

## Supplement

**Table S1.** Models from the Coupled Model Intercomparison Project Phase 5 (CMIP5) used for this study.

Model Name	Institute
ACCESS1-0	Commonwealth Scientific and Industrial Research,Organization (CSIRO) and Bureau of Meteorology (BOM), Australia
BCC-CSM1-1	Beijing Climate Center, China Meteorological Administration
BNU-ESM	College of Global Change and Earth System Science, Beijing Normal University
CanESM2	Canadian Centre for Climate Modelling and Analysis
CNRM-CM5	Centre National de Recherches Météorologiques / Centre Européen de Recherche et Formation Avancée en Calcul Scientifique
CSIRO-Mk3.6.0	Commonwealth Scientific and Industrial Research Organization in collaboration with Queensland Climate Change Centre of Excellence
GFDL-CM3	<a href="#">NOAA Geophysical Fluid Dynamics Laboratory</a>
GISS-E2-R	<a href="#">NASA Goddard Institute for Space Studies</a>
HadGEM2-CC	Met Office Hadley Centre (additional HadGEM2-ES realizations contributed by Instituto Nacional de Pesquisas Espaciais)
HadGEM2-ES	Met Office Hadley Centre (additional HadGEM2-ES realizations contributed by Instituto Nacional de Pesquisas Espaciais)
INM-CM4	Institute for Numerical Mathematics
IPSL-CM5A-LR	Institut Pierre-Simon Laplace
IPSL-CM5A-MR	Institut Pierre-Simon Laplace
IPSL-CM5B-LR	Institut Pierre-Simon Laplace
MIROC-ESM	Japan Agency for Marine-Earth Science and Technology, Atmosphere and Ocean Research Institute (The University of Tokyo), and National Institute for Environmental Studies
MIROC-ESM-CHEM	Japan Agency for Marine-Earth Science and Technology, Atmosphere and Ocean Research Institute (The University of Tokyo), and National Institute for Environmental Studies
MIROC5	Atmosphere and Ocean Research Institute (The University of Tokyo), National Institute for Environmental Studies, and Japan Agency for Marine-Earth Science and Technology
MRI-CGCM3	Meteorological Research Institute
NorESM1-M	Norwegian Climate Centre

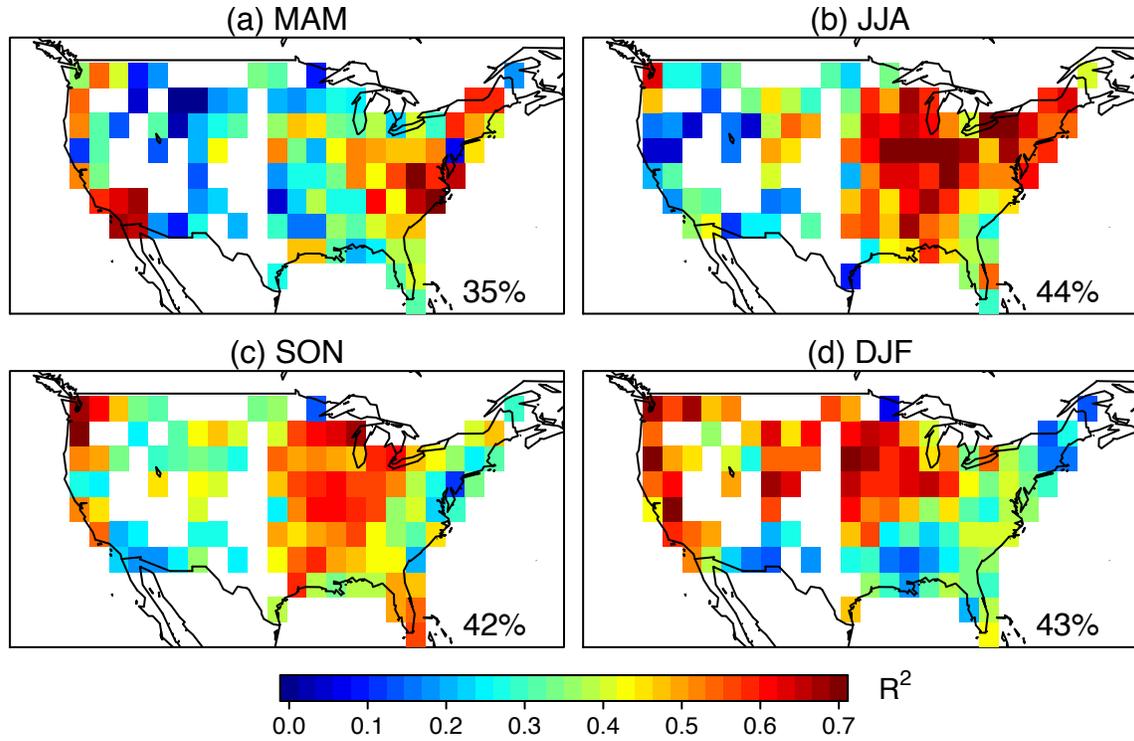
**Table S2.** Models from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) used for this study (Lamarque et al, 2013).

