



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

A. P. K. Tai, L. J. Mickley, and D. J. Jacob

School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts, USA

Correspondence to: A. P. K. Tai (pkamostai@gmail.com)

Received: 6 June 2012 – Published in Atmos. Chem. Phys. Discuss.: 20 July 2012

Revised: 5 November 2012 – Accepted: 24 November 2012 – Published: 3 December 2012

Abstract. Studies of the effect of climate change on fine particulate matter (PM_{2.5}) air quality using general circulation models (GCMs) show 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 PM_{2.5} in the eastern US arising from less frequent frontal ventilation, and a likely decrease albeit with greater inter-GCM variability in the Pacific Northwest due to more frequent maritime inflows. Potentially larger regional effects of 2000–2050 climate change on PM_{2.5} may arise from changes in temperature, biogenic emissions, wildfires, and vegetation, but are still unlikely to affect annual PM_{2.5} by more than $0.5 \mu\text{g m}^{-3}$.

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 (PM_{2.5}). Change in ozone is largely driven by change in temperature, but for PM_{2.5} the dependence on meteorological variables is far more complex, including different sensitivities for different 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 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 effect of climate change on these frequencies, and from there we deduce the corresponding effect of climate change on PM_{2.5}.

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 $\Delta PM_{2.5} = (dPM_{2.5}/dT)\Delta T$. This assumes that the local $dPM_{2.5}/dT$ relationships will remain unchanged, and that the same meteorological modes will remain dominant for PM_{2.5} variability. The physical meaning of $dPM_{2.5}/dT$ 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).

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 yr were calculated for each grid cell. Such spatial averaging can smooth out local effects and yields more robust correlation statistics of PM_{2.5} with synoptic weather (Tai et al., 2012). Figure 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. 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-yr mean PM_{2.5}. 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% of 12-yr mean PM_{2.5}. Relative interannual variability is largest in the western US but there it could be driven in part by forest fires (Park et al., 2007).

We follow the approach of Tai et al. (2012) to determine the dominant meteorological modes for interannual PM_{2.5} variability on the $4^\circ \times 5^\circ$ grid. Daily meteorological variables for 1981–2010 (Table 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.

Table 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.

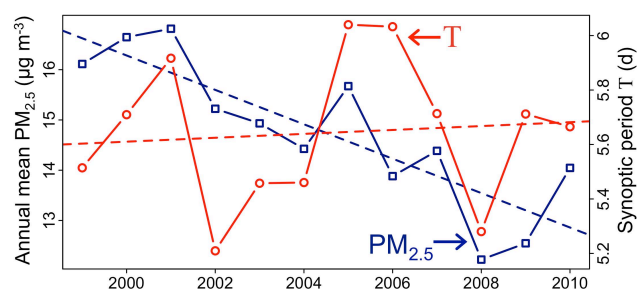


Fig. 1. Observed 1999–2010 time series of annual mean PM_{2.5} and synoptic period T of the dominant meteorological mode (cold frontal passage) for the $4^\circ \times 5^\circ$ grid square centered over Chicago at 42° N, 87.5° W (asterisk in Fig. 2). Linear regression lines are shown as dashed. The detrended variables have a correlation of $r = 0.62$.

Following Tai et al. (2012), we decomposed the daily time series of the meteorological variables (Table 1) for each $4^\circ \times 5^\circ$ grid cell to produce time series of eight principal components (U_1, \dots, U_8):

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

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, in-

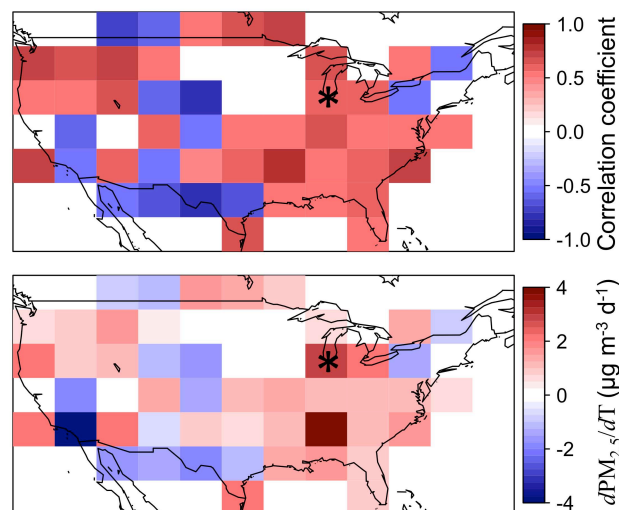


Fig. 2. Interannual correlation of annual mean PM_{2.5} with the period T of the dominant meteorological mode for 1999–2010 observations: correlation coefficients (top) and reduced-major-axis regression slopes $dPM_{2.5}/dT$ (bottom). Only values significant with 90 % confidence (p -value < 0.1) are shown. The asterisk marks the Chicago grid cell for which the time series of PM_{2.5} and T are shown in Fig. 1.

