The $PM_{2.5}$-climate penalty: quantifying sensitivities in a global climate model

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Abstract

Climate change can influence fine particulate matter concentrations ($PM_{2.5}$) through changes in air pollution meteorology. The extent to which climate change can exacerbate or alleviate air pollution in the future is an important aspect of robust climate and air pollution policy decision-making. To examine the influence of climate on $PM_{2.5}$, we use the Geophysical Fluid Dynamics Laboratory Coupled Model version 3 (GFDL CM3), a fully-coupled chemistry-climate model, combined with future emissions and concentrations provided by the four Representative Concentration Pathways (RCPs). For each of the RCPs, we conduct future simulations in which emissions of aerosols and their precursors are held at present-day levels while other climate forcing agents evolve in time, such that only climate (and thus meteorology) can influence $PM_{2.5}$ surface concentrations. We find a small increase in global

$PM_{2.5}$ of about 0.21 μg m^{-3} (5%) for RCP8.5, a scenario with maximum warming. Changes in global $PM_{2.5}$ are at a maximum in the fall and are mainly controlled by sulfate followed by organic aerosol with minimal influence of black carbon. RCP2.6 is the only scenario that projects a decrease in global $PM_{2.5}$ with future climate changes, albeit only by -0.06 μg m^{-3} (1.5%) by the end of the 21st century. Regional and local changes in $PM_{2.5}$ are larger, reaching upwards of 2 μg m^{-3} for polluted (eastern China) and dusty (western Africa) locations on an annually averaged basis in RCP8.5. Using multiple linear regression, we find that future $PM_{2.5}$ concentrations are most sensitive to local temperature, surface wind, and precipitation, in that order. Fine particulate matter concentrations are robustly positively associated with temperature, while negatively related with precipitation and wind speed. Present-day (2006-2015) modeled sensitivities of $PM_{2.5}$ to meteorological variables are evaluated against observations and found to agree reasonably well with observed sensitivities (within 10-50% over the eastern United States for several variables), although the modeled $PM_{2.5}$ is less sensitive to precipitation than in the observations due to weaker convective scavenging. We conclude that the hypothesized "climate penalty" of future increases in fine particulate matter is relatively minor on a global scale compared to the influence of emissions on $PM_{2.5}$ concentrations.

1 Introduction

In 2012, poor air quality resulted in up to 7 million premature deaths worldwide (WHO, 2014). Particles less than 2.5 μm in diameter, known as $PM_{2.5}$, are the major contributor to poor air quality and have been linked numerous times to increases in mortality (Dockery et al., 1993; Dominici et al., 2006; Pope, 1999; Pope et al., 2004, 2006). The chemical composition of $PM_{2.5}$ is dependent on many factors, but in general, inorganic species such as sulfate, nitrate, and ammonium along with a plethora of organic species comprise the majority of the particulate mass (Jimenez et al., 2009a). Emissions of particulate matter and their precursors, from sources such as energy use and biomass burning, are the dominant contributors to $PM_{2.5}$ concentrations in the atmosphere (Chan and Yao, 2008; West et al., 2013). However, $PM_{2.5}$ levels depend on meteorological factors such as temperature, precipitation, and wind speed, suggesting that $PM_{2.5}$ can be influenced by climate change. For example, wet deposition is the main sink for atmospheric particulate matter, so increases in precipitation (particularly frequency of precipitation) will lead to decreases in particle concentrations (Bernard et al., 2001; Dawson et al., 2007; Jacob and Winner, 2009; Jimenez-Guerrero et al., 2011; Tai et al.,

2010). $PM_{2.5}$ is also thought to be negatively sensitive to wind speed, as decreases in ventilation will cause build-up of $PM_{2.5}$ levels (Jacob and Winner, 2009; Porter et al., 2015). $PM_{2.5}$ may be positively associated with relative humidity (RH), as RH increases result in increased aqueous uptake of semi-volatile materials (such as nitrate and organic aerosol); however, an anti-correlation has been reported between sulfate and cloud cover, which calls the positive RH-$PM_{2.5}$ relationship into question (Koch et al., 2003; Wise and Comrie, 2005). Warmer temperatures can increase reaction rates for oxidation of sulfur dioxide (SO_2) to sulfate (SO_4) aerosol by the hydroxyl radical (OH), but warmer temperatures are also less favorable for condensation of semi-volatile materials to the aerosol phase. With increased warming, variable changes in precipitation, and other significant climate changes projected over the remainder of the 21st century, it is important to understand the effect that these changes might have on $PM_{2.5}$ and air quality.

Previous modeling studies on the effects of climate change on future air quality have yielded mixed results. The majority of these studies have employed offline meteorological fields that are then fed into a chemical transport or general circulation model. A review by Jacob and Winner (2009) reported mostly an increase in $PM_{2.5}$ from climate change, ranging from ±0.1 – 1 μg m^{-3}, depending on the time horizon, region, and species (Heald et al., 2008; Jacobson, 2008; Liao et al., 2006; Racherla and Adams, 2006; Spracklen et al., 2009; Tagaris et al., 2007; Unger et al., 2006). However, two papers featured in the review, Pye et al. (2009) and Avise et al. (2009), reported a change of -0.3 to +0.3 μg m^{-3} (2050 vs 2000, US, sulfate, annual mean) and -1 μg m^{-3} (2050 vs 2000, US, $PM_{2.5}$, July mean), respectively. More recently, Jiang et al. (2013) reported an annually averaged change in 2050 of -1.5 to +0.8 μg m^{-3} for aerosol concentrations in eastern China due to climate change alone. Megaritis et al. (2013) reported strictly decreases in aerosol concentrations in Central Europe in 2050 due to climate change because of dominating decreases in ammonium nitrate and organic aerosol levels. Reflecting the uncertainty and discrepancies in these various estimates, the Intergovernmental Panel on Climate Change (IPCC) assigns “no confidence level” to the overall impact of climate change on $PM_{2.5}$, and only “low confidence” to the expectation that increases in precipitation lead to decreases in $PM_{2.5}$ (Kirtman et al. 2013) because of confounding effects of the timing and location of changes in precipitation and $PM_{2.5}$ (Fang et al., 2011). Although there is a significant amount of variability of the magnitude and even the sign of the effect of climate on future air quality, virtually all studies agree that the magnitude of climate-driven changes in surface $PM_{2.5}$ are dwarfed by emissions-driven changes (Fiore et

al., 2015; Gonzalez-Abraham et al., 2015; Hedegaard et al., 2013; Jiang et al., 2013; Kelly et al., 2012; Trail et al., 2014; Unger et al., 2006; Val Martin et al., 2015; West et al., 2013).

Here, we build upon previous studies in several ways. First, we utilize a fully coupled chemistry-climate model (GFDL CM3, see Sect. 2) with online meteorology, whereas almost all previous studies have relied on offline (input) meteorology. The offline meteorology approach allows for shorter simulations (i.e., beginning and end simulations for the time domain). A shortcoming of this method is that the short simulations make it more difficult to separate climate change-induced signal versus climate variability in surface $PM_{2.5}$ concentrations. Our work employs a three-member ensemble of 95-year simulations, allowing us to better estimate the climate-forced signal in surface $PM_{2.5}$ from the noise. Second, we conduct these full 21st century simulations using all four of the Representative Concentration Pathways (RCPs) for future climate and emissions (modified such that aerosol and precursor emissions are held fixed at 2005 levels). To our knowledge this is the first study to analyze the effect of future climate on $PM_{2.5}$ using all four RCPs in fully transient simulations in a chemistry-climate model. Third, we focus on both global analyses and specific regions, which exhibit larger $PM_{2.5}$ changes and also contain large population centers. Fourth, we use a multiple linear regression (MLR) model to estimate sensitivities of surface $PM_{2.5}$ to several meteorological parameters, allowing for analysis of the magnitude and sign of the $PM_{2.5}$-meteorology association, following the method of Tai et al. (2010). We also compare the present-day modeled MLR sensitivities to observed sensitivities based on $PM_{2.5}$ and meteorological data taken over the United States from 1998-2008 (Tai et al., 2010). Finally, we include both annual and seasonal analyses as well as speciated results for individual $PM_{2.5}$ components. Although previous studies may have partially covered some of our goals, we aim to provide a comprehensive, robust analysis as an additional piece of evidence to a growing understanding of the effect of climate change on surface $PM_{2.5}$.

2 Models and analysis

2.1 GFDL CM3

We employ the Geophysical Fluid Dynamics Laboratory Coupled Model version 3 (GFDL CM3) in this work (Donner et al., 2011; Naik et al., 2013). CM3 is a fully coupled chemistry-climate model, featured in the latest Coupled Model Intercomparison Project (CMIP) as well as the Intergovernmental Panel on Climate Change Assessment Report 5 (IPCC AR5). A brief

description of the model is provided below. CM3 has been extensively evaluated against observations including many of the model variables considered in this work. Further model description and evaluation details can be found in Donner et al. (2011) as well as Naik et al. (2013) and references therein.