Model	Type	Resolution(lat/lon/#levels), top level	Modelling Center
NCAR-CAM3.5	CCM	1.875/2.5/L26, 3.5 hPa	NCAR,USA
GFDL-AM3	CCM	2/2.5/L48, 0.017 hPa	UCAR/NOAA,GFDL, USA
MIROC-CHEM	CCM	2.8/2.8/L80, 0.003 hPa	FRCGC, JMSTC Japan
GISS-ModelE2*	CCM	2/2.5/L40, 0.14 hPa	NASA-GISS,USA

\*We use an updated GISS simulation relative to their ACCMIP contributions, forced in atmospher-only mode using the ACCMIP emissions (Lamarque et al., 2010), observed daily sea-surface temperatures and sea-ice from Reynolds et al. (2007), and with winds nudged to the Modern-Era Retrospective Analysis for Research and Applications (MERRA) meteorological reanalysis (Rienecker et al., 2011).

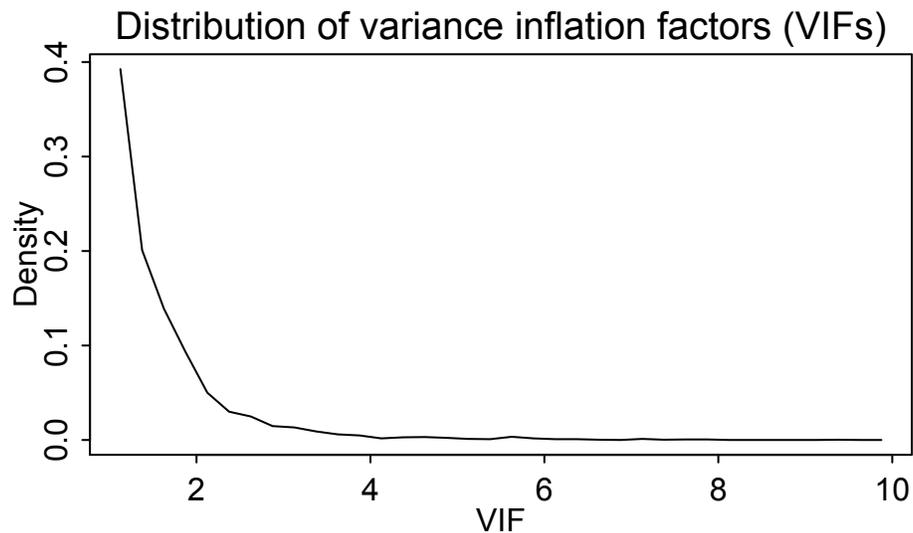
## 1. Cross-validation results in different seasons

Cross-validated  $R^2$  between observed and predicted  $PM_{2.5}$  in different seasons



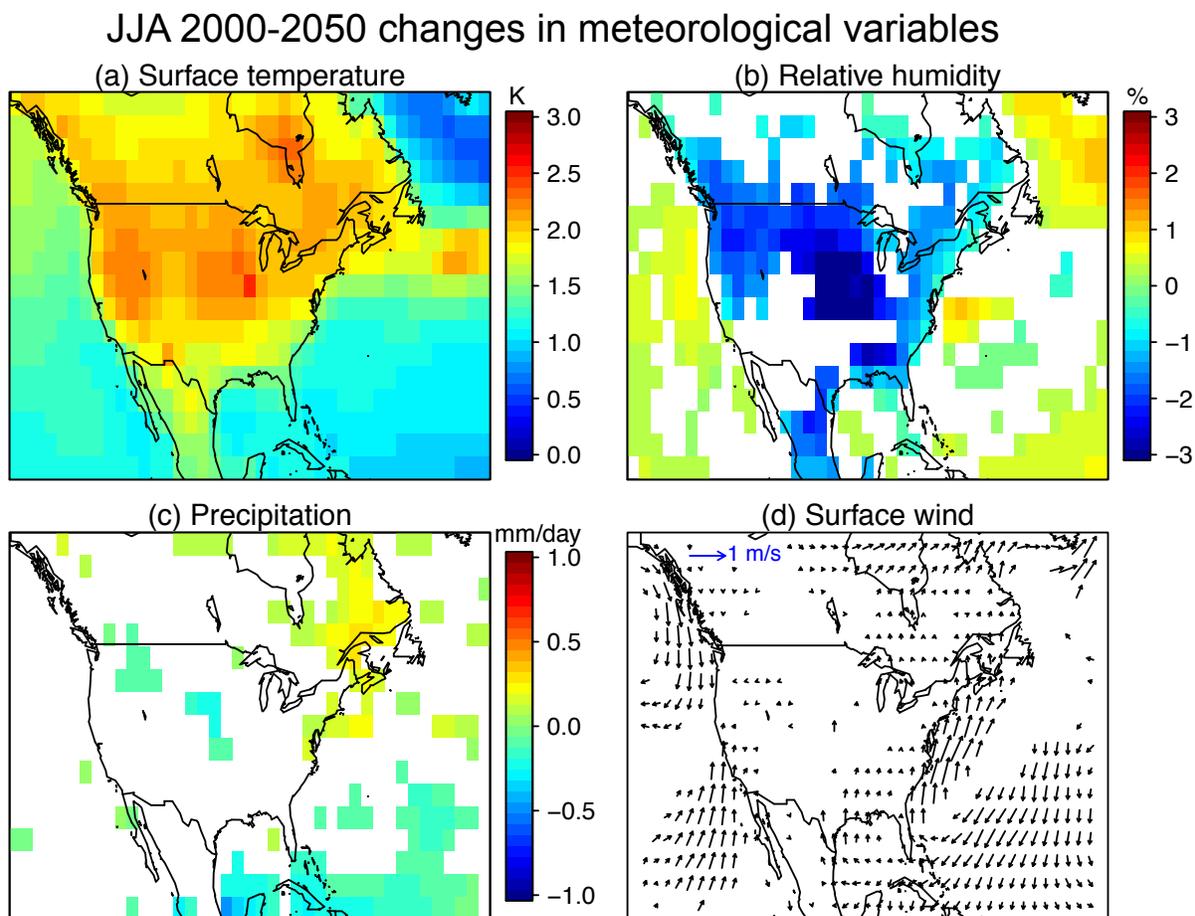
**Figure S1.** Cross-validated coefficients of determination ( $R^2$ ) between observed and predicted 1999-2013 monthly  $PM_{2.5}$  in different seasons across the United States, calculated with both local meteorology and patterns of synoptic circulation. Spatially averaged coefficients of determination are shown inset.