cluding justification for extracting median frequency instead of mean.

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 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 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. The mean values of T range from 5 to 9 days (Fig. 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

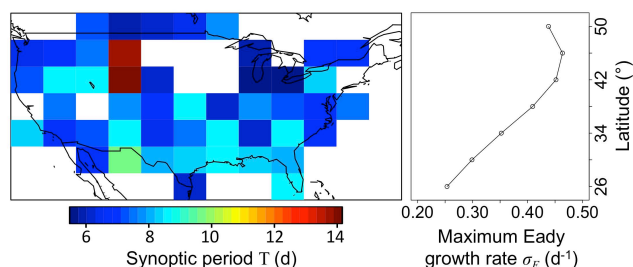


Fig. 3. Mean synoptic periods T of the dominant meteorological modes for interannual PM_{2.5} variability in NCEP/NCAR Reanalysis 1 observations for 1981–2000. Also shown is the latitudinal profile of maximum Eady growth rate σ_E as calculated by Eq. (2) for 0–180° W and 850–500 hPa.

accompanying precipitation (Tai et al., 2012). In other parts of the western US with weak frontal activity, the physical interpretation of $dPM_{2.5}/dT$ is less clear, and the PM_{2.5} data may not be representative of the $4^\circ \times 5^\circ$ scale 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 in these regions.

3 GCM simulations of meteorological modes relevant to PM_{2.5}

We first 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. 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 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 $4^\circ \times 5^\circ$, recognizing that such regridding might have some effect on the GCM meteorological modes and their variability. 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 compares the GCM median periods T of the dominant meteorological modes with the NCEP/NCAR observations of Fig. 3. The models show strong skill in reproducing the spatial variability of T , especially in the eastern US. We see from Fig. 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):

$$\sigma_E = 0.31 \frac{g}{NT} \left| \frac{\partial T}{\partial y} \right| \quad (2)$$

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. 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 (see the Supplement). 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. 5 for the Chicago grid cell (see the Supplement for the correlation for other grid cells). Thus the ability of the GCMs to reproduce T and its variability reflects their ability to reproduce atmospheric baroclinicity.

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 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. 3) gives a measure of 2000–2050 climate change. The top panel of Fig. 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 ($\Delta PM_{2.5}$) obtained by $\Delta PM_{2.5} = (dPM_{2.5}/dT)\Delta T$ where $dPM_{2.5}/dT$ is the observed local relationship (Fig. 2). If two or more modes are similarly dominant in a given grid cell, we calculate an average effect from these modes. Figure 7 shows the aggregated results for nine regions in the US including the spread across GCMs.