The model consists of a cubed sphere finite-volume dynamical core with a horizontal grid consisting of 6 faces with roughly a 200-km by 200-km spatial resolution. Vertical resolution consists of 48 levels extending from the surface up to about 80 km (0.01 hPa). Anthropogenic emissions of reactive gases and aerosols and their precursors are based on estimates from Lamarque et al. (2010) for the historical period (1860-2000) and from van Vuuren et al. (2012) and Lamarque et al. (2011) for the RCP projections (2006-2100). Tropospheric chemistry is interactive with emissions and radiation modules and is based on Horowitz et al. (2003) with updates from Horowitz (2006). Aerosol species include sulfate, black carbon, primary organic aerosol (POA), secondary organic aerosol (SOA), sea salt, and mineral dust. Sulfate is formed via oxidation of sulfur dioxide and dimethyl sulfide (DMS) by the hydroxyl radical (OH), ozone (O_3), and hydrogen peroxide (H_2O_2). SOA formation is parameterized by apportioning fixed yields of terpene emissions and butane oxidation as biogenic and anthropogenic SOA, respectively. The model's SOA source of 40 Tg C yr^{-1} globally (natural plus anthropogenic) is likely to be an underestimate (Heald et al., 2008). Additionally, CM3 lacks inclusion of sophisticated treatment of SOA chemistry and includes few SOA precursors. Advanced treatment of SOA chemistry and precursor species is undergoing development at GFDL. Hydrophobic organic carbon aerosols (OA) and black carbon (BC) aerosols are converted to hydrophilic aerosol with an e-folding time of 1.44 days. Sea salt and mineral dust aerosol are treated with a five-section size distribution ranging from 0.1 to 10 μm dry radius, and have emissions that depend on model meteorology. Changes in climate do not feedback on biogenic VOC or wildfire emissions, or partitioning of semi-volatile organics from the gas phase into SOA, which are not temperature-dependent in this configuration of CM3. The model also does not include any changes in the dust source availability in response to climate-induced changes in soil moisture or vegetation. These omissions may lead to errors in our estimate of the total influence of climate change on future $PM_{2.5}$ levels (Heald et al., 2008; Spracklen et al., 2009). Particularly, increases in future wildfires and biogenic VOC emissions due to temperature changes will likely result in a larger estimate PM2.5 climate penalty.

Species included in our surface $PM_{2.5}$ definition are limited to sulfate, BC, OA, and dust. We do not include sea salt particles in our analysis as our focus is mostly on anthropogenic aerosols which impact human health more directly and comprise the vast majority of $PM_{2.5}$ in major world population centers (Jimenez et al., 2009b; Vallius et al., 2005; Yang et al., 2011). Dust and sea salt are also typically coarser in size such that a large portion of their mass would fall outside of the 2.5 μm upper size cutoff (Pérez et al., 2008). Dust and sea salt levels have been found to comprise no more than 10% of total $PM_{2.5}$ mass concentration in several Chinese megacities (Yang et al., 2011), but in five Mediterranean European cities, dust contributed anywhere between 5-32% of total $PM_{2.5}$ (Salameh et al., 2015). Thus, our inclusion of dust is important for estimating climate driven $PM_{2.5}$, especially considering dust emission changes are driven by climate (wind speed) alone in CM3. We do not currently consider nitrate aerosol in $PM_{2.5}$ changes, due to the relatively small current contribution of nitrate to total aerosol mass in CM3 and the simplified manner in which nitrate chemistry and thermodynamics is treated in the model (Paulot et al., 2016). However, in the future nitrate may play a larger role in $PM_{2.5}$ due to decreasing sulfate (Bellouin et al., 2011), and a more advanced treatment of nitrate including its interaction with radiation and clouds is undergoing development at GFDL (Paulot et al., 2016).

2.2 Simulations with 21st century climate scenarios

We use GFDL CM3 to evaluate the effect of future climate changes on $PM_{2.5}$ concentrations. Since these simulations were previously carried out by Westervelt et al. (2015), we follow the same simulation naming convention as used in that work. Simulations made up of three ensemble members for each RCP were run from 2006-2100. These simulations are denoted RCPx.x_2005AER, where x.x = 2.6, 4.5, 6.0, or 8.5 for each of the four RCPs. In each simulation, anthropogenic and biomass burning emissions of aerosols and their precursors are held fixed at 2005 levels throughout the 95 years of the simulation, while other climate forcing agents including greenhouse gases follow the RCP scenario, thus allowing only changes in climate and meteorology to perturb aerosol levels. Tracer concentrations and climate variables for each of the three ensemble members for each RCP were also initialized from the ending conditions of 1860-2005 transient historical runs in CM3. We use these historical simulations from 1980-2005 in which aerosol emissions are allowed to vary in time for comparison with climate-only changes in the 21st century. We present all results as ensemble means.

The Representative Concentration Pathways (RCPs) contain emissions projections for all long- and short-lived climate forcers, including the aerosol and aerosol precursor species SO_2, OA, and BC. The four pathways include a strong mitigation scenario (RCP2.6), two stabilization scenarios in which radiative forcing stabilizes shortly after 2100 (RCP4.5 and RCP6), and one high emissions/no mitigation scenario (RCP8.5). Details of the implementation of the RCPs into GFDL CM3 can be found in Westervelt et al. (2015). Since recent GHG emissions are tracking at or above RCP8.5 (Peters et al., 2012; Sanford et al., 2014) we will focus mainly on RCP8.5_2005AER, and RCP8.5_2005AER should be viewed as the most realistic scenario as of now, leading to the largest warming and other climate signals. We also present RCP2.6_2005AER as a lower bound. The other RCPs are included mostly in the Supplementary Information.

2.3 Multiple linear regression

In order to determine the magnitude and sign of the effect of various climate parameters on surface $PM_{2.5}$ concentrations, we use a multiple linear regression (MLR) approach (Tai et al., 2010). Unlike a correlation coefficient from a single linear regression, MLR allows for analysis of multiple independent variables (meteorological variables) affecting the dependent variable ($PM_{2.5}$ concentrations) at the same time. The form of the model is:

$$y = \beta_0 + \sum_{i=1}^{6} \beta_i x_i + interaction\ terms \quad \text{(Equation 1)}$$

In Eq. 1, the dependent variable y represents $PM_{2.5}$ concentrations (annual or seasonal mean) and is a function of the coefficient β_0 (y-intercept), the slope coefficients β_i, and the independent variables x_i. Each of the independent variables x_i represents a climate or meteorological variable that determines the values of y, or the $PM_{2.5}$ concentration. We chose six meteorological variables for inclusion in the MLR model: surface temperature (K), precipitation (mm d^{-1}), total cloud amount (in all vertical layers) (%), 10 m wind speed magnitude (m s^{-1}), sea level pressure (hPa) and relative humidity (%). Both the modeled $PM_{2.5}$ and meteorological variables are monthly averages beginning in January 2006 and ending in December 2100. The slope coefficients β_i represent the amount of change in $PM_{2.5}$ concentrations for a unit change in the meteorological variables x_i if all other climate variables x are held constant. These values can be interpreted as "sensitivities" of $PM_{2.5}$ to a change in climate, and have units of μg m^{-3} D^{-1}, where D refers to the units for the particular meteorological variable (e.g., temperature in K). Positive coefficients refer to an increase in

$PM_{2.5}$ concentrations for an increase in the meteorological variable (positive correlation), whereas negative values refer to decreases in $PM_{2.5}$ (anti-correlation). Interaction terms, which refer to the product of two or more independent variables and represent the higher-order linear effects when meteorological variables are interacting with each other in affecting $PM_{2.5}$, are found to be small and thus excluded from the analysis.

We also present MLR slope coefficients for observations over the United States domain for comparison against model results. Meteorological monthly mean data (derived from daily data) from 1998 to 2008 from the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis 1 is used for the MLR estimatation (http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.html) (Kalnay et al., 1996). The Reanalysis data is gridded on a 2.5º by 2.5º latitude-longitude grid. For $PM_{2.5}$, we calculated monthly means of daily mean surface concentrations of total $PM_{2.5}$ from the EPA Air Quality System (EPA-AQS, http://www.epa.gov/aqs), which operates a network of about 1000 stations in the United States. The modeled and observed $PM_{2.5}$ and meteorological values were detrended and deseasonalized by subtracting the three-month running means from each original month. Details on both the meteorological and PM data can be found in Tai et al. (2010), which applied the same methodology but on the daily deviation of $PM_{2.5}$ from a 30-day moving mean to focus on the synoptic scale.

3 $PM_{2.5}$ changes

We first present our $PM_{2.5}$ changes over time with our 2005 fixed aerosol emissions simulations (RCPx.x_2005AER) in Sect 3.1, Fig. 2, and Table 1. Then, spatial distributions of the change of $PM_{2.5}$ from the beginning of the 21st century to the end ($\Delta PM_{2.5}$, Eq. 2) are presented in Figs. 3 and 4. Finally, we present the regional results in Sect. 3.2 and Fig. 3 and Tables 2 and 3. Region definitions are provided in Fig. 1.