## 2. Distribution of variance inflation factors (VIFs)



**Figure S2.** The distribution of variance inflation factors (VIFs) of all variables in each gridbox and each month, calculated from the regression model using the best variable combination of both local meteorology and synoptic patterns.

### 3. Changes of meteorological variables from 2000 to 2050



**Figure S3.** Seasonal changes in June-July-August (a) mean temperature, (b) relative humidity, (c) precipitation and (d) surface wind field from 2000-2019 to 2050-2069, as projected by 19 CMIP5 climate models following the RCP4.5 scenario. White space indicates regions where fewer than 14 of the models show a consistent sign of change.

# DJF 2000-2050 changes in meteorological variables

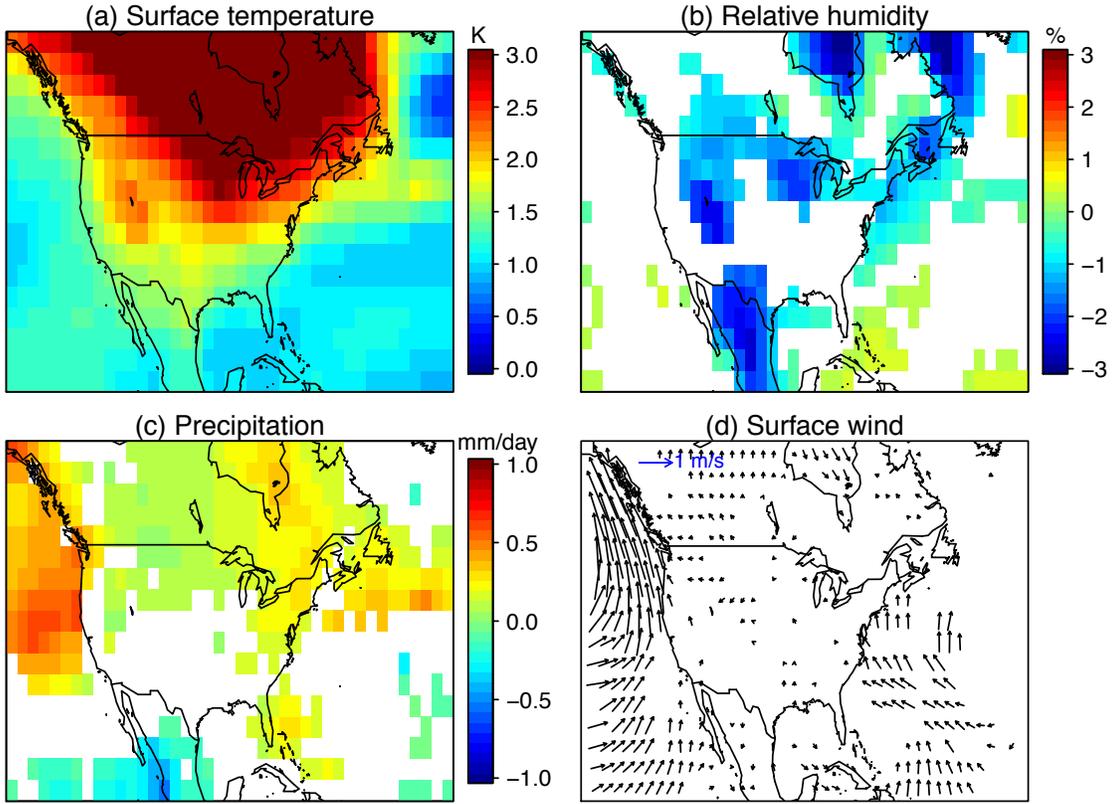