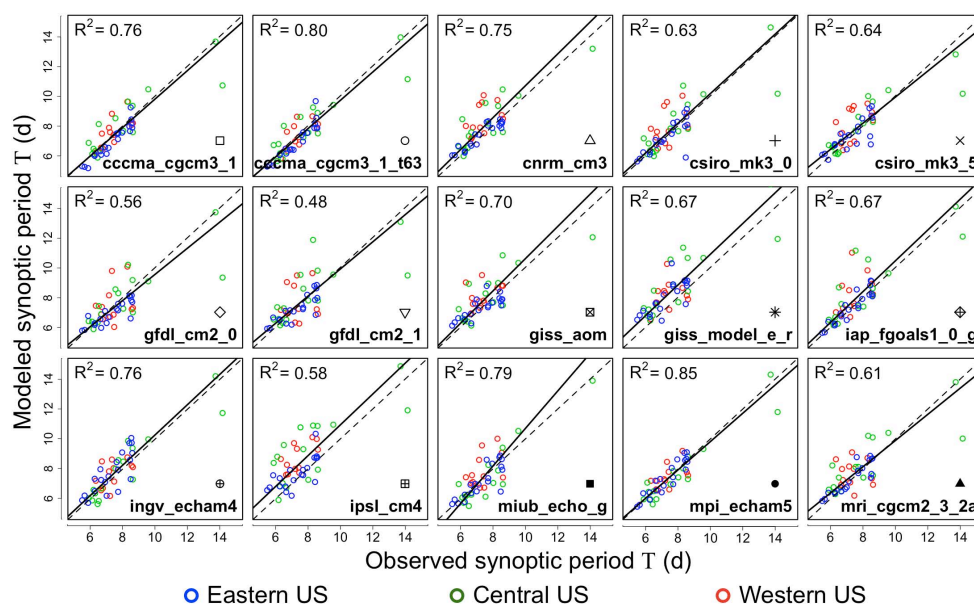


Fig. 4. Scatterplots of modeled vs. observed synoptic periods T of the dominant meteorological modes for interannual PM_{2.5} variability in the US for 1981–2000. Observed values are from NCEP/NCAR Reanalysis 1, and modeled values are from 15 IPCC AR4 GCMs. GCM names are given in each panel, and the symbol above each name is used to identify the model in Figs. 5 and 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° W), central (110 – 95° W), and western (west of 110° 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.

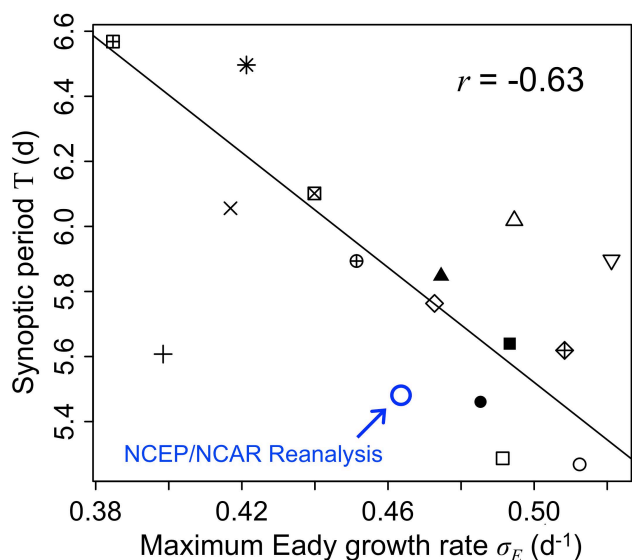


Fig. 5. Relationship between atmospheric baroclinicity and synoptic period T of the dominant meteorological mode for PM_{2.5} variability in the Chicago grid cell as simulated by 15 IPCC AR4 GCMs for 1981–2000. The observed value from the NCEP/NCAR Reanalysis 1 is also indicated. Baroclinicity is measured 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). Correlation coefficient and reduced-major-axis regression slope are also shown.

We see from Fig. 6 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 increase of about $0.1 \mu\text{g m}^{-3}$ (North-east, Midwest, and Southeast in Fig. 7). In the Northwest (Pacific and Interior NW in Fig. 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, albeit with greater inter-model variability than in the eastern US. This reflects more frequent ventilation by maritime inflows and scavenging by the associated precipitation, and 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, and the skill of GCMs to reproduce present-day synoptic periods is generally lower (Fig. 4).

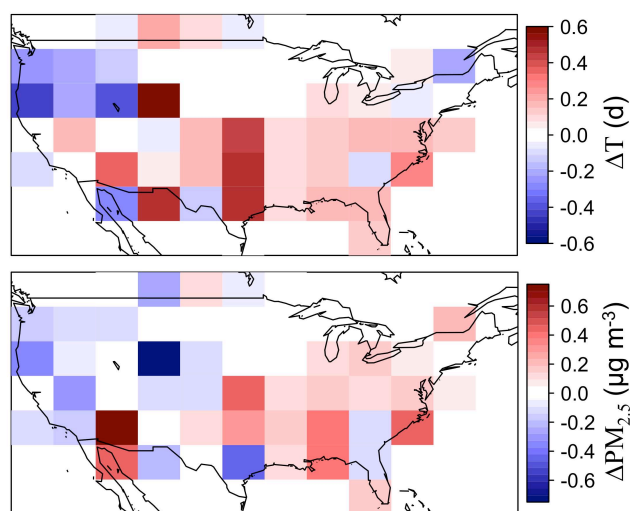


Fig. 6. Projected 2000–2050 changes in the periods of the dominant meteorological modes for PM_{2.5} variability (top), and implied changes in annual mean PM_{2.5} (bottom). The changes in synoptic periods (ΔT) are weighted averages from the ensemble of IPCC AR4 GCMs calculated using the Bayesian-REA 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. 2. When two or more meteorological modes have similar correlation with annual PM_{2.5}, an average effect from these modes is calculated.

