due to reduced synoptic periods, reflecting more frequent ventilation by maritime inflows and scavenging by the associated precipitation. This is consistent with the general IPCC finding of increasing westerly flow over the western parts of mid-latitude continents in the future climate (Christensen et al., 2007; Meehl et al., 2007). Projections for other parts of the western US are more uncertain. As pointed out earlier, the physical meaning of synoptic periods in the West is less clear than in the East.

GCM-CTM studies in the literature have reported $\pm 0.1-1 \mu\text{g m}^{-3}$ changes in annual mean PM_{2.5} resulting from 2000-2050 climate change, with no consistency across studies (Jacob and Winner, 2009). As pointed out in the *Sect. 4.1*, such inconsistency is to be expected since individual studies used a single future-climate realization from a single GCM. Our multi-model ensemble analysis allows us to conclude with greater confidence that changes in synoptic circulation brought about by climate change will degrade PM_{2.5} air quality in the eastern US but that the effect will be small ($\sim 0.1 \mu\text{g m}^{-3}$). Effects in the western US are potentially larger but of uncertain sign even when the ensemble of IPCC GCMs is considered.

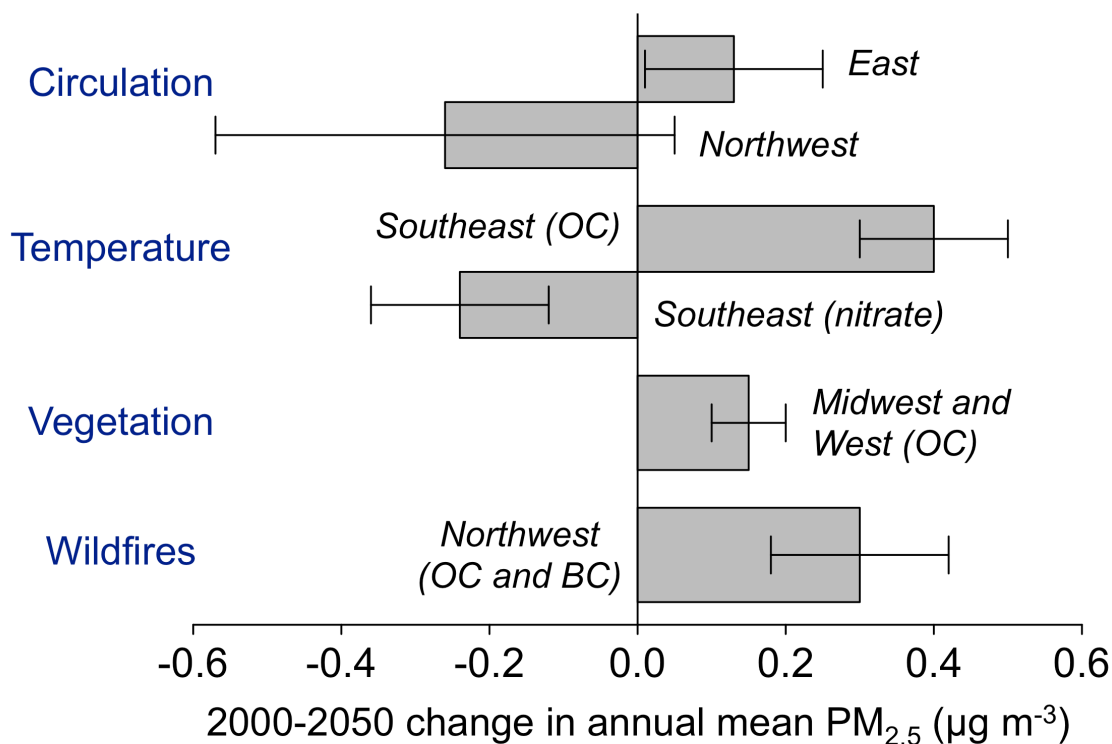


Figure 4.8. Summary of projected effects of 2000-2050 climate change on annual $PM_{2.5}$ in the US as driven by changes in circulation (including precipitation), temperature (biogenic emissions and PM volatility), vegetation dynamics, and wildfires. The affected regions and $PM_{2.5}$ components are identified (OC \equiv organic carbon; BC \equiv black carbon). Error bars represent either the approximate range or standard deviation of the estimate. Estimates are from several studies: this work (circulation); Heald et al. (2008), Pye et al. (2009) and Tai et al. (2012) (temperature); Wu et al. (2012) (vegetation); Spracklen et al. (2009) and Yue et al. (2012) (wildfires). All studies used the IPCC SRES A1B scenario for 2000-2050 climate forcing.