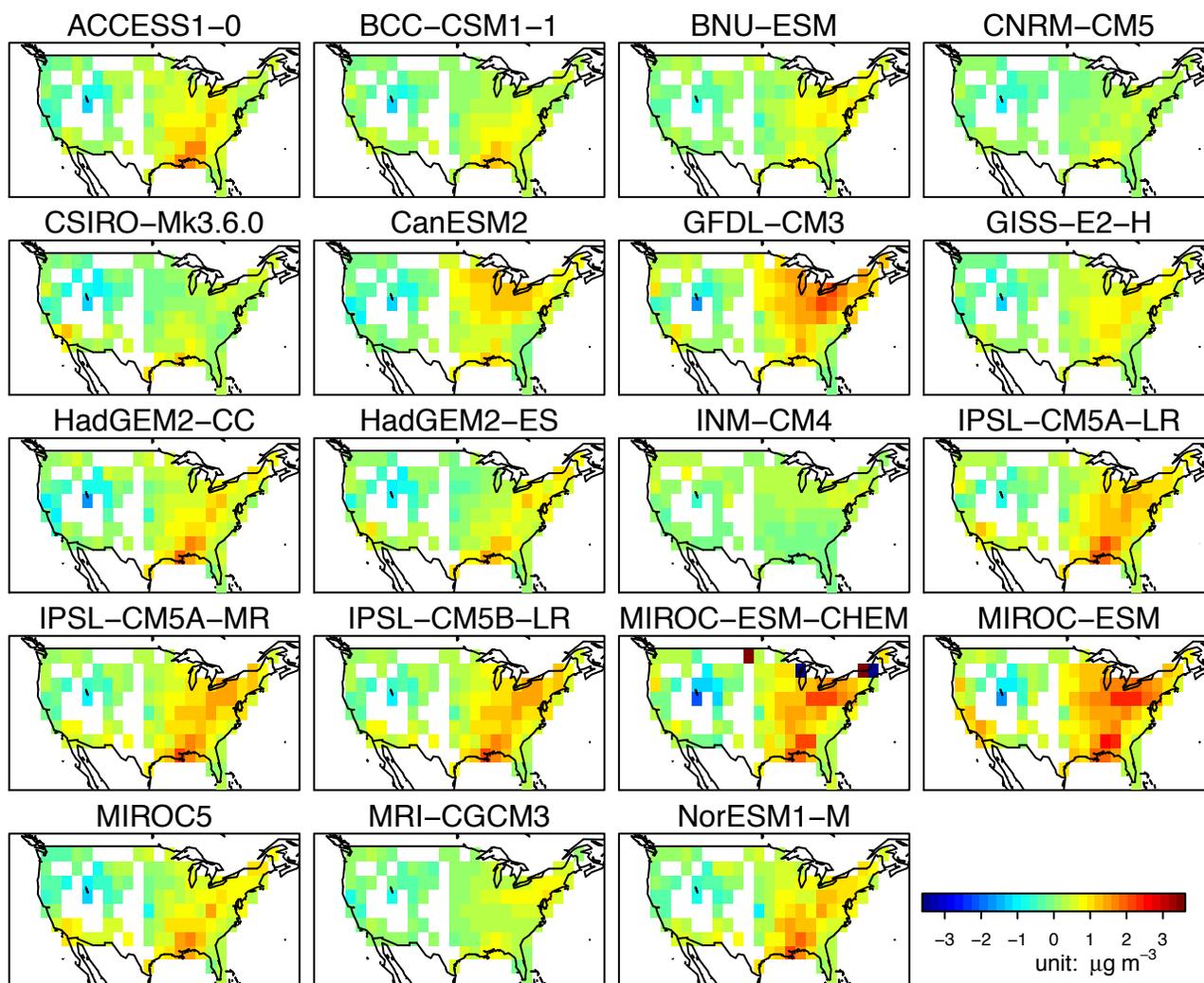


Figure S4. Same as Figure S3 but for December-January-February.

#### 4. Projected PM<sub>2.5</sub> changes in each CMIP5 model

Effects of 2050s climate change on annual mean PM<sub>2.5</sub> in each CMIP5 model



**Figure S5.** Effects of climate change from 2000-2019 to 2050-2069 on annual mean PM<sub>2.5</sub> concentrations, calculated with observed relationships of PM<sub>2.5</sub> and meteorology and with meteorology projected by each of the 19 CMIP5 models. White areas denote the regions with no PM<sub>2.5</sub> observations. For those models providing an ensemble of simulations for the RCP4.5 scenario, only one simulation was chosen for application to our model.