GCM-CTM studies in the literature have reported ± 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 Introduction, 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 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 (including modulation of precipitation frequency) and from previous studies for other climatic factors. 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 changes in 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 (Tagaris et al., 2007; Pye et al., 2009), but increase organic PM by $\sim 0.4 \mu\text{g m}^{-3}$ due to increased biogenic emissions (Heald et al., 2008).

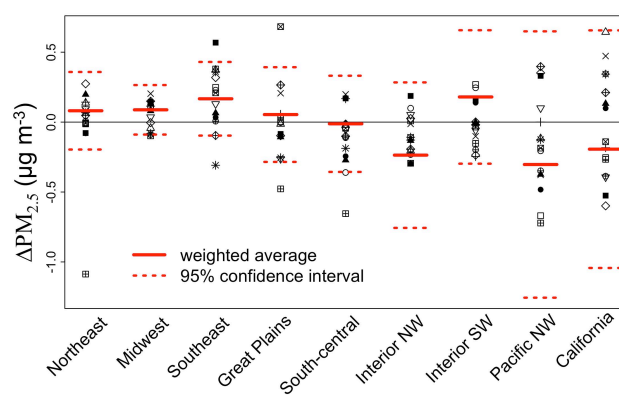


Fig. 7. 2000–2050 regional changes in annual mean PM_{2.5} ($\Delta PM_{2.5}$) due to changes in the periods of dominant meteorological modes for nine US regions. Regional division follows that of Tai et al. (2012). Symbols represent individual IPCC AR4 GCMs (see Fig. 4). Weighted averages and confidence intervals are calculated using the Bayesian-REA approach from Tebaldi et al. (2004, 2005).

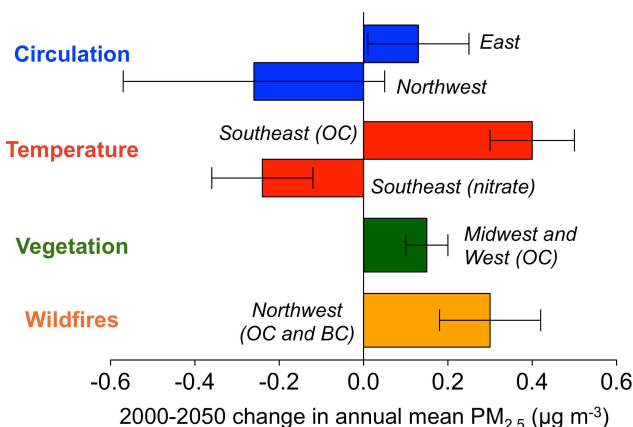


Fig. 8. Summary of projected effects of 2000–2050 climate change on annual PM_{2.5} in the US as driven by changes in circulation, temperature (biogenic emissions and PM volatility), vegetation dynamics, and wildfires. The affected regions and PM_{2.5} components are identified (OC \equiv organic carbon; BC \equiv black carbon). Error bars represent either the approximate range or standard deviation of the estimate. Estimates are from this work (circulation); Tagaris et al. (2007), Heald et al. (2008) and Pye et al. (2009) (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.

Wu et al. (2012) projected a 0.1 – $0.2 \mu\text{g m}^{-3}$ increase in organic PM in the Midwest and western US due to climate-driven changes in ecosystem type. 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. Tagaris et al. (2007) and Avise et al. (2009) predicted an average decrease of summertime PM_{2.5} by $\sim 10\%$ and $\sim 1 \mu\text{g m}^{-3}$ by 2050, respectively,

caused primarily by higher precipitation in their GCMs, but trends in precipitation in most of the US are highly uncertain (Christensen et al., 2007). All in all, none of these effects (or their ensemble) is likely to affect annual mean PM_{2.5} by more than 0.5 $\mu\text{g m}^{-3}$ ($\sim 3\%$ of the current annual standard of 15 $\mu\text{g m}^{-3}$). Therefore, for PM_{2.5} regulatory purpose on an annual mean basis, 2000–2050 climate change will represent only a modest penalty or benefit for air quality managers toward the achievement of PM_{2.5} air quality goals. Of potentially greater concern would be the effect of increased wildfires on daily PM_{2.5} in the western US (Spracklen et al., 2009).

