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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

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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 μg 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.

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 (± 1 μg 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 SO_2 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

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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. Data and methods

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 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 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.

Fig. 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.

Table 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 θ (dimensionless)[f]
x_9	North-south wind direction indicator sin θ (dimensionless)[f]

[a] All meteorological parameters are 24-h averages. Except for daily total precipitation and cloud cover, all data are from NCEP/NCAR Reanalysis 1 with spatial resolution of 2.5° × 2.5°.
[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° × 0.25°.
[d] Obtained from NCEP/NCAR Reanalysis 1, regridded from original spatial resolution in T62 Gaussian grid with 192 × 94 points.
[e] Calculated from the horizontal wind vectors (u, v).
[f] θ is the angle of the horizontal surface wind vector counterclockwise from the east.

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). Fig. 2 shows the site locations in 2005 and the regional division used in this work.

Interpolated 2.5° × 2.5° 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:

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

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).

Fig. 3 shows the annual mean concentrations of total $PM_{2.5}$ and the five major $PM_{2.5}$ components, interpolated on the 2.5° × 2.5° 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.

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 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

$$y = \beta_0 + \sum_{k=1}^{9} \beta_k x_k + \text{interaction terms} \quad (2)$$

where y is the deseasonalized and detrended concentration of total $PM_{2.5}$ or its components for each grid square, (x_1, …, x_9) is the ensemble of meteorological variables in Table 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.

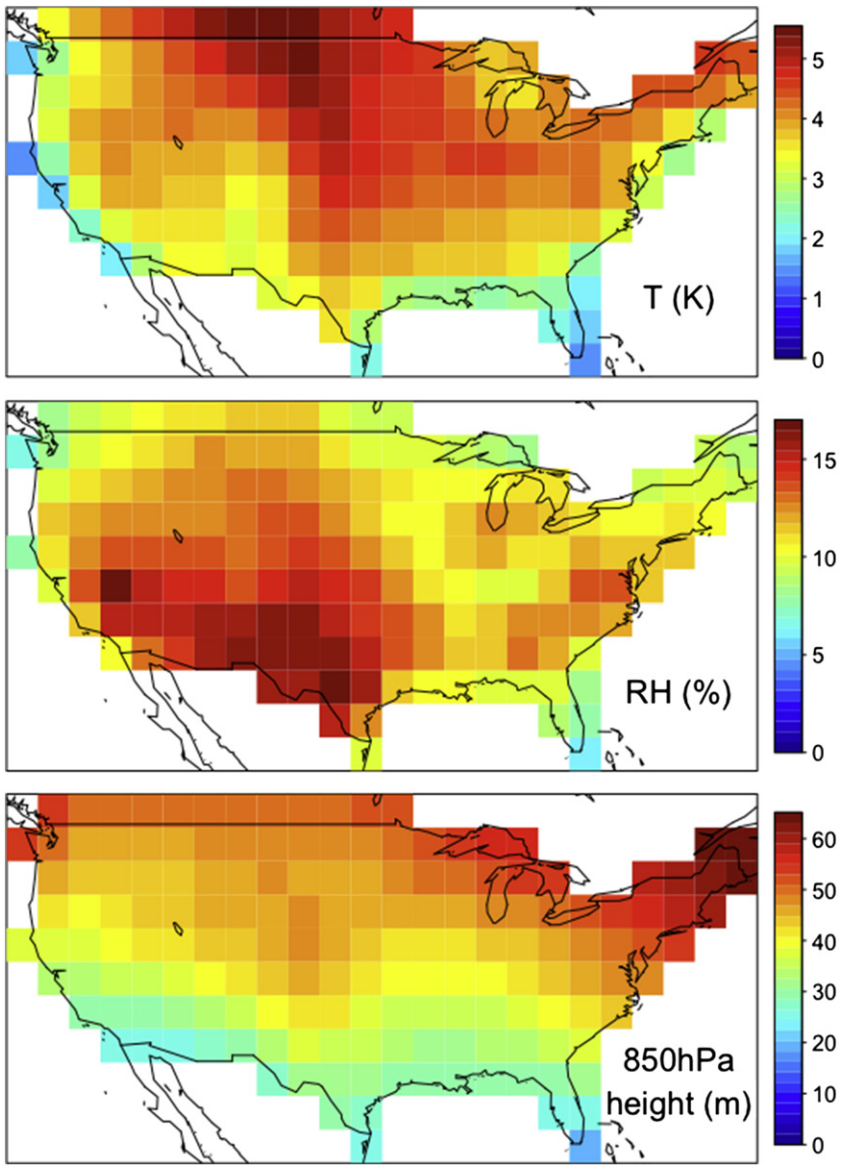

Fig. 1. Daily variability of surface air temperature, relative humidity and 850-hPa geopotential height in the US. Figure shows standard deviations for deseasonalized and detrended observations from 1998 to 2008.