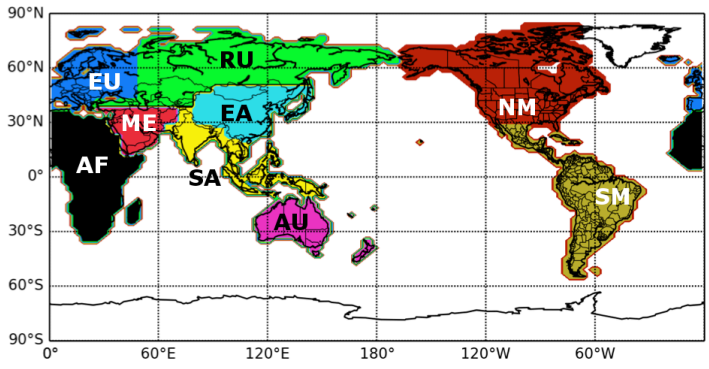

Figure 1: Region definitions. North America (NM), South and Central America (SM), Europe (EU), Russia (RU), East Asia (EA), South Asia (SA), Australia (AU), Africa (AF), Middle East (ME)

We report values of population-weighted surface $\Delta PM_{2.5}$ as the change in concentration from a ten-year ensemble mean of 2006-2015 to a ten-year ensemble mean of 2091-2100, i.e. the change from the beginning to the end of the 21st century. All results are for surface-level $PM_{2.5}$. In equation form:

$$\Delta PM_{2.5} = PM_{2.5}(2091-2100) - PM_{2.5}(2006-2015) \quad \text{(Equation 2)}$$

Population weighted concentrations were calculated by summing the product of population and concentration for a specific grid cell divided by the total population:

$$C_w = \frac{\sum_j C_j P_j}{\sum_j P_j} \quad \text{(Equation 3)}$$

where C_j indicates concentration at each grid cell, P_j is population at each grid cell, and C_w is the weighted concentration. Population data was obtained from the CIESEN 2005 Gridded Population product (http://sedacciesincolumbiaedu/plue/gpw).

3.1 Global mean $\Delta PM_{2.5}$ from 1980-2100

Figure 2 shows the time evolution of globally and annually averaged speciated and total surface $PM_{2.5}$ from 1980-2100. "Total" $PM_{2.5}$ refers here to the sum of sulfate (and its associated ammonium), black carbon, organic aerosol, and dust less than 2.5 μm in diameter. Results from the historical runs are shown in purple from 1980-2005, followed by results from the four RCPx.x_2005AER simulations for the rest of the 21st century. For the historical time series in $PM_{2.5}$, aerosol and precursor emissions were not fixed; they instead were allowed to vary according to the historical emissions inventory of Lamarque et al. (2010). Starting in 2005, all anthropogenic and biomass burning aerosol emissions are held fixed at 2005 levels for the remainder of the simulation of the 21st century, allowing us to isolate only the effect of meteorology. The results plotted for the 25-year period 1980-2005, during which emissions vary, has about the same range or an even larger range of $PM_{2.5}$ concentrations ($\Delta PM_{2.5}$ of ~0.15 μg m^{-3}) as the effect of meteorology over the entire 21st century for any of the RCPs (0.21 μg m^{-3} for RCP8.5_2005AER). This is particularly noticeable in the black

carbon and organic aerosol time series (Fig. 2, b and c) in which the variable-emissions trend from 1980-2005 increases dramatically, followed by little to no increase for the next 95 years due to meteorology only. For sulfate (Fig. 2a) and the sum of sulfate, BC, OA, and dust, the $\Delta PM_{2.5}$ from 2006 to 2100 is of comparable magnitude to the emissions-driven changes of $\Delta PM_{2.5}$ from 1980-2005, at least in RCP8.5 and RCP2.6. Thus, since a century of climate-driven changes in $PM_{2.5}$ are less than or equal to emissions-driven changes over only 25 years, we conclude that changes in emissions are the dominant driver for future $PM_{2.5}$ concentrations, with fairly minor meteorological changes, at least on the global, annual scale. In particular, black carbon aerosols are very weakly affected by precipitation perhaps owing to their lower water affinity than sulfate aerosols, which are most strongly affected. Additionally, production of secondary aerosols such as sulfate is climate-dependent, whereas emissions of primary aerosols such as BC are not as affected by climate.

Westervelt et al. (2015) found similar results for aerosol optical depth (AOD) which was far more sensitive to RCP emissions reductions than climate changes with emissions held fixed (see Fig. S3 of Westervelt et al. (2015)).

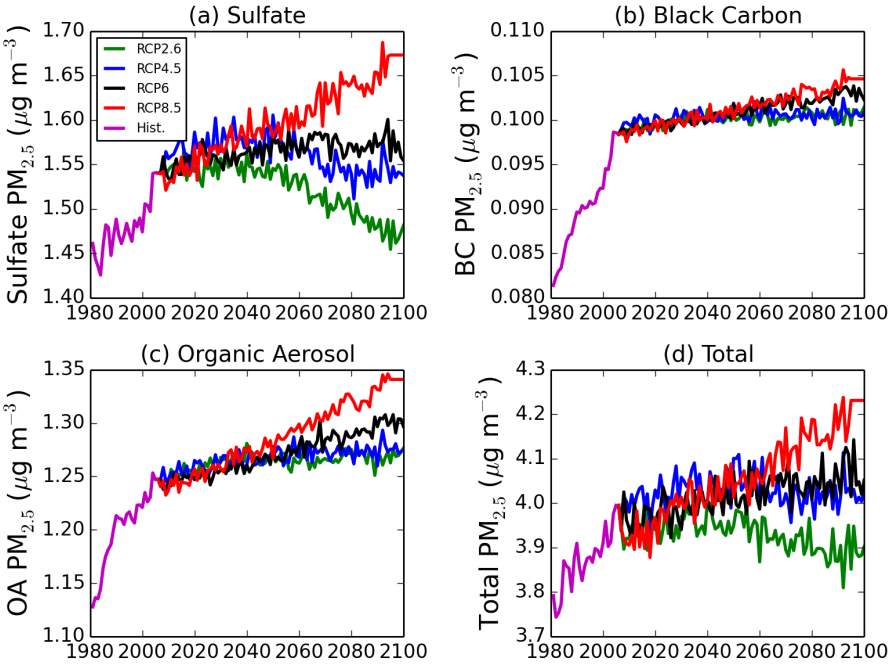

Figure 2: Global, annual mean trend in $PM_{2.5}$ concentration for sulfate (a), black carbon (b), organic aerosol (c), and total (including fine dust) (d) for each of the RCPx.x_2005AER simulations. Emissions vary in time only for the 1980-2005 pre-RCP time period.

As shown in Fig. 2, $PM_{2.5}$ concentrations are weakly affected by meteorology on a global scale. The high-warming scenario, RCP8.5, projects a consistent but slight increase in $PM_{2.5}$ as the 21st century progresses. This increase in $PM_{2.5}$ can be considered a "climate penalty" (Wu et al., 2008), due to increases in $PM_{2.5}$ caused solely by climate change (Fang et al., 2013). Table 1 presents the $\Delta PM_{2.5}$ concentrations differences for ten-year averages from the beginning to the end of the century. Table 1 and Fig. 2 clearly show that the meteorological impact on $PM_{2.5}$ is strongly dependent on the RCP scenario. For example, total $\Delta PM_{2.5}$ for RCP2.6_2005AER is -55.4 ng m^{-3} and +210 ng m^{-3} for RCP8.5_2005AER, indicating that even the sign of the change (increase or decrease in $PM_{2.5}$) is dependent on the particular climate change scenario. Exceptions to this are OA and BC, which have a positive $\Delta PM_{2.5}$ for all RCPs. The order of the four trends over the 21st century, especially visible in the sulfate and total $PM_{2.5}$ concentrations (Fig. 2, a and d), mirrors the order of warming in each of the simulations. In general, RCP8.5_2005AER results in the largest $\Delta PM_{2.5}$, followed by RCP6_2005AER, RCP4.5_2005AER, and RCP2.6_2005AER. This order comes expected, as RCP8.5_2005AER results in the largest amount of warming and other meteorological change (e.g., precipitation), while RCP2.6_2005AER features stringent climate policies and thus relatively little warming (Table 1).

Our globally averaged $\Delta PM_{2.5}$ results are comparable with Fang et al. (2013), although their estimate (0.28 μg m^{-3}) using a prototypical version of GFDL AM3 (atmospheric component only) and an older set of emissions scenarios (IPCC SRES) is slightly above the RCP8.5_2005AER global average of 0.21 μg m^{-3}. This small positive enhancement in $PM_{2.5}$ due to climate change also falls in the range of PM sensitivity to climate ($\pm 0.1 - 1$ μg m^{-3}) as defined by the Jacob and Winner (2009) review paper and reiterated by Dawson et al. (2014).

3.2 Spatial distribution of seasonal and annual $\Delta PM_{2.5}$

Spatial distributions of seasonally and annually averaged total $\Delta PM_{2.5}$ (end of 21st century minus present day) are shown for RCP8.5_2005AER in Fig. 3.