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 analyzing 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 $d\text{PM}_{2.5}/dT$ 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 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 PM_{2.5} regulatory purposes. Of more potential concern would be the effect of increased wildfires on daily PM_{2.5}.

An important caveat in our approach is the assumption that the $d\text{PM}_{2.5}/dT$ will remain unchanged and that the same meteorological modes will remain dominant for PM_{2.5} variability in the future climate. Very large changes in emissions could affect the validity of these assumptions. This could be explored in future work using GCM-CTM studies with perturbed emissions.

Supplementary material related to this article is available online at: <http://www.atmos-chem-phys.net/12/11329/2012/acp-12-11329-2012-supplement.pdf>.

Acknowledgements. This work was supported by a Mustard Seed Foundation Harvey Fellowship to Amos P. K. Tai, and the NASA Applied Sciences Program through the NASA Air Quality Applied Sciences Team (AQAAT). We acknowledge the modeling groups, the Program for Climate Model Diagnosis and Intercomparison (PCMDI) and the WCRP's Working Group on Coupled Modelling (WGCM) for their roles in making available the WCRP CMIP3 multi-model dataset. Support of this dataset is provided by the Office of Science, US Department of Energy. We also thank Susan C. Anenberg and Carey Jang from US EPA for their thoughtful inputs and suggestions.

Edited by: C. H. Song

References

- Avise, J., Chen, J., Lamb, B., Wiedinmyer, C., Guenther, A., Salathé, E., and Mass, C.: Attribution of projected changes in summertime US ozone and PM_{2.5} concentrations to global changes, *Atmos. Chem. Phys.*, 9, 1111–1124, doi:10.5194/acp-9-1111-2009, 2009.
- Bengtsson, L., Hodges, K. I., and Roeckner, E.: Storm tracks and climate change, *J. Climate*, 19, 3518–3543, 2006.
- Christensen, J. H., Hewitson, B., Busuioc, A., Chen, A., Gao, X., Held, I., Jones, R., Kolli, R. K., Kwon, W.-T., Laprise, R., Magaña Rueda, V., Mearns, L., Menendez, C. G., Raisanen, J., Rinke, A., Sarr, A., and Whetton, P.: Regional climate projections, in: *Climate change 2007: The physical science basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, New York, NY, USA, 847–940, 2007.
- Cooper, O. R., Moody, J. L., Parrish, D. D., Trainer, M., Ryerson, T. B., Holloway, J. S., Hubler, G., Fehsenfeld, F. C., Oltmans, S. J.,