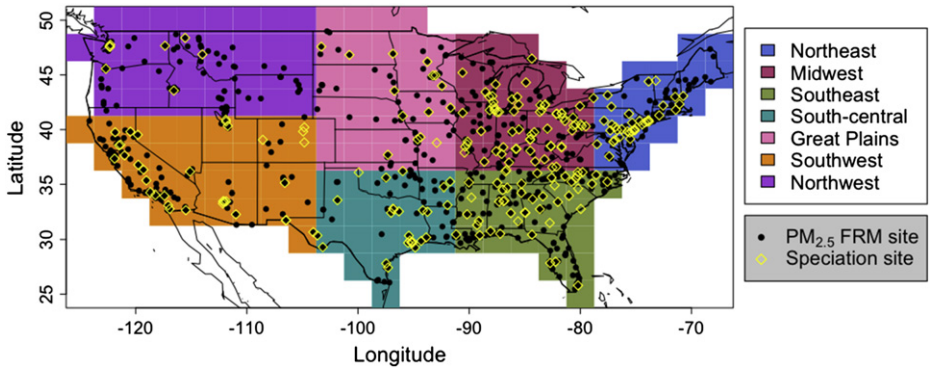

Fig. 2. Locations of EPA Air Quality System $PM_{2.5}$-monitoring sites in 2005. Black dots denote total $PM_{2.5}$ monitors where data are collected with Federal Reference Method (FRM); yellow diamonds denote monitors in chemical speciation network (SLAMS + STN). US regional divisions used in our analysis are also shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

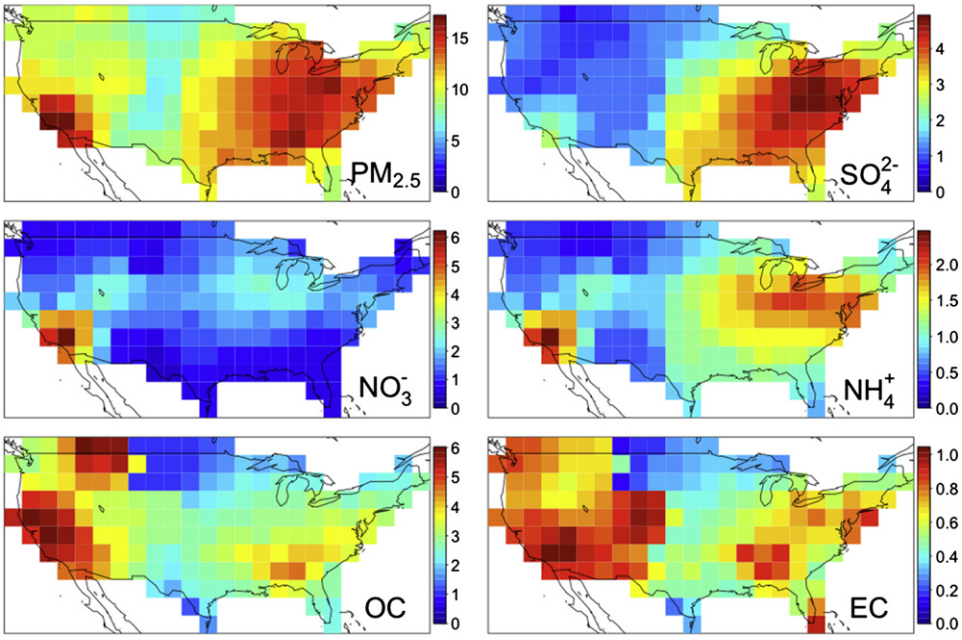

Fig. 3. Annual mean concentrations of total $PM_{2.5}$ and its five major components, interpolated on a 2.5° × 2.5° grid as described in text. Concentrations are in units of μg m^{-3} and averaged over 1998–2008 for total $PM_{2.5}$ and 2000–2008 for individual species. OC concentrations were adjusted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

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 $PM_{2.5}$ that can be accounted for with the MLR model (Kutner et al., 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.