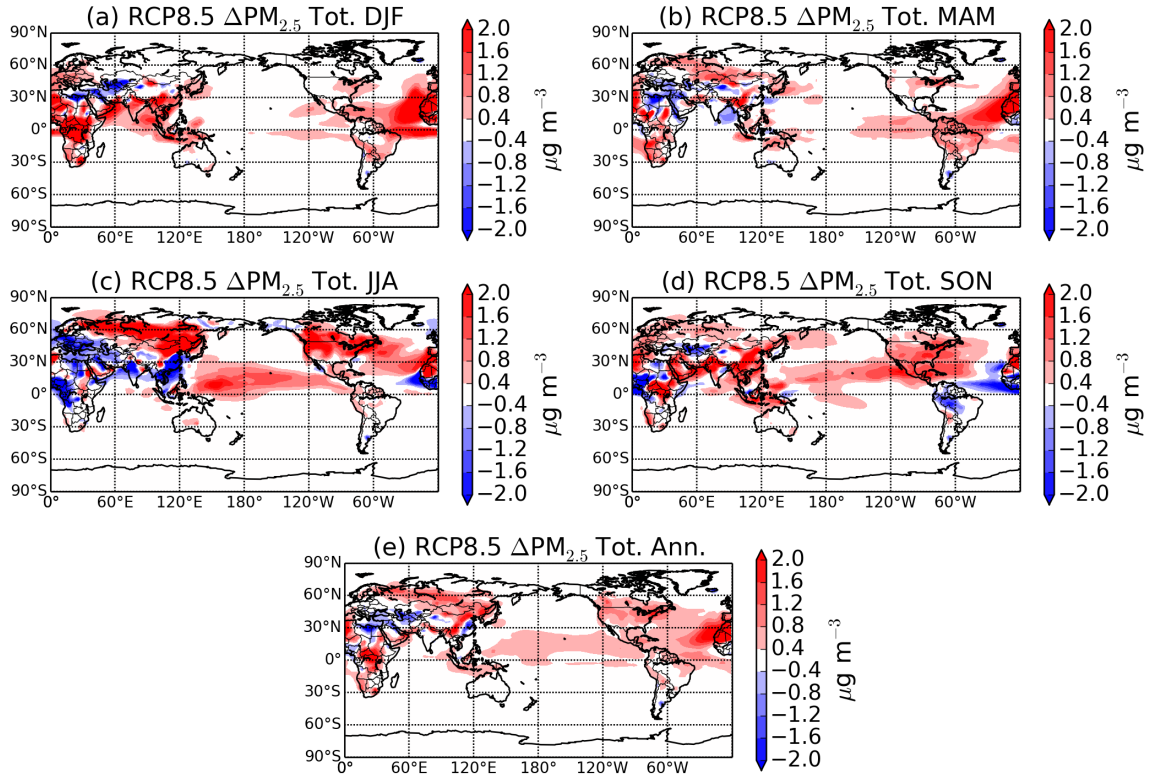

Figure 3: Seasonal (a-d) and annual (e) average $\Delta PM_{2.5}$ for a difference of 2091-2100 average and a 2006-2015 average in RCP8.5_2005AER

In each season, the impact of meteorology on $PM_{2.5}$ concentrations is largely an increase in most regions, with some exceptions. While annual globally averaged $\Delta PM_{2.5}$ were less than 0.3 μg m^{-3}, over certain regions, annual concentration changes can reach 1-2 μg m^{-3}. Increases are typically found over populated continental regions such as Europe, China, and North America. Changes (mainly increases) over Western Africa reflect increases in dust $PM_{2.5}$ due to increases in wind speed. $PM_{2.5}$ increases over the tropical Pacific Ocean are caused by decreases in wet deposition in CM3 associated with a warming climate (Westervelt et al., 2015). In certain seasons, $PM_{2.5}$ concentration changes are even larger, resulting in a larger "climate penalty". For example, in the autumn months (SON, Fig. 3d) total $\Delta PM_{2.5}$ differences are upwards of +2 μg m^{-3}, with a global average increase of 0.31 μg m^{-3}, the largest of any season. Pye et al. (2009) also found peak aerosol concentrations in fall when looking solely at meteorological driven changes. In SON, sulfate $PM_{2.5}$ increases more than any other season with organic aerosol increases being at second highest values out of the four seasons, which explains the large increases in $PM_{2.5}$. Total $PM_{2.5}$ increases are at a minimum global mean value of 0.16 μg m^{-3} in the northern hemisphere summer months (JJA) due to a

corresponding minimum in the sulfate aerosol increases (dominant species by mass), despite a summertime peak in the increases for organic aerosol. Details of speciated seasonal results for sulfate, OA, and BC are shown in the Supplement Fig. S1. The seasonal averages reveal pockets of negative $\Delta PM_{2.5}$ (aerosol decreases) that are largely masked in the annual spatial distribution. We find that not only the magnitude but also the sign of $\Delta PM_{2.5}$ is highly dependent on the season in some areas. For example, South America exhibits positive $\Delta PM_{2.5}$ in DJF and SON but negative $\Delta PM_{2.5}$ in the other months. The speciated results (Fig. S1) reveal that these decreases are driven entirely by decreases in organic aerosol, as sulfate and black carbon either increase or do not change. This finding implies a strong meteorological influence on organic aerosol aerosol in the northern hemisphere winter (DJF) and fall (SON). Fang et al. (2013) also found decreases in $PM_{2.5}$ in South America and attributed the change to changing precipitation patterns. We confirm that $PM_{2.5}$ is negatively associated with precipitation in South America and globally, which is discussed in more detail in Sect. 4.

The spatial distribution of $\Delta PM_{2.5}$ in RCP2.6_2005AER differs from RCP8.5_2005AER (Fig. 4 vs. Fig. 3), with large decreases over most of the continents balanced with pockets of increases in RCP2.6_2005AER.

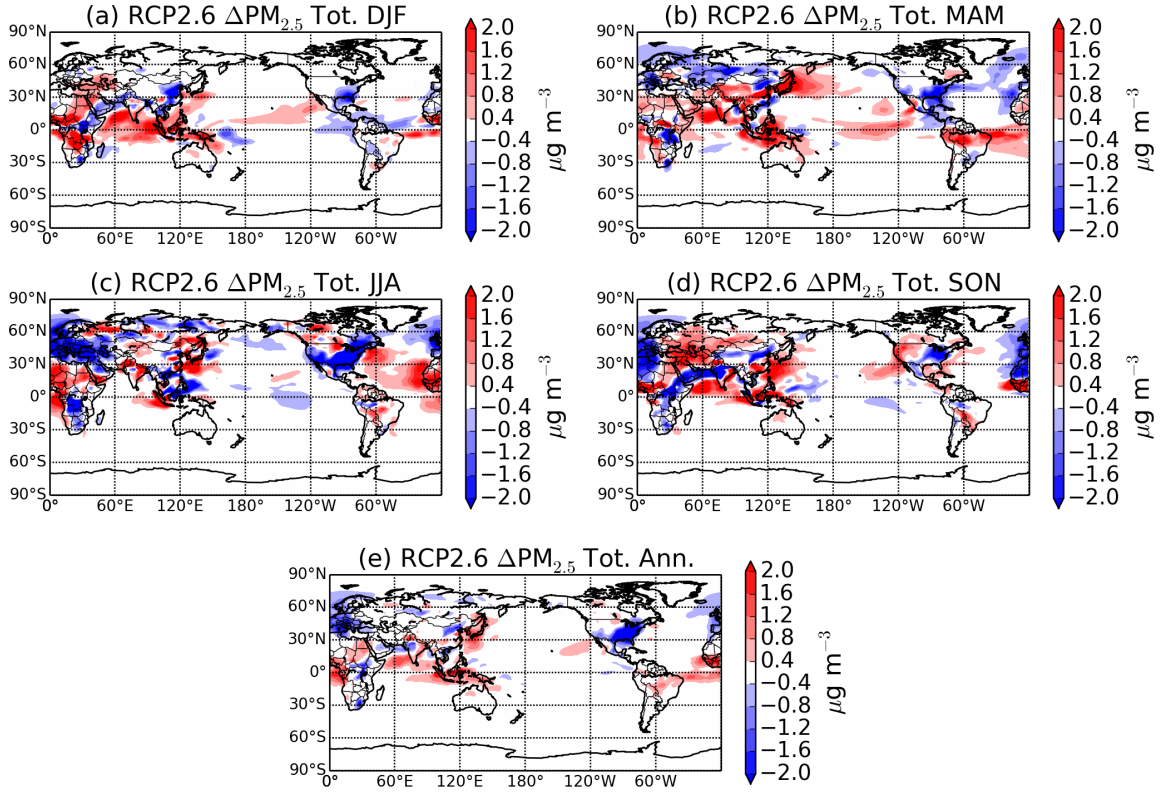

Figure 4: Same as Figure 3, but for RCP2.6_2005AER

Annually, this results in a slight overall decrease in global mean $PM_{2.5}$ due to meteorological changes (-0.03 μg m^{-3}), suggesting a slight "climate benefit" overall. Decreases are occurring largest over the summer months (JJA, Fig. 4c), in which concentration decreases are found over nearly all of Europe and US, for a global average of -0.08 μg m^{-3}. As shown in Supplement Fig. S2, the large summer decreases across Europe and the eastern US come largely from sulfate decreases, with secondary influence of organic aerosol. Globally averaged JJA sulfate $\Delta PM_{2.5}$ is about -0.08 μg m^{-3}, whereas BC and OA show essentially zero change globally, with decreases in some regions offset by increases. The large decreases in the eastern US and Europe are consistent across all seasons, with the slight exception of European $\Delta PM_{2.5}$ in the winter months (DJF). However, as found for RCP8.5_2005AER, $\Delta PM_{2.5}$ can be very different for the same location in different seasons. As is the case with RCP8.5_2005AER, the largest changes in magnitude (positive or negative) in global mean $PM_{2.5}$ occur in the summer and fall, with relative minima in the spring and winter. Annual and seasonal spatial distributions of $\Delta PM_{2.5}$ for RCP4.5_2005AER and RCP6.0_2005AER are shown in Supplemental Figs. S3 and S4, and fall between the results of RCP2.6_2005AER and RCP8.5_2005AER.