- and Evans, M. J.: Trace gas signatures of the airstreams within North Atlantic Cyclones: case studies from the North Atlantic regional experiment (NARE '97) aircraft intensive, *J. Geophys. Res.-Atmos.*, 106, 5437–5456, 2001.
- Dawson, J. P., Adams, P. J., and Pandis, S. N.: Sensitivity of PM_{2.5} to climate in the Eastern US: a modeling case study, *Atmos. Chem. Phys.*, 7, 4295–4309, doi:10.5194/acp-7-4295-2007, 2007.
- EPA: Our nation's air: Status and trends through 2010, U.S. EPA Office of Air Quality Planning and Standards, Research Triangle Park, NC, 2012.
- Geng, Q. Z. and Sugi, M.: Possible change of extratropical cyclone activity due to enhanced greenhouse gases and sulfate aerosols – study with a high-resolution AGCM, *J. Climate*, 16, 2262–2274, 2003.
- Giorgi, F. and Mearns, L. O.: Calculation of average, uncertainty range, and reliability of regional climate changes from AOGCM simulations via the “reliability ensemble averaging” (REA) method, *J. Climate*, 15, 1141–1158, 2002.
- Heald, C. L., Henze, D. K., Horowitz, L. W., Feddema, J., Lamarque, J. F., Guenther, A., Hess, P. G., Vitt, F., Seinfeld, J. H., Goldstein, A. H., and Fung, I.: Predicted change in global secondary organic aerosol concentrations in response to future climate, emissions, and land use change, *J. Geophys. Res.-Atmos.*, 113, D05211, doi:10.1029/2007jd009092, 2008.
- Holton, J. R.: An introduction to dynamic meteorology, 4th ed., International geophysics series, 88, Elsevier Academic Press, Burlington, MA, xii, 535 pp., 2004.
- Jacob, D. J. and Winner, D. A.: Effect of climate change on air quality, *Atmos. Environ.*, 43, 51–63, 2009.
- Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Leetmaa, A., Reynolds, R., Jenne, R., and Joseph, D.: The NCEP/NCAR 40-year reanalysis project, *B. Am. Meteorol. Soc.*, 77, 437–471, 1996.
- Kistler, R., Kalnay, E., Collins, W., Saha, S., White, G., Woollen, J., Chelliah, M., Ebisuzaki, W., Kanamitsu, M., Kousky, V., van den Dool, H., Jenne, R., and Fiorino, M.: The NCEP-NCAR 50-year reanalysis: Monthly means CD-ROM and documentation, *B. Am. Meteorol. Soc.*, 82, 247–267, 2001.
- Kleeman, M. J.: A preliminary assessment of the sensitivity of air quality in California to global change, *Climatic Change*, 87, S273–S292, doi:10.1007/S10584-007-9351-3, 2008.
- Lambert, S. J. and Fyfe, J. C.: Changes in winter cyclone frequencies and strengths simulated in enhanced greenhouse warming experiments: Results from the models participating in the IPCC diagnostic exercise, *Clim. Dynam.*, 26, 713–728, 2006.
- Lang, C. and Waugh, D. W.: Impact of climate change on the frequency of Northern Hemisphere summer cyclones, *J. Geophys. Res.-Atmos.*, 116, D04103, doi:10.1029/2010JD014300, 2011.
- Li, Q. B., Jacob, D. J., Park, R., Wang, Y. X., Heald, C. L., Hudman, R., Yantosca, R. M., Martin, R. V., and Evans, M.: North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone, *J. Geophys. Res.-Atmos.*, 110, D10301, doi:10.1029/2004jd005039, 2005.
- Liao, H., Chen, W. T., and Seinfeld, J. H.: Role of climate change in global predictions of future tropospheric ozone and aerosols, *J. Geophys. Res.-Atmos.*, 111, D12304, doi:10.1029/2005jd006852, 2006.
- Lindzen, R. S. and Farrell, B.: A simple approximate result for the maximum growth-rate of baroclinic instabilities, *J. Atmos. Sci.*, 37, 1648–1654, 1980.
- Malm, W. C., Schichtel, B. A., Pitchford, M. L., Ashbaugh, L. L., and Eldred, R. A.: Spatial and monthly trends in speciated fine particle concentration in the United States, *J. Geophys. Res.-Atmos.*, 109, D03306, doi:10.1029/2003jd003739, 2004.
- Meehl, G. A., Stocker, T. F., Collins, W. D., Friedlingstein, P., Gaye, A. T., Gregory, J. M., Kitoh, A., Knutti, R., Murphy, J. M., Noda, A., Raper, S. C. B., Watterson, I. G., Weaver, A. J., and Zhao, Z.-C.: Global climate projections, in: *Climate change 2007: The physical science basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, Cambridge, UK and New York, NY, USA, 2007.
- Mickley, L. J., Jacob, D. J., Field, B. D., and Rind, D.: Effects of future climate change on regional air pollution episodes in the United States, *Geophys. Res. Lett.*, 31, L24103, doi:10.1029/2004gl021216, 2004.
- Murazaki, K. and Hess, P.: How does climate change contribute to surface ozone change over the United States?, *J. Geophys. Res.-Atmos.*, 111, D05301, doi:10.1029/2005jd005873, 2006.
- Nakicenovic, N. and Swart, R.: Special report on emissions scenarios: A special report of Working Group III of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, New York, 599 pp., 2000.
- Park, R. J., Jacob, D. J., and Logan, J. A.: Fire and biofuel contributions to annual mean aerosol mass concentrations in the United States, *Atmos. Environ.*, 41, 7389–7400, 2007.
- Pinto, J. G., Ulbrich, U., Leckebusch, G. C., Spanghel, T., Reyers, M., and Zacharias, S.: Changes in storm track and cyclone activity in three sres ensemble experiments with the ECHAM5/MPI-OM1 GCM, *Clim. Dynam.*, 29, 195–210, 2007.
- Pye, H. O. T., Liao, H., Wu, S., Mickley, L. J., Jacob, D. J., Henze, D. K., and Seinfeld, J. H.: Effect of changes in climate and emissions on future sulfate-nitrate-ammonium aerosol levels in the United States, *J. Geophys. Res.-Atmos.*, 114, D01205, doi:10.1029/2008jd010701, 2009.
- Rind, D., Lerner, J., Jonas, J., and McLinden, C.: Effects of resolution and model physics on tracer transports in the NASA Goddard Institute for Space Studies general circulation models, *J. Geophys. Res.-Atmos.*, 112, D09315, doi:10.1029/2006jd007476, 2007.
- Spracklen, D. V., Mickley, L. J., Logan, J. A., Hudman, R. C., Yevich, R., Flannigan, M. D., and Westerling, A. L.: Impacts of climate change from 2000 to 2050 on wildfire activity and carbonaceous aerosol concentrations in the western United States, *J. Geophys. Res.-Atmos.*, 114, D20301, doi:10.1029/2008jd010966, 2009.
- Tagaris, E., Manomaiphiboon, K., Liao, K. J., Leung, L. R., Woo, J. H., He, S., Amar, P., and Russell, A. G.: Impacts of global climate change and emissions on regional ozone and fine particulate matter concentrations over the United States, *J. Geophys. Res.-Atmos.*, 112, D14312, doi:10.1029/2006jd008262, 2007.
- Tai, A. P. K., Mickley, L. J., and Jacob, D. J.: 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, 2010.