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

3.1. Total $PM_{2.5}$

Fig. 4 shows the relationships of total $PM_{2.5}$ with meteorological variables, as measured by the MLR coefficients β_k in Eq. (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 μg m^{-3} 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 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.

Fig. 4 also shows that high $PM_{2.5}$ concentrations are correlated with high 850-hPa geopotential height (anticyclonic conditions), decreasing sea-level pressure ($dSLP/dt < 0$), 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.

Fig. 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 et al., 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° × 2.5° grid squares (Fig. 2).

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 m s^{-1}, daily mean 500 hPa wind <13 m s^{-1}, and daily total precipitation <0.01 cm d^{-1} (Wang and Angell, 1999). The result is shown in Fig. 6. Total $PM_{2.5}$ is on average 2.6 μg m^{-3} higher on a stagnant day. Fig. 6 also shows the average number of stagnant days per year, highlighting the severity of stagnation in the Southwest.

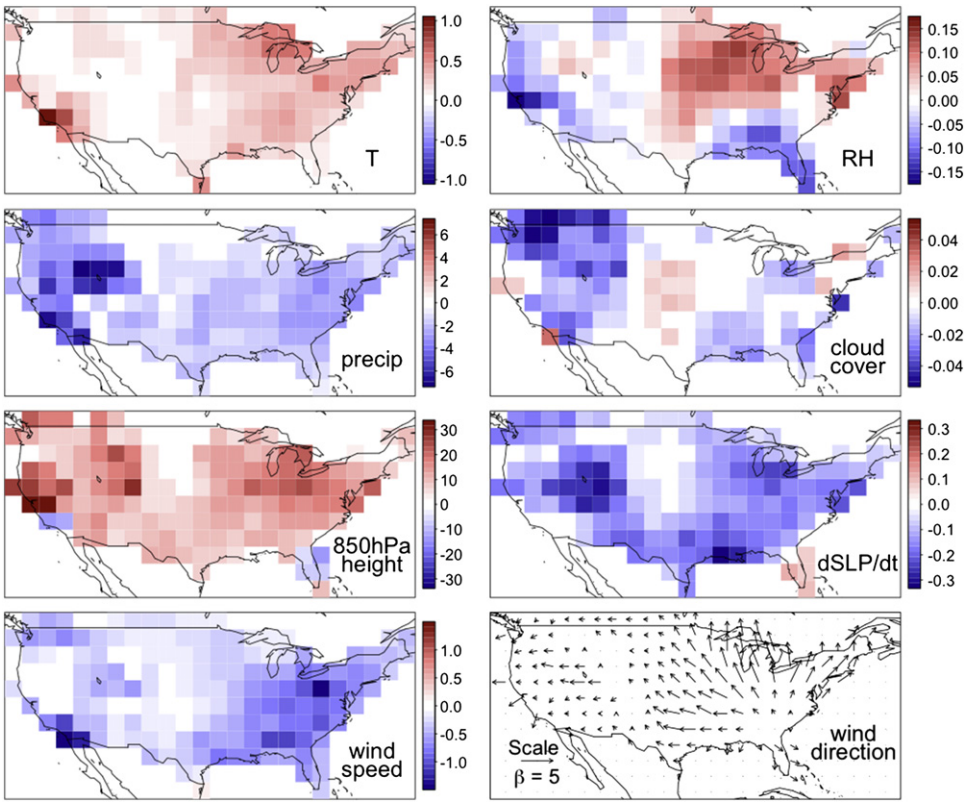

Fig. 4. Correlations of total $PM_{2.5}$ with meteorological variables. Figure shows multiple linear regression coefficients, β_k, in units of μg m^{-3} D^{-1}, where D is dimension of each meteorological variable listed in Table 1. Wind direction panel shows vector sums of regression coefficients β_8 and β_9. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p-value < 0.05).