3.3 Regional average $\Delta PM_{2.5}$

Figure 5 presents the total $PM_{2.5}$ (sulfate + OA + BC + fine dust) annual mean time series from 1980-2100 for four key regions (North America, East Asia, South Asia, and Russia) as defined by Fig. 1. Total $\Delta PM_{2.5}$ is presented in Table 2 for RCP8.5_2005AER and Table 3 for RCP2.6_2005AER for all of the regions in Fig. 1. Four regions were chosen for further analysis because they represent some of the largest $PM_{2.5}$ changes driven by meteorology and include major population centers. As with Fig. 2, the historical model emissions are varying in time (not fixed) from 1980-2005. These historical trends are quite different for each of the regions. For example, North America had largely begun to enact and enforce air quality regulations by the latter part of the 20th century, which is reflected by a decreasing trend in the $PM_{2.5}$ concentrations. East and South Asia (Fig 5, b and c) $PM_{2.5}$ increases by up to 3 to 4 μg m^{-3}, reflecting rapid industrialization in these parts of the world. Concentrations of $PM_{2.5}$ in Russia (Fig. 5, panel d) first increase until about 1990 and then decrease strongly for the next decade, indicative of a slowing economy after the collapse of the Soviet Union. By the beginning of the 21st century, $PM_{2.5}$ are again trending upwards due to economic recovery in Russia.

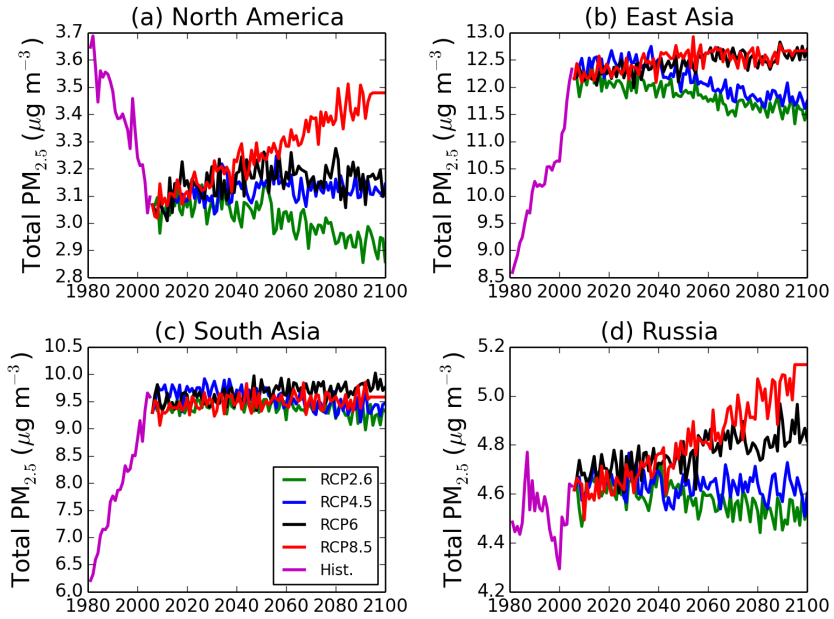

Figure 5: Annually averaged timeseries of total $PM_{2.5}$ concentration for each of the RCPx.x_2005AER simulations for the North American region (a), East Asia (b), South Asia (c), and Russia (d) (see Fig. 1 for region definitions).

Compared to the global time series in Fig. 2 and Table 1, 21st century meteorologically-driven changes in regional $PM_{2.5}$ are much larger. For example, for RCP8.5_2005AER East Asia (average of China, Japan, and Korea) has an annual average $\Delta PM_{2.5}$ of about 0.4 μg m^{-3}, and the value for Russia is 0.44 μg m^{-3} (see Table 2). The largest regional increase is in Africa at 0.64 μg m^{-3}, however this is one of the few regions where dust $PM_{2.5}$ concentrations increase due to climate change (wind speed increases), whereas they decrease significantly over East Asia, South Asia, Europe, Middle East, and Russia. Grid boxes over certain populous cities in these regions exhibit $\Delta PM_{2.5}$ of up to 1 to 2 μg m^{-3}, representing a more significant climate penalty. Though subject to uncertainty and variability in specific studies, epidemiologists have surmised up to a 6% and 11% increase in all-cause and cardiovascular mortality for every incremental 10 μg m^{-3} increase in daily $PM_{2.5}$ concentrations (Hoek et al., 2013), suggesting that our projected climate-driven concentration increases may exacerbate local air pollution related mortality. However, these meteorologically-driven $PM_{2.5}$ changes represent less than a 10% change in total $PM_{2.5}$ over highly polluted areas such as eastern China. Additionally, the linear relationship between increased mortality and $PM_{2.5}$ cited above does not hold in highly polluted environments with high base concentrations. Still, in less or

moderately polluted areas, such as the Pacific Northwest U.S., the relative increase in $PM_{2.5}$ can be as high as 40%.

As was the case with the global results, the total $\Delta PM_{2.5}$ from the beginning of the 21st century to the end is much smaller than the change from 1980-2005, highlighting the dominance of emissions over meteorology in driving $PM_{2.5}$ changes at the regional level. The order of the $PM_{2.5}$ concentrations in 2100 also reflects differences inherent to the RCPs, with RCP8.5_2005AER having the largest changes in meteorological values such as temperature, precipitation, etc., and therefore the largest impact of changing climate on $PM_{2.5}$. Regional increases in temperature and precipitation for each of the RCPs can be found in Westervelt et al. (2015). This order is most prevalent in the North America and Russia regions. $PM_{2.5}$ concentrations in South Asia increase the most in RCP6_2005AER, and East Asian $PM_{2.5}$ trend splits into two regimes in which RCP8.5_2005AER and RCP6.0_2005AER increase throughout the century nearly identically, while RCP4.5_2005AER and RCP2.6_2005AER decrease nearly identically by the same amount. Indeed the $\Delta PM_{2.5}$ for East Asia in RCP2.6_2005AER and RCP4.5_2005AER is -0.51 and -0.55 μg m^{-3}, and +0.40 and +0.43 μg m^{-3} for RCP8.5_2005AER and RCP6.0_2005AER (see Tables 2-3 and Tables S1-S2).

Speciated results shown in Tables 2-3 provide some insight into which components of $PM_{2.5}$ are driving the 21st century trends. For RCP8.5_2005AER, the increases are nearly half driven by sulfate and half driven by organic aerosol for many regions, with a few exceptions. South America has a trivial organic aerosol increase (0.014 μg m^{-3}) such that 87% of the $\Delta PM_{2.5}$ increase is due to sulfate, and Europe has a smaller sulfate increase such that only 35% of the increase is caused by sulfate increases. BC plays a small role in total PM2.5 changes due to a lack of atmospheric abundance by mass, contributing at most 6-9% in East and South Asia but significantly less elsewhere. For RCP2.6_2005AER, overall $\Delta PM_{2.5}$ concentrations are negative in all regions except Africa. This is entirely caused by strong sulfate decreases, which occur in all regions. On the other hand, positive values of organic aerosol $\Delta PM_{2.5}$ (increases) are still predicted for all but one region (South America). We therefore conclude that organic aerosol concentrations primarily increase due to climate change and that this increase is robust across virtually all RCPs and all regions. BC concentration changes, although minor, are also consistently positive. Sulfate $\Delta PM_{2.5}$ changes may be either positive or negative depending on the RCP (and thus the climate scenario). Sulfate $\Delta PM_{2.5}$ tends to be positive (increases) globally and across most regions for RCP8.5_2005AER and

RCP6.0_2005AER, the two scenarios with the most extreme climate changes. Sulfate $\Delta PM_{2.5}$ is largely negative (decreases) for RCP4.5_2005AER and RCP2.6_2005AER.

4 $PM_{2.5}$ dependence on meteorology

4.1 Modeled $PM_{2.5}$

In order to determine the relationship between changing meteorology and $PM_{2.5}$ changes, we use the multiple linear regression (MLR) model described in Sect. 2.3. Figures 6 and 7 show plots of the annual average coefficients β_i, or slope coefficients, for RCP8.5_2005AER and RCP2.6_2005AER for the 21st century. These coefficients are essentially "sensitivities" of $PM_{2.5}$ to unit changes in each of 6 meteorological variables assuming all other meteorological variables are held fixed. For a seasonal analysis of the $PM_{2.5}$ dependence on meteorological parameters, Figs. S5 and S6 show DJF and JJA averages for RCP8.5_2005AER. Figure 6 provides a broad look at the sign of the relationship between each meteorological variable and $PM_{2.5}$, with red values representing a positive PM-meteorological correlation, and blue values representing a negative correlation. The MLR model explains 50% to 80% of the variation in $PM_{2.5}$ for RCP8.5_2005AER, with the largest percentages found over the continents. "The correlation matrix shown in the supplemental information shows that each of the independent variables (the meteorological variables) are not strongly correlated with one another. Thus, we consider multicollinearity to be minimal."