- Tai, A. P. K., Mickley, L. J., Jacob, D. J., Leibensperger, E. M., Zhang, L., Fisher, J. A., and Pye, H. O. T.: 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, *Atmos. Chem. Phys.*, 12, 3131–3145, doi:10.5194/acp-12-3131-2012, 2012.
- Tebaldi, C., Mearns, L. O., Nychka, D., and Smith, R. L.: Regional probabilities of precipitation change: A Bayesian analysis of multimodel simulations, *Geophys. Res. Lett.*, 31, L24213, doi:10.1029/2004gl021276, 2004.
- Tebaldi, C., Smith, R. L., Nychka, D., and Mearns, L. O.: Quantifying uncertainty in projections of regional climate change: A Bayesian approach to the analysis of multimodel ensembles, *J. Climate*, 18, 1524–1540, 2005.
- Thishan Dharshana, K. G., Kravtsov, S., and Kahl, J. D. W.: Relationship between synoptic weather disturbances and particulate matter air pollution over the United States, *J. Geophys. Res.-Atmos.*, 115, D24219, doi:10.1029/2010jd014852, 2010.
- Ulbrich, U., Pinto, J. G., Kupfer, H., Leckebusch, G. C., Spanghel, T., and Reyers, M.: Changing Northern Hemisphere storm tracks in an ensemble of IPCC climate change simulations, *J. Climate*, 21, 1669–1679, doi:10.1175/2007jcli1992.1, 2008.
- Ulbrich, U., Leckebusch, G. C., and Pinto, J. G.: Extra-tropical cyclones in the present and future climate: A review, *Theor. Appl. Climatol.*, 96, 117–131, 2009.
- Wilks, D. S.: *Statistical methods in the atmospheric sciences*, 3rd ed., International geophysics series, 100, Elsevier/Academic Press, Amsterdam, Boston, xix, 676 pp., 2011.
- Wu, S., Mickley, L. J., Kaplan, J. O., and Jacob, D. J.: Impacts of changes in land use and land cover on atmospheric chemistry and air quality over the 21st century, *Atmos. Chem. Phys.*, 12, 1597–1609, doi:10.5194/acp-12-1597-2012, 2012.
- Yin, J. H.: A consistent poleward shift of the storm tracks in simulations of 21st century climate, *Geophys. Res. Lett.*, 32, L18701, doi:10.1029/2005gl023684, 2005.
- Yue, X., Mickley, L. J., Logan, J. A., and Kaplan, J. O.: Ensemble projections of wildfire activity and carbonaceous aerosol concentrations over the western United States in the mid-21st century, in preparation, 2012.