### 5. The 90<sup>th</sup> and 10<sup>th</sup> percentile changes in annual mean PM<sub>2.5</sub> by 2050s

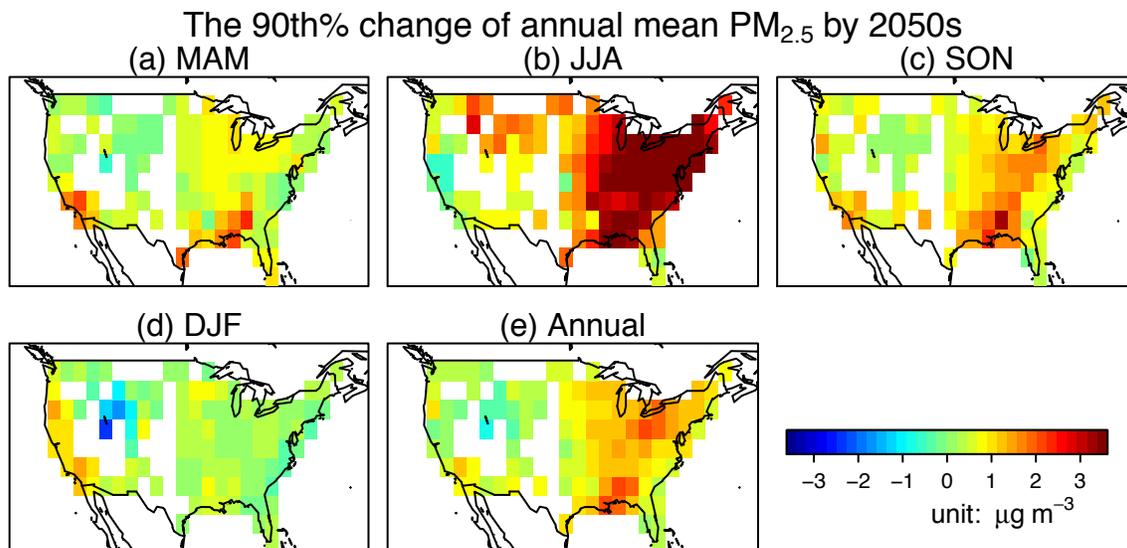


Figure S6. Same as Figure 4, but for the 90<sup>th</sup> percentile changes of PM<sub>2.5</sub> concentrations, calculated with meteorology projected by the ensemble of 19 CMIP5 models.

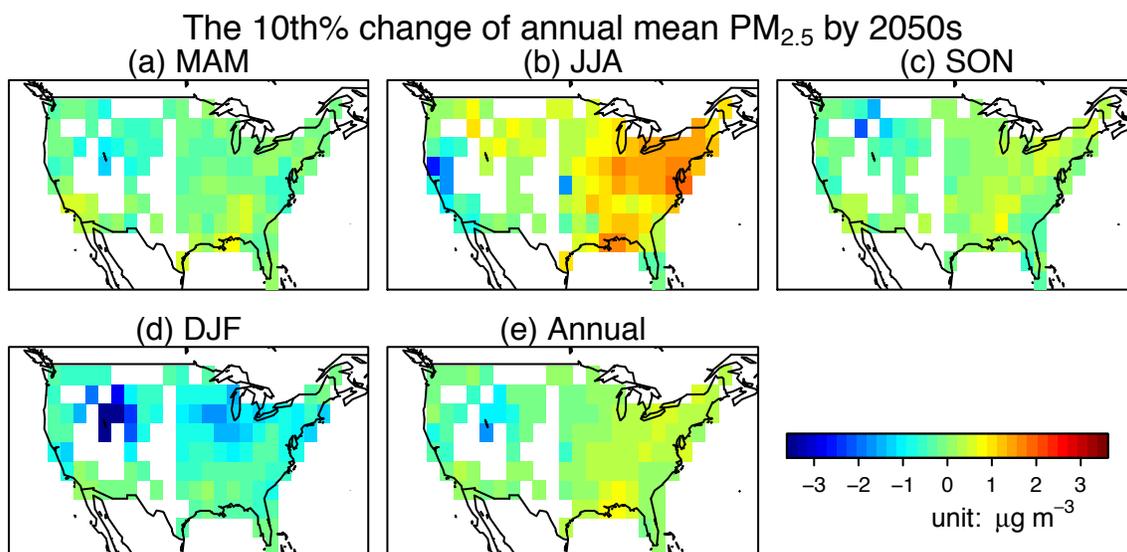


Figure S7. Same as Figures 4 and S7, but for the 10<sup>th</sup> percentile changes of PM<sub>2.5</sub> concentrations, calculated with meteorology projected by the ensemble of 19 CMIP5 models.