3.2. $PM_{2.5}$ components vs. temperature

Fig. 7 shows the deseasonalized relationships of the major $PM_{2.5}$ components with surface air temperature, as measured by the MLR coefficient β_1 in Eq. (2). We do not show ammonium as it is mainly the counter-ion for sulfate and nitrate. The relationships in Fig. 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 ng m^{-3} 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 ng m^{-3} 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

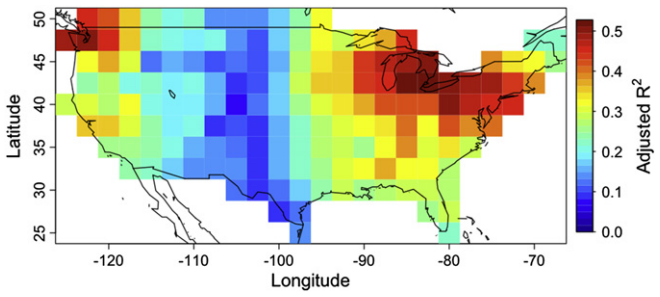

Fig. 5. Coefficients of determination (R^2) for multiple linear regression of deseasonalized and detrended 1998–2008 total $PM_{2.5}$ concentrations on meteorological variables of Table 1. Values are adjusted to account for different number of explanatory terms at each location.

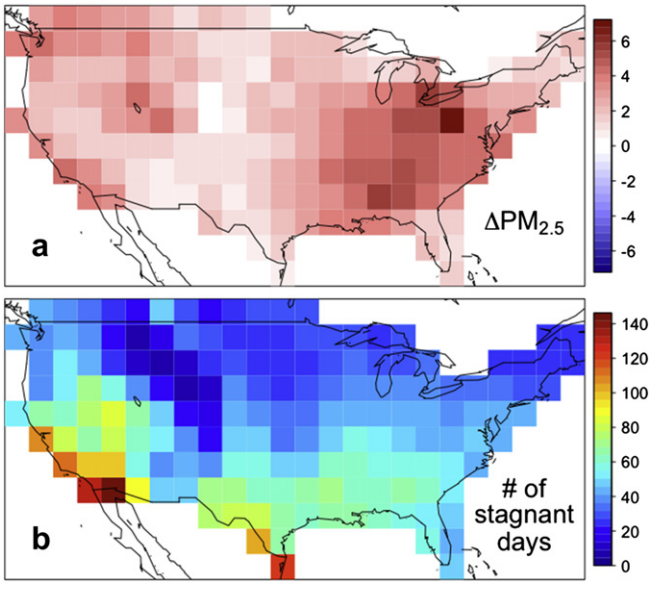

Fig. 6. (a) Average differences in deseasonalized total $PM_{2.5}$ concentrations on stagnant vs. non-stagnant days, based on deseasonalized and detrended 1998–2008 observations. Stagnation is defined following Wang and Angell (1999). Only differences with 95% confidence (p-value < 0.05) are shown. (b) Number of stagnant days per year averaged over 1998–2008.

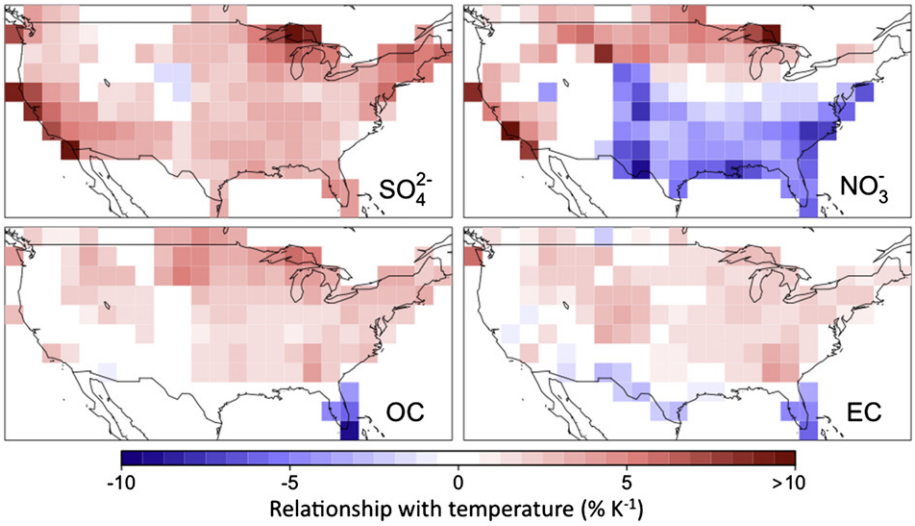

Fig. 7. Correlations of $PM_{2.5}$ components with surface air temperature. Figure shows multiple linear regression coefficients β_1, normalized to annual mean concentrations of Fig. 3. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p-value < 0.05).

temperature is mainly determined by joint association with southerly flow, stagnation, and ventilation of pollution by cold fronts, rather than by chemistry.

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 correlation is much weaker in winter. The Dawson et al. (2007) CTM sensitivity study found an average OC response of -14 ng m^{-3} K^{-1} in summer and -13 ng m^{-3} K^{-1} in winter driven by volatility, but they did not account for variability of biogenic VOC emissions or wildfires.