Figure 4.8 summarizes the projected effects of 2000-2050 climate change on annual $PM_{2.5}$ in the US, drawing from this work for circulation changes and from previous studies for other effects. Tai et al. (2012) pointed out that increasing mean

temperature, independently from changes in circulation, could have a large effect on $PM_{2.5}$ in the Southeast and some parts of the western US through biogenic emissions, wildfires, and nitrate aerosol volatility. Temperature-driven changes in the Southeast may reduce ammonium nitrate by $\sim 0.2 \mu\text{g m}^{-3}$ due to increased volatility (Pye et al., 2009; Tai et al., 2012), but increase organic PM by $\sim 0.4 \mu\text{g m}^{-3}$ due to increased biogenic emissions (Heald et al., 2008; Tai et al., 2012). Wu et al. (2012) projected a $0.1\text{-}0.2 \mu\text{g m}^{-3}$ increase in organic PM in the Midwest and western US due to climate-driven changes in vegetation composition. Spracklen et al. (2009) and Yue et al. (2012) projected a $\sim 1 \mu\text{g m}^{-3}$ increase in summertime carbonaceous aerosols in the Northwest due to increased wildfire activities. All in all, none of these effects (or their ensemble) is likely to affect annual mean $PM_{2.5}$ by more than $0.5 \mu\text{g m}^{-3}$. Therefore, for $PM_{2.5}$ regulatory purpose on an annual mean basis, 2000-2050 climate change will unlikely represent any significant penalty or benefit for air quality managers toward the achievement of $PM_{2.5}$ air quality goals.

4.5. Conclusions

$PM_{2.5}$ air quality depends on a number of regional meteorological variables that are difficult to simulate in general circulation models (GCMs). This makes projections of the effect of 21st-century climate change on $PM_{2.5}$ problematic. Consideration of a large ensemble of future-climate simulations using a number of independent GCMs can help to reduce the uncertainty. However, this is not computationally practical in the standard GCM-CTM studies where a chemical transport model (CTM) is coupled to the

GCM for explicit simulation of air quality. We presented here an alternative method by first using climatological observations to identify the dominant meteorological modes driving $PM_{2.5}$ variability, and then using CMIP3 archived data from 15 GCMs to diagnose the effect of 2000-2050 climate change on the periods of these modes.

We focused on projections of annual mean $PM_{2.5}$ over a $4^\circ \times 5^\circ$ grid covering the contiguous US. We showed that the observed 1999-2010 interannual variability of $PM_{2.5}$ across the US is strongly correlated with the periods (T) of the dominant synoptic-scale meteorological modes, particularly in the eastern US where these modes correspond to frontal passages. The observed local relationship $dPM_{2.5}/dT$ then provides a means to infer changes in $PM_{2.5}$ from GCM-simulated changes in T. We find that all GCMs have significant skill in reproducing T and its spatial distribution over the US, reflecting their ability to capture the baroclinicity of the atmosphere. Inter-model differences in synoptic periods can be largely explained by differences in baroclinicity.

We then examined the 2000-2050 trends in synoptic periods T across the continental US as simulated by the ensemble of GCMs for the SRES A1B greenhouse warming scenario. We find a general slowing down of synoptic circulation in the eastern US, as measured by an increase in T. We infer that changes in circulation driven by climate change will likely increase annual mean $PM_{2.5}$ in the eastern US by $\sim 0.1 \mu\text{g m}^{-3}$, reflecting a more stagnant mid-latitude troposphere and less frequent ventilation by frontal passages. We also project a likely decrease by $\sim 0.3 \mu\text{g m}^{-3}$ in the Northwest due to more frequent ventilation by maritime inflows. Potentially larger regional effects of climate change on $PM_{2.5}$ air quality may arise from changes in temperature, biogenic

emissions, wildfires, and vegetation. Overall, however, it is unlikely that 2000-2050 climate change will modify annual mean $\text{PM}_{2.5}$ by more than $0.5 \mu\text{g m}^{-3}$. These climate change effects, independent of changes in anthropogenic emissions, represent a relatively minor penalty or benefit for $\text{PM}_{2.5}$ regulatory purpose. Of more concern would be the effect of increased fires on daily $\text{PM}_{2.5}$.

4.6. Acknowledgements

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