6. Time series of regional PM<sub>2.5</sub> changes from 2000 to 2069

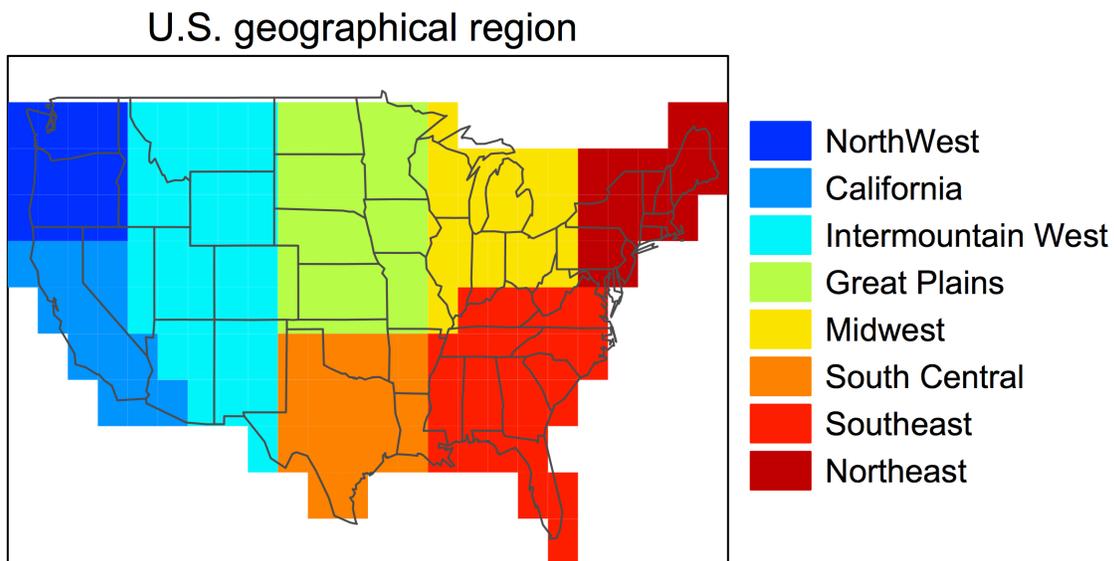
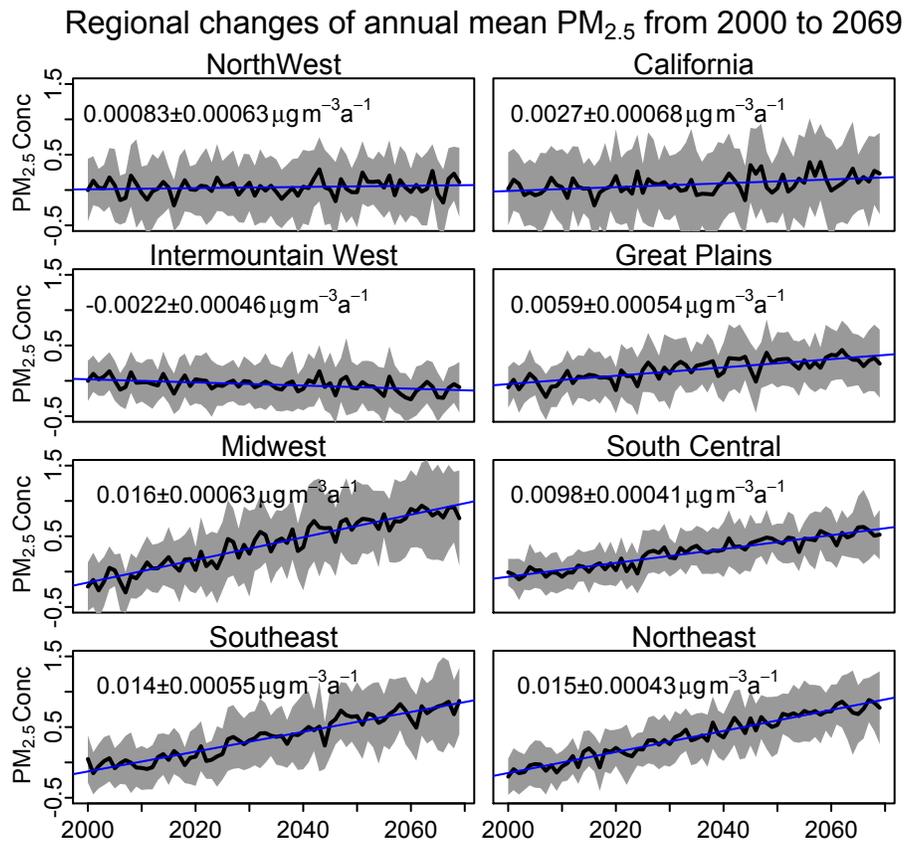


Figure S8. The eight U.S. geographical regions used for Figures S9-11.



**Figure S9.** Regional trends in annual mean PM<sub>2.5</sub> concentrations from 2000 to 2069, calculated with observed relationships of PM<sub>2.5</sub> and meteorology and with meteorology projected by an ensemble of 19 CMIP5 models. Shading denotes one standard deviation the mean change across models. For those regions with significant trends in PM<sub>2.5</sub>, the slopes of the timeseries over the 70-yr timeframe are shown inset. Figure S8 defines the eight regions.