For RCP8.5_2005AER, the $PM_{2.5}$-temperature relationship is robustly positive across the continents and oceans, with minor exceptions in locales of East and South Asia. This is consistent with studies compiled by Jacob and Winner (2009). In GFDL CM3, this positive correlation likely comes temperature-dependent reaction rates and changes in oxidant abundances, which can enhance oxidation of aerosol precursors (such as SO_2) with higher temperature, leading to more $PM_{2.5}$. There are other possible feedbacks of temperature on PM, such as the relationship between organic aerosol volatility and temperature, but these mechanisms are not included in this version of the model. $PM_{2.5}$ sensitivity to temperature can also manifest in changes in stagnation and circulation. Taking the average slope coefficient in the eastern US as 0.15 $\mu g\ m^{-3}\ K^{-1}$ and projected RCP8.5 temperature increases of 4 K leads to a $PM_{2.5}$ increase of 0.6 $\mu g\ m^{-3}$, which would represent most if not all of the observed $PM_{2.5}$ increase due to all meteorological variables in that region.

Similar scaling arguments can be made for the other meteorological parameters. $PM_{2.5}$ is also found to be sensitive to precipitation, which is consistently anti-correlated due to the large scavenging sink of wet deposition. Precipitation changes are typically more positive than negative from the beginning to the end of the 21st century (increases) although there is significant variation across regions. As expected, the relationship between $PM_{2.5}$ and precipitation is largely negative (anti-correlated), indicating that areas with precipitation decreases yield $PM_{2.5}$ increases and vice versa. The largest negative sensitivities (Fig. 6b) are found over the Middle East, parts of Africa, and Europe. The strong sensitivity of $PM_{2.5}$ to precipitation over most of Africa and the Middle East, some of the driest regions in the world, suggests that precipitation is an important $PM_{2.5}$ sink even in areas where it infrequently happens. Using an average value of the slope coefficients of roughly -1.0 $\mu g\ m^{-3}\ mm^{-1}$ d and a precipitation increase in Europe of about 0.1 mm d^{-1} results in a -0.1 $\mu g\ m^{-3}$ change in $PM_{2.5}$ due to precipitation only, suggesting that the precipitation effect is moderately strong but likely not larger than the influence of temperature (~0.5 $\mu g\ m^{-3}$) in CM3. The moderate sensitivity of $PM_{2.5}$ to precipitation is in part caused by a weak convective scavenging sink in this version of CM3 (Paulot et al., 2016; Fang et al. 2011).

We also find in the model that $PM_{2.5}$ is consistently anti-correlated with wind speed (Fig. 6d), as negative values of the slope coefficients appear over most continental regions. Lower wind speeds lead to less ventilation, causing an increase in $PM_{2.5}$, as is discussed in Jacob and Winner (2009). Similarly, higher wind speeds cause more mixing and dilution, which may decrease $PM_{2.5}$ levels. An exception is in Saharan Africa, the Middle East, and Australia, which have a large positive correlation between wind speed and $PM_{2.5}$. This is likely due to the strong wind speed dependence of dust emissions. Sea salt is not included in $PM_{2.5}$ in this study, but would result in large positive correlations with wind speed over the Southern Ocean. Typical wind speed changes from the beginning to the end of the century are much larger over the oceans and are typically no greater than ±0.4 m s^{-1} over the continents. With wind speeds in CM3 slightly decreasing through the 21st century over the continents (due to decreases in temperature gradients), sensitivities of -0.5 $\mu g\ m^{-3}\ m^{-1}$ s could lead to $PM_{2.5}$ increases of up to 0.2 $\mu g\ m^{-3}$, which is a substantial portion of projected regional and global $\Delta PM_{2.5}$. Since relatively humidity, sea level pressure, and cloud cover sensitivities are not substantial (discussed in detail below), we conclude that after temperature, wind speed decreases may be the second-most important contributor to $PM_{2.5}$ increases.

Fig. 6e shows the slope coefficients β_i for the relationship between $PM_{2.5}$ and sea level pressure. We find that the slopes are positive nearly everywhere, with somewhat modest sensitivities (~0.1-0.2 μg m^{-3} hPa^{-1}) except for in the tropical Pacific near Malaysia, Indonesia, and the Philippines. Despite being a typically low-pressure region at present, future shifting of the Hadley circulation and the ITCZ due to greenhouse gas forcing could lead to an increase in sea level pressure which in turn causes the strong sensitivity of $PM_{2.5}$ over the remainder of the 21st century seen here. Generally, high-pressure systems can lead to stable atmospheric conditions and a shallow mixing layer, resulting in poor ventilation and therefore an increase in $PM_{2.5}$.

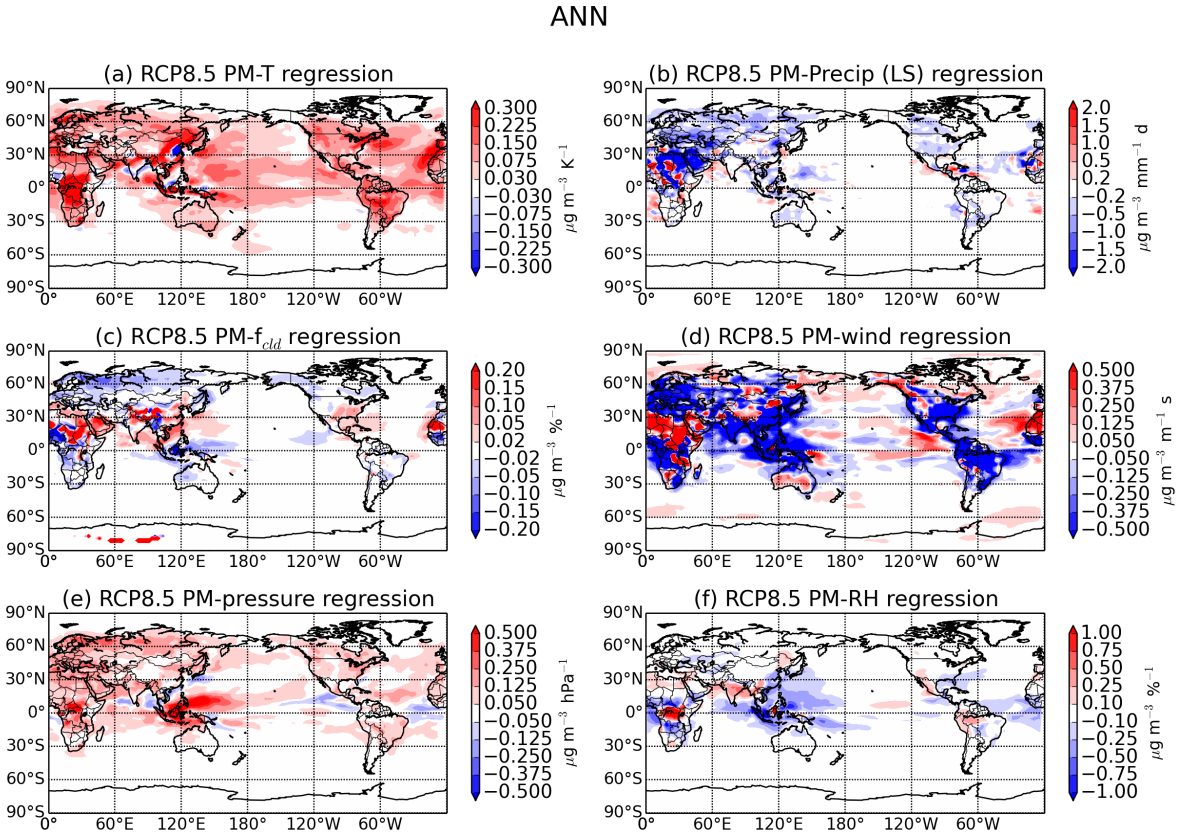

Figure 6: Multiple linear regression coefficients (β_i, "slope coefficients") for the dependence of $PM_{2.5}$ on six meteorological parameters in RCP8.5_2005AER

The effect of relative humidity on $PM_{2.5}$ (Fig. 6f) is small and mostly limited to tropical regions in South Asia. In these regions RH is found to be mostly anti-correlated with dry $PM_{2.5}$, with minor exceptions most notably in India and southwestern North America. Since RH-dependent inorganic aerosol formation is not currently included in this version of the

model, changes in moisture and cloud cover likely explain this negative relationship. Indeed, the relationship between total cloud fraction and $PM_{2.5}$ is found to be negative in this region (Fig. 6c), as well as large-scale anti-correlation across most of Eurasia. In regions with this anti-correlation, clouds can partially inhibit photochemical oxidation of aerosol precursors (such as sulfur dioxide and VOCs) from occurring. Regions where cloudiness and $PM_{2.5}$ are positively correlated, such as India and the southeastern United States, are likely indicative of enhancements in in-cloud production of sulfate aerosols. However, with relatively small slope coefficients in both the PM-f_{cld} and PM-RH regressions, the impact of relative humidity on $PM_{2.5}$ in CM3 is small.

Figure 7 shows the same annually averaged slope coefficients β_i except for RCP2.6_2005AER. There are not many major qualitative differences from RCP8.5_2005AER (Fig. 6) in the RCP2.6_2005AER coefficients across each of the six parameters. Some exceptions include temperature in Fig. 7a, which for RCP2.6_2005AER includes many regions of negative correlation with $PM_{2.5}$. There are also some significant differences in the PM-wind regression (Fig. 7d), especially over North America where sensitivity values are much less negative in RCP2.6 compared to RCP8.5_2005AER. There are several major differences in the trends in meteorological variables in RCP2.6_2005AER and RCP8.5_2005AER; for example, global temperatures in RCP2.6_2005AER are only projected to increase by 2 K at most, compared to 4-5 K in RCP8.5_2005AER. These smaller changes in temperature in RCP2.6_2005AER result in smaller increases in $PM_{2.5}$, which combined with large increases in precipitation in RCP2.6_2005AER, result in an overall decrease in RCP2.6_2005AER $PM_{2.5}$ (Fig. 2). The regression plots for both RCPs are largely consistent. Intuitively, the magnitude or sign of the meteorological perturbation should not fundamentally change the nature of $PM_{2.5}$ sensitivity to meteorology. In other words, whether or not precipitation increases or decreases by a certain amount, $PM_{2.5}$ should respond in a way such that the sensitivities (slope coefficients) are consistent with each other since the underlying mechanisms driving the relationship do not change, i.e. $PM_{2.5}$ concentration decreases for precipitation increases and vice versa.