3.3. $PM_{2.5}$ components vs. relative humidity

Fig. 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). The coefficients for sulfate and nitrate are generally positive. For sulfate this likely reflects the dominant source from in-cloud SO_2 oxidation and the association with moist southerly flow shown by the wind patterns in Fig. 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.

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. 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).

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

Fig. 9 shows the normalized vector sums of MLR coefficients β_8 and β_9 in Eq. (2), which indicate the wind direction most strongly associated with high concentrations of PM components. This dramatically illustrates the role of SO_2 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 NH_3 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.

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

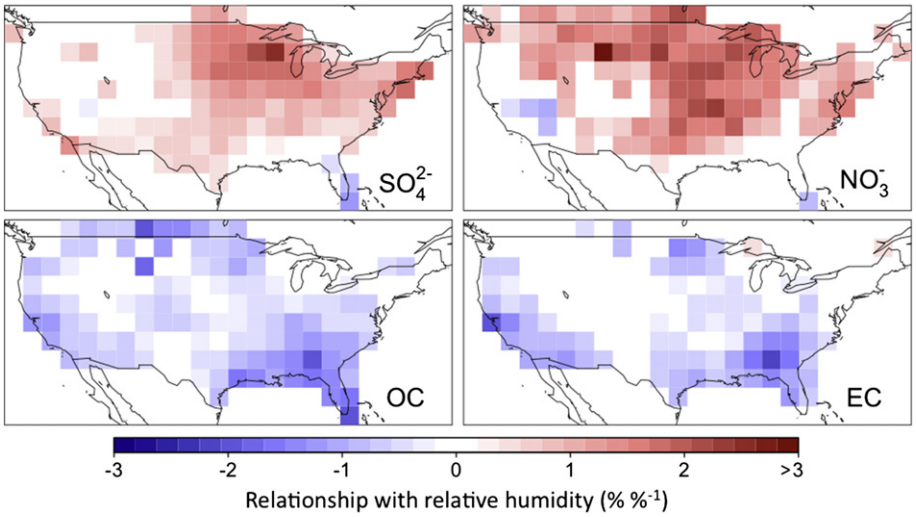

Fig. 8. Same as Fig. 7 but for correlations of $PM_{2.5}$ components with surface air relative humidity.

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. 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 anti-correlation 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 anti-correlation 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. 6, this translates to an average increase of 0.24 μg m^{-3} in summer mean $PM_{2.5}$ concentrations with a maximum increase of 0.93 μg m^{-3} in the Midwest.

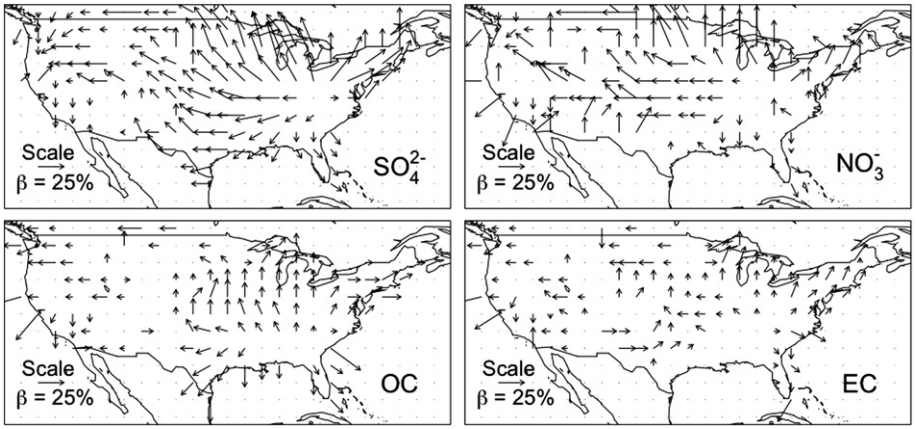

Fig. 9. Correlations of $PM_{2.5}$ components with wind direction. Figure shows vector sums of multiple linear regression coefficients β_8 and β_9, normalized to annual mean concentrations of Fig. 3. Length of arrows (in units of % per unit sine or cosine) indicates magnitude of correlation. Values are for deseasonalized and detrended variables and are only shown when significant with 95% confidence (p-value < 0.05).

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 $\mu g\ m^{-3}$ 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 NH_3 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 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. 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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