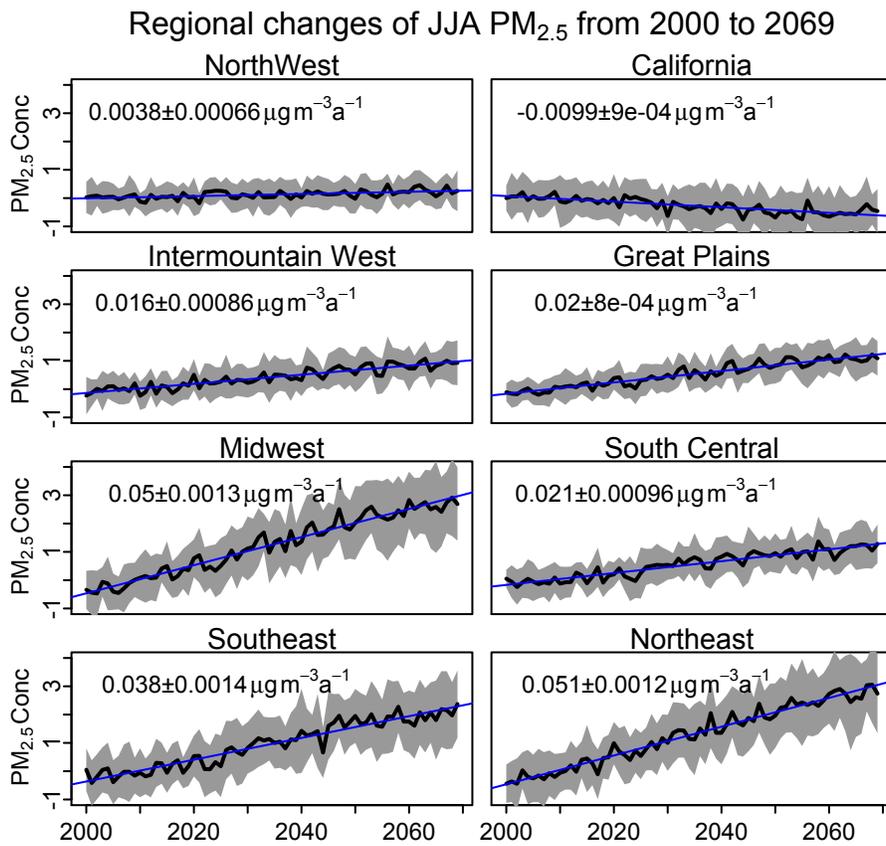


Figure S10. Similar as Figure S9, but for trend in JJA  $PM_{2.5}$  concentrations.

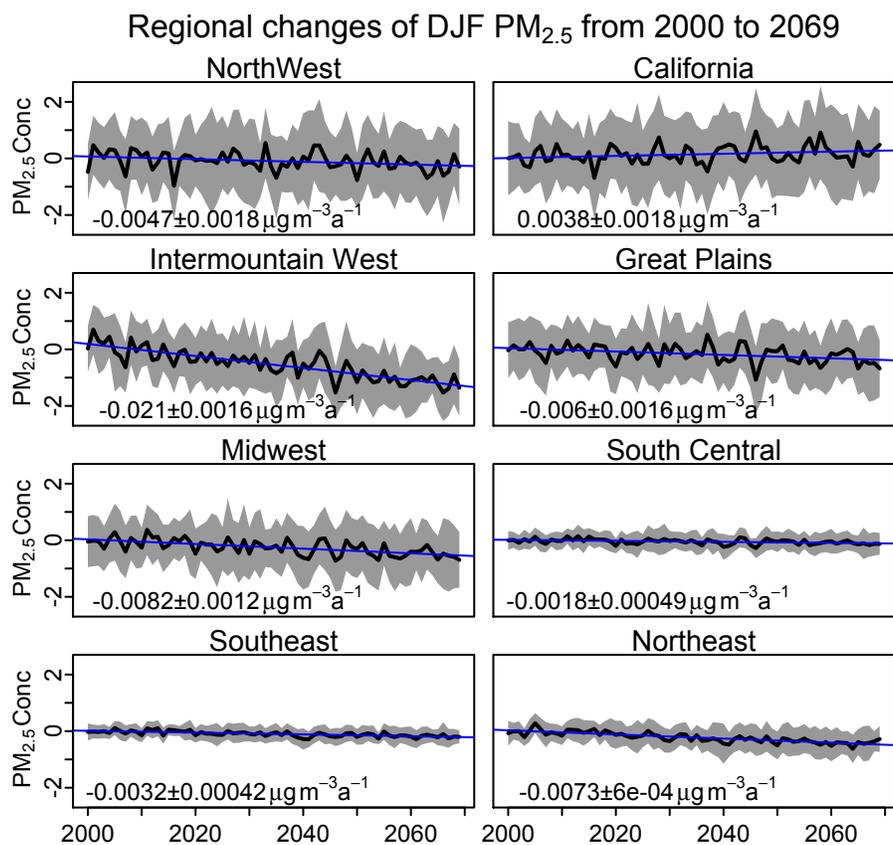
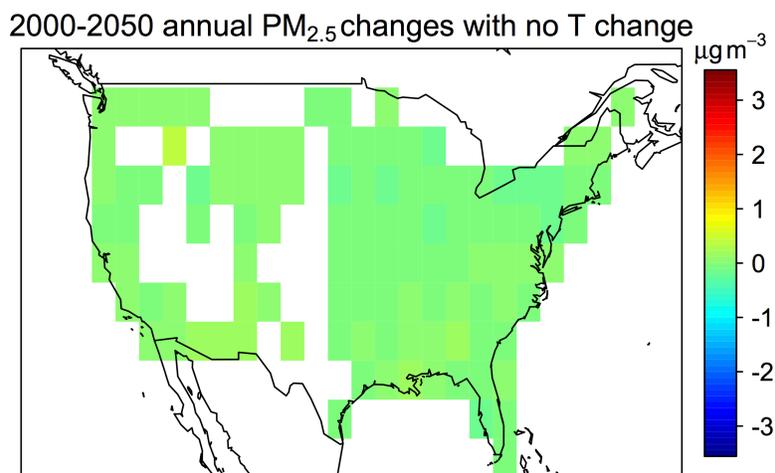


Figure S11. Similar as Figure S9, but for trends in DJF  $PM_{2.5}$  concentrations.