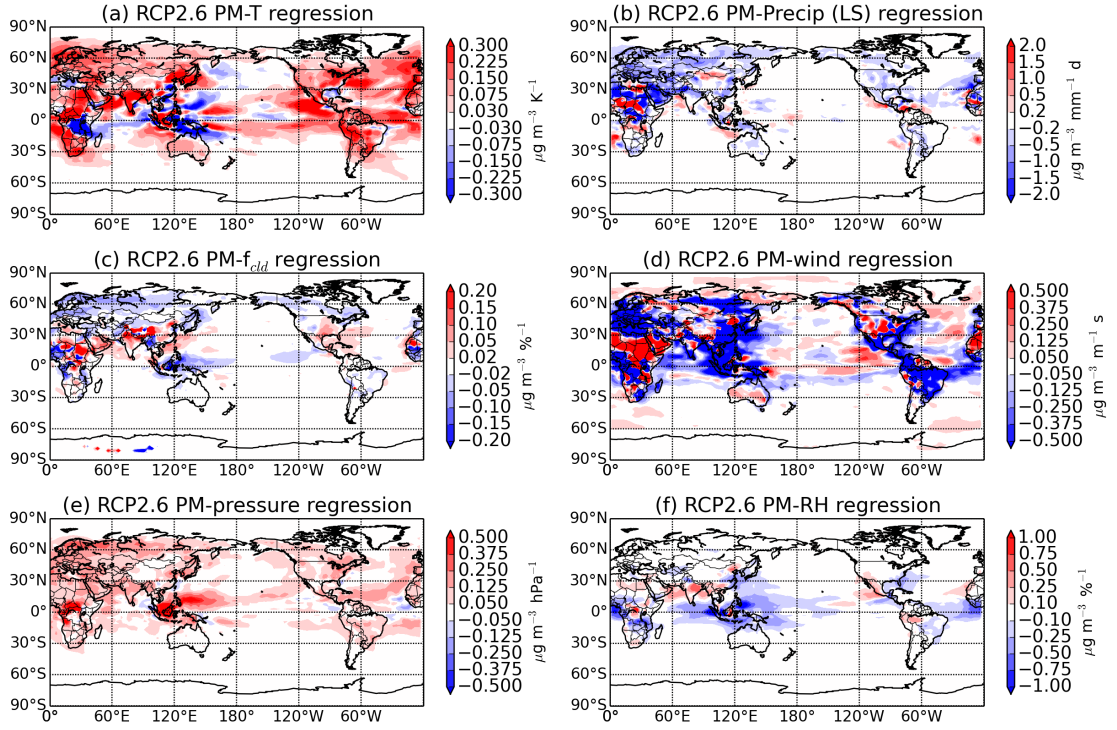

Figure 7: Same as Figure 6 but for RCP2.6_2005AER

4.2 Comparison of modeled $PM_{2.5}$ sensitivities to observations

Though several modeling studies have performed similar regression analyses for $PM_{2.5}$ and have compared results to each other with mixed agreement, none have evaluated their model against regression coefficients derived from observations. Here, we conduct such an analysis. Our goal is to compare monthly $PM_{2.5}$ sensitivities in the model and observations for similar time periods with longer-term trends removed (e.g. from emissions) in order to isolate the impact on meteorological variability on $PM_{2.5}$ concentrations. We do not aim to compare $PM_{2.5}$ sensitivities over time periods influenced by emissions trends. Figure 8 shows the modeled and observed MLR slope coefficients over the United States. Monthly mean modeled slopes from 2006-2015 in the RCP8.5_2005AER simulations are compared to monthly mean slopes from 1998-2008 in the observations. Both observed and modeled monthly values are detrended and deseasonalized by subtracting the 3-month moving average from each original monthly value. These monthly model slopes over the US therefore different from those described in Sect 4.2 in that they represent the short term variability in

early 21st century $PM_{2.5}$ sensitivity rather than longer term climate-driven changes. This conversion is necessary to facilitate consistent comparison with the observations. As can be seen in Figs. 8a and 8b, GFDL CM3 captures the $PM_{2.5}$-T relationship well in the eastern and midwestern US, with large positive slopes across the entire domain. Modeled slope coefficients for temperature are uniformly positive across the Western US, whereas the observations suggest slopes near zero or at times negative, with the exception of southern California and parts of the mountain west. The domain average modeled and observed slopes are 0.09 and 0.07 $\mu g\ m^{-3}\ K^{-1}$, respectively. For precipitation (Figs. 8c and 8d), GFDL CM3 is underestimating the magnitude of the slope coefficients, although capturing the sign (anti-correlation) correctly in the mountain west and northeast in particular. $PM_{2.5}$ in GFDL CM3 is much less sensitive to changes in precipitation than seen in the observations. CM3 shows some spatial agreement in the sign of the slopes in certain locales such as the upper midwest and parts of the eastern US, but overall the model fails to capture the magnitude of the slopes. This is evidenced by the domain mean model slope of -0.06 $\mu g\ m^{-3}\ mm^{-1}$ d versus -0.41 $\mu g\ m^{-3}\ mm^{-1}$ d in the observations. This discrepancy is likely due to weak convective removal of aerosols in this version of CM3, which has recently been investigated by Paulot et al. (2016).

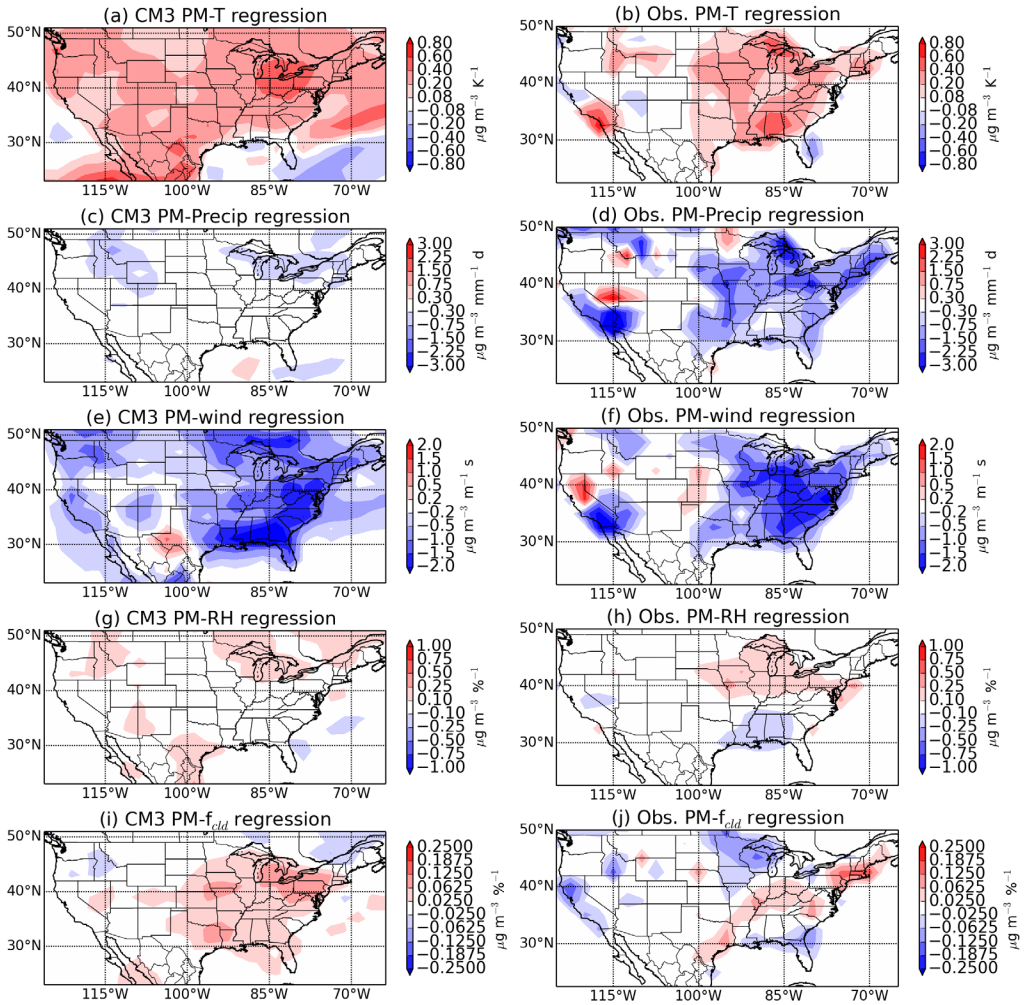

Figure 8: Detrended and deseasonalized monthly multiple linear regression coefficients over the United States for CM3 (left column) and observations from EPA-AQS and NCEP reanalysis (right column).

$PM_{2.5}$-wind sensitivities in both the model and the observations agree well spatially in sign and magnitude primarily in the Eastern US. Model predicted slopes are strongly negative across most of the US, with the largest values occurring in the eastern and midwestern US (-1 to -2 μg m^{-3} m^{-1} s). Observed values of PM-wind slopes are large over California, whereas the model predicts modest negative slopes (-1 μg m^{-3} m^{-1} s) over this region. The domain averages in the model and the observations are in excellent agreement at -0.294 and -0.286 μg m^{-3} m^{-1} s. The $PM_{2.5}$ versus RH in both the model and the observations agree in that most of the $PM_{2.5}$ domain is largely insensitive to RH. However, observed PM-RH regression slopes in the model and observations correlate poorly (domain mean of -0.02 and 0.01 μg m^{-3} $\%^{-1}$ in the model and observations, respectively). Finally, total cloud amount is positively correlated

with $PM_{2.5}$ in most of the eastern US in CM3, but the observations suggest positive slopes only sporadically in the eastern US. Domain mean modeled and observed slopes for $PM_{2.5}$-f_{cld} are both small in magnitude but disagree in sign (0.005 and -0.006 μg m^{-3} $\%^{-1}$). However, CM3 is able to capture an observed negative relationship over the Pacific northwestern US. Overall, the model is able to capture the relationship between $PM_{2.5}$ and temperature and wind speed over the United States with skill, suggesting that the model is useful for projections of the impact of climate change on future air quality at large regional scales. One caveat to this finding is the slightly differing time periods over which the model was run and the observations were reported.

5 Conclusions

Using the GFDL CM3 global chemistry-climate model, we have assessed the impact of climate change on $PM_{2.5}$ concentrations from present-day until the end of the 21st century. We conducted simulations of 21st century climate in which anthropogenic and biomass burning emissions were held fixed at 2005 levels (denoted RCPx.x_2005AER), in order to isolate the effects of changing climate on $PM_{2.5}$ concentrations. We use multiple linear regression modeling to establish associations between changes in meteorological variables and $PM_{2.5}$. Our analysis focuses on multiple spatial domains (global and regional averages) as well as multiple temporal domains (annual and seasonal averages).

We find that climate impacts on global $PM_{2.5}$ are relatively small, ranging from an annual $PM_{2.5}$ decrease of 0.055 μg m^{-3} to an increase of 0.21 μg m^{-3} from present-day to the end of the 21st century ($\Delta PM_{2.5}$), depending on the choice of RCP future climate scenario. Simulations using three of the four RCPx.x_2005AER scenarios project a "climate penalty" for $PM_{2.5}$, meaning future climate will raise $PM_{2.5}$ levels, albeit by small amounts (up to 5%). RCP8.5_2005AER, which projects a future with the most extreme climate impacts, has the largest positive $\Delta PM_{2.5}$ both globally and in most regions, while the stringent climate mitigation scenario of RCP2.6_2005AER results in modest $PM_{2.5}$ decreases due to increases in precipitation and decreases in windspeed that dominate modest temperature changes. In general, the trends in climate-driven $PM_{2.5}$ closely mirror the inherent projected climate of each of the RCPs, as one would expect. Global $\Delta PM_{2.5}$ is found to be largest in the fall months (SON) and at a minimum in the summer (JJA), largely because of sulfate aerosol, which is found to be the most susceptible to climate change. Sulfate is also found to both increase and decrease in concentration depending on the climate scenario, whereas OA and BC aerosol

almost always increase over the course of the 21st century regardless of the scenario due to a lower sensitivity to precipitation than sulfate. Regional averages of $\Delta PM_{2.5}$ are much larger, and specific population centers may see annual average increases of up to 2 μg m^{-3} due to climate changes only. Although seemingly small, epidemiology studies have found up to a 10% increase in mortality for every 10 μg m^{-3} increase in $PM_{2.5}$, suggesting that these regional changes are not trivial (Hoek et al., 2013). Additionally, current $PM_{2.5}$ standards are expected to become more stringent in the future, amplifying the relative contribution of small changes like those that are due to climate if the sensitivities to climate are independent of emissions. Still, global climate-driven changes in $PM_{2.5}$ are found to be minor compared to emissions-driven changes, as changes in PM from 1980-2005 (while emissions were changing) are equal to or sometimes larger than changes from 2005-2100 (while emissions were held fixed), despite occurring over a much shorter time period.

$PM_{2.5}$ is found to be positively correlated with and sensitive to changes in temperature. Temperature increases are likely the major factor explaining $PM_{2.5}$ increases in RCP8.5_2005AER. As a caveat, the GFDL CM3 model lacks certain aerosol-temperature feedbacks that may be negative, such as the effect of increasing temperature on semi-volatile species such as nitrate aerosol and on source of biogenic SOA. $PM_{2.5}$ is inversely related to precipitation, with increasing precipitation rates leading to decreases in $PM_{2.5}$ concentrations. $PM_{2.5}$ is also anti-correlated with wind speed, although projected wind speed decreases over the century will result in less mixing and more stagnation, ultimately yielding higher $PM_{2.5}$ levels. $PM_{2.5}$ is found to be mostly anti-correlated with relative humidity, although this is found to play a minor role in influencing $PM_{2.5}$ concentrations. We find that PM2.5 over the 21st century is most sensitive to temperature, wind speed, and precipitation in that order, although precipitation estimates may be biased low due to weak convective scavenging. We present a model-observation comparison of $PM_{2.5}$ sensitivity to climate variables over the United States where observations of $PM_{2.5}$ and meteorology are available. We find overall some agreement between the model and observations, especially for the PM-temperature and PM-wind sensitivities, which in addition to being dominant on the global scale, also represent the largest sensitivities in the US.

Our model results are consistent with recent findings that climate impacts on $PM_{2.5}$ are relatively minor compared to emissions impacts (West et al., 2013). We are also in agreement with the majority of studies that have found the impact of climate on $PM_{2.5}$ more likely to be a

"penalty" (increase in concentrations) rather than a "benefit" (decrease) (Jacob and Winner, 2009; Fiore et al. 2015). Since we find $PM_{2.5}$ to mostly increase in the future due to climate, air pollution control and climate policies will need to be strong in order to avoid exacerbation of air-pollution related mortality. This is especially true for locations where we find the "climate penalty" to be more severe (1-2 $\mu g\ m^{-3}$). To our knowledge this is the first study to utilize a fully coupled chemistry-climate model, all four RCPs, and full century transient simulations to determine the $PM_{2.5}$ concentrations driven by meteorological changes in a future climate. This work has focused on relatively broad temporal and spatial domains, with the finest resolution being at the regional and seasonal domains. We emphasize that local meteorology may impact local $PM_{2.5}$ much more strongly, as has been shown for winter 2013 in Beijing, China (Wang et al., 2014). Additionally, although these findings are consistent with other models, these results are of a single GCM. Similar experiments with fixed aerosol emissions in order to isolate the "climate penalty" could be conducted perhaps as part of a Model Intercomparison Project (MIP) in order to fully determine the robustness of these results. Future work could focus on incorporating temperature-dependent aerosol volatility, feedbacks on biogenic VOC and other natural emissions, and better treatment of ammonium nitrate thermodynamics and chemistry. However, this is unlikely to change the overall results of this work, such as the finding that $PM_{2.5}$ is much more strongly influenced by emissions than it is climate.

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Table 1: Global average change in population-weighted fine partiulate matter concentration ($\Delta PM_{2.5}$, ng m^{-3}) and temperature (K) between end of century (2091-2100 mean) and beginning of the century (2006-2015) for each RCPx.x_2005AER simulation.

	Sulfate	OA	BC	Dust	Total	ΔT
RCP2.6_2005AER	-57.8	12.6	1.40	-11.6	-55.4	0.8
RCP4.5_2005AER	-19.8	15.6	0.61	6.41	2.88	1.8
RCP6.0_2005AER	17.7	43.1	3.20	19.7	83.9	2.5
RCP8.5_2005AER	104	82.0	4.47	10.7	210	3.9

Table 2: Average change in fine particulate matter ($\Delta PM_{2.5}$, ng m^{-3}) between the end of the century (2091-2100 mean) and present day (2006-2015 mean) for each region (see Fig. 1 for region definitions) for RCP8.5_2005AER

	Sulfate	OA	BC	Dust	Total
North America	148	191	4.61	4.34	387
South America	128	13.8	4.97	116	262
East Asia	245	276	37.8	-159	401
South Asia	132	183	20.5	-37.2	299
Europe	67.7	186	7.34	-68.1	193
Russia	234	282	6.86	-81.0	441
Australia	132	41.5	2.23	-43.5	132
Africa	281	258	21.3	80.1	640
Middle East	159	142	12.1	-295	17.7

Table 3: Same as Table 2 but for RCP2.6_2005AER

	Sulfate	OA	BC	Dust	Total
North America	-177	28.3	0.595	1.05	-147
South America	-18.6	-9.79	0.006	10.9	-17.5
East Asia	-548	57.4	7.42	-27.6	-510
South Asia	-234	68.5	9.61	21.7	-134
Europe	-328	89.5	9.74	8.0	-221
Russia	-82.2	59.7	1.10	-3.3	-24.7
Australia	-44.7	18.7	0.657	107	81.1
Africa	-26.5	164	12.5	31.9	182
Middle East	-163	18.4	0.744	16.9	-126