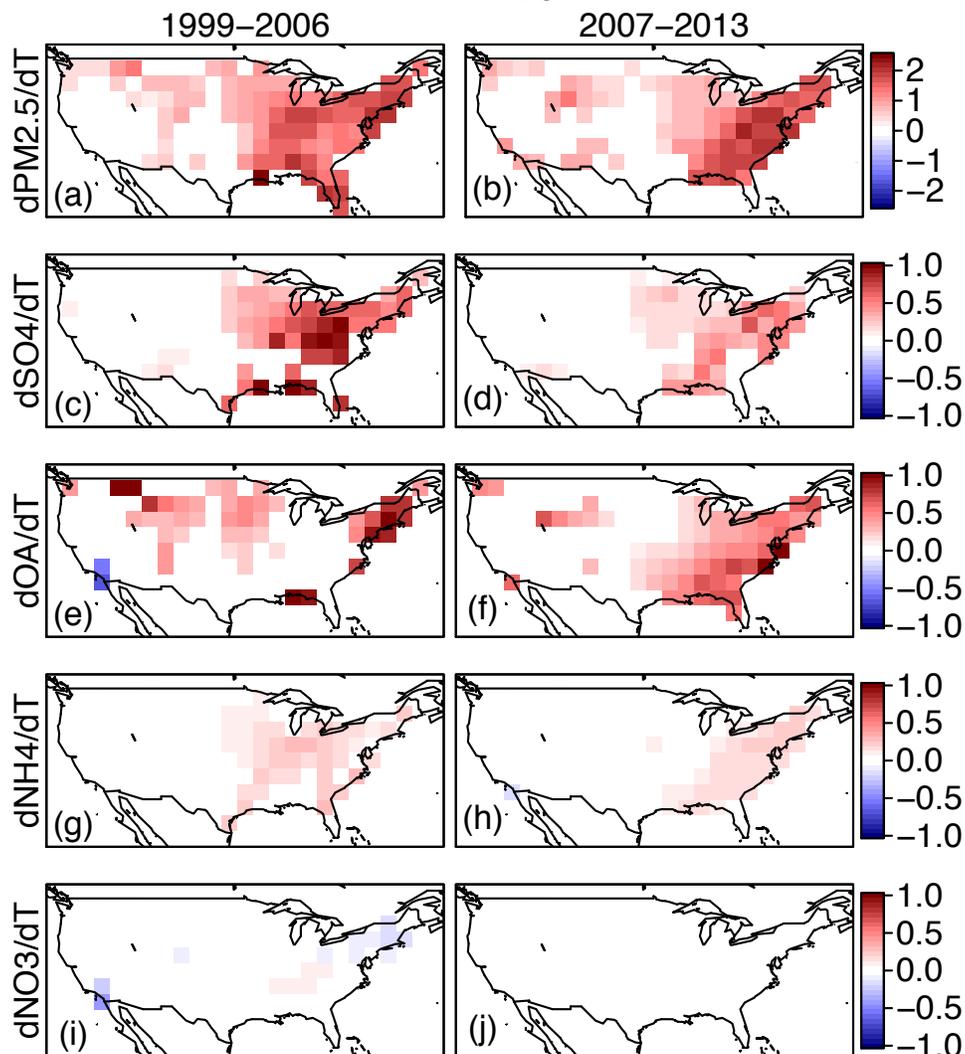
**7. Projected  $PM_{2.5}$  changes in the future climate excluding the influence from changing temperature.**



**Figure S12.** Response of annual mean  $PM_{2.5}$  concentrations to 2000-2050 climate change when changes in surface temperature are excluded. The figure shows the mean response as projected by an ensemble of 19 CMIP5 models. For this sensitivity test, surface temperatures in the statistical model for 2050-2069 are kept the same as for 2000-2019. White areas indicate regions with no  $PM_{2.5}$  observations.

## 8. Slopes of JJA $\text{PM}_{2.5}$ and temperature in 1999-2006 and 2007-2013

Slopes of JJA  $\text{PM}_{2.5}$  and temperature in AQS for 1999-2006 and 2007-2013 ( $\mu\text{g m}^{-3} \text{K}^{-1}$ )



**Figure S13.** The slopes of detrended (a-b) monthly mean  $\text{PM}_{2.5}$  and (c-j) different  $\text{PM}_{2.5}$  components with surface air temperature for 1999-2013 summer months. Left column shows slopes for 1999-2006 with relatively high  $\text{NO}_x$  emissions, and right column shows slopes for 2007-2013 with relatively low  $\text{NO}_x$  emissions. Organic aerosol (OA) in Panel (e-f) is inferred from the measured organic carbon (OC) component using an OA/OC mass ratio of 1.8 (Canagaratna et al., 2015). White areas indicate either missing data or grid boxes where the slope is not significant at the 0.10 level. We note that the observation network has fewer sites in 1999 and 2000 than more recent years.