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Isolating the Meteorological Impact of 21st Century GHG Warming on the Removal and Atmospheric Loading of Anthropogenic Fine Particulate Matter Pollution at Global Scale

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Abstract Particulate matter with the diameter smaller than 2.5 μm (PM2.5) poses health threats to human population. Regardless of efforts to regulate the pollution sources, it is unclear how climate change caused by greenhouse gases (GHGs) would affect PM2.5 levels. Using century-long ensemble simulations with Community Earth System Model 1 (CESM1), we show that, if the anthropogenic emissions would remain at the level in the year 2005, the global surface concentration and atmospheric column burden of sulfate, black carbon, and primary organic carbon would still increase by 5%–10% at the end of 21st century (2090–2100) due to global warming alone. The decrease in the wet removal flux of PM2.5, despite an increase in global precipitation, is the primary cause of the increase in the PM2.5 column burden. Regionally over North America and East Asia, a shift of future precipitation toward more frequent heavy events contributes to weakened wet removal fluxes. Our results suggest climate change impact needs to be accounted for to define the future emission standards necessary to meet air quality standard.

Key Points:
- Future global warming due to GHG can lead to an overall increase of air pollution burden, if the emission level does not change
- The heavy precipitation increase does not effectively remove more aerosol and the decrease in moderate rainfall leads to pollution buildup
- In future, more stringent particulate matter emission control is needed for North America and East Asia to meet today's air quality standard

Supporting Information:
- Supporting Information S1.

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1. Introduction

Climate change in the 21st century, primarily driven by anthropogenic greenhouse gas (GHG) emission, has the potential to negatively impact air quality through multiple mechanisms (Jacob & Winner, 2009; Von Schneidemesser et al., 2015), which thus poses additional threats to human health (Ebi & McGregor, 2008). Research efforts have been made to explore various *meteorological* factors that may contribute to air pollutant changes under global warming condition (Doherty et al., 2013; Kleeman, 2007; Racherla & Adams, 2006; Weaver et al., 2009), which is the main purpose of this paper. These future *meteorological* factors operate in parallel with emission changes in air pollutants that have a direct influence on air pollution level (Avise et al., 2009; Lam et al., 2011; Liao et al., 2006; Pye et al., 2009).

Climate change signals that contribute to increases in surface ozone levels over specific regions such as Northeast United States (U.S.) have been documented (Fiore et al., 2015; Pfister et al., 2014). The effect of climate change on particulate matter (PM), however, remains unclear across observational and modeling studies, with reported changes often conflicting in sign over certain regions (Dawson et al., 2014; Garcia-Menendez et al., 2015; Isaksen et al., 2009; Jacob & Winner, 2009).

Modeling approaches may shed light on the mechanisms of climate change-driven pollutant changes. Previous studies have used chemical transport models driven by prescribed meteorological conditions under global warming conditions (Avise et al., 2009). By perturbing various meteorological parameters, such as temperature, relative humidity, and solar insolation, the sensitivity of pollutant level to climate change can be assessed (Dawson et al., 2007; Kleeman, 2007). However, these types of studies are flawed in that they fail to account for the covariance of any single meteorological variables with the synoptic meteorological conditions (Tai et al., 2010, 2012). Another modeling approach is to use coupled chemistry–climate models (e.g., Doherty et al., 2013), as we have adopted in this study.

This paper particularly focuses on global PM2.5 loading and its wet removal process. Our analysis here differs from previous studies in two aspects. Firstly, we utilize an Earth system global model to conduct transient

climate change simulations throughout the 21st century forced by increasing GHG concentrations along the trajectory of RCP8.5 (Representative Concentration Pathway; Riahi et al., 2011), while keeping the emissions of aerosols and/or their precursors fixed at present-day level (2005). Also, we perform an ensemble of 15 members to minimize the contribution of natural variability in estimating decadal average changes. Previous studies with global chemistry–climate models had at most a small number of time-slice simulations (Allen et al., 2016), or, used simplified tracers in physical climate models (Fang et al., 2013). Secondly, we document the common mechanisms in changes of PM2.5 across three Northern Hemisphere (NH) mid-latitude land regions (North America, Europe, and East Asia), rather than limiting the discussions over a specific region (e.g., Kleeman, 2007; Zhu & Liang, 2012).

We describe the model and experiments in Section 2. The changes in PM2.5 loading and the associated mechanisms are explained in Section 3. Highlights of the results and discussions are in Section 4.

2. Methods

2.1. The Global Chemistry–Climate Model

The Community Earth System Model Version 1 (CESM1) is an Earth system model consisting of the atmosphere, land, ocean, and sea-ice components (Hurrell et al., 2013). The resolution of both atmosphere and ocean models is nominal 1° by 1° for all model experiments, identical to Kay et al. (2015). The 21st-century climate projections with CESM1 are described in detail by Meehl et al. (2013). The anthropogenic forcing in CESM1 can include GHGs as well as time- and space-evolving tropospheric ozone, stratospheric ozone, aerosols, and tropospheric oxidants (Lamarque et al., 2011). The land and the atmosphere are fully interactive in the model simulations, including the biogeochemistry component. But CO_2 concentration is prescribed rather than predicted from emissions in these simulations.

The prognostic modal aerosol scheme in CESM1 includes three internally mixed modes (Ghan et al., 2012) and accounts for numerous processes, such as aerosol nucleation, coagulation, condensational growth, gas-phase and aqueous-phase chemistry, emission, dry deposition and gravitational settling, in-cloud and below-cloud scavenging (Liu et al., 2012). The Aitken mode includes sulfate and secondary organic matter; and the accumulation model includes sulfate, primary and secondary organic matter, black carbon. Nitrate is not simulated in this version of the model. Each mode includes both interstitial and cloud-borne component of each chemical species. The performance of aerosol simulation has been extensively evaluated (Ghan et al., 2012; Liu et al., 2012), and we have only briefly discussed the notable biases in the supplement section (Figure S1 and S2).

Aerosols in the coarse mode (mainly natural aerosols such as soil dust and sea salts) are included in the model simulation, but all results presented here do not account for coarse aerosols because they are deemed less important for human health impact (Pope & Dockery, 2006). Instead, we only consider PM2.5 component (PM with size smaller than 2.5 μm) in the Aitken and accumulation modes: sulfate, black carbon, primary organics, and secondary organics. We particularly focus on sulfate aerosols for detailed analysis, which contribute to more than 50% of the total PM2.5 amount in our simulations.

2.2. Model Experiments

The 21st-century simulations start from the end of the year 2005 and run through 2100. The experiments consist of 15 ensemble members, differing solely in their atmospheric initial conditions by a temperature difference at random round-off level (order of 10^{-14}°C), in order to capture internal climate variability at shorter time scales (Kay et al., 2015).

The simulations are forced by RCP8.5 scenario (Riahi et al., 2011), except that all emission of sulfate, BC, primary organic matter (POM), and SOM aerosols (or their precursors) and atmospheric oxidants are fixed at the year of 2005 level (RCP8.5_FixAerosol2005). Note that in RCP scenarios database only the decadal change of aerosol emission is considered, so the year 2005 emission level here represents the average level during 2000–2010, and should not be interpreted as the emission at a single year (Lamarque et al., 2011).

The differences in the mean climate (temperature and precipitation) between late and early 21st century in the RCP8.5_FixAerosol2005 experiment are largely due to the increase in GHG concentrations, and not due to declining aerosol emissions as originally specified in RCP8.5. Therefore, changes of PM2.5 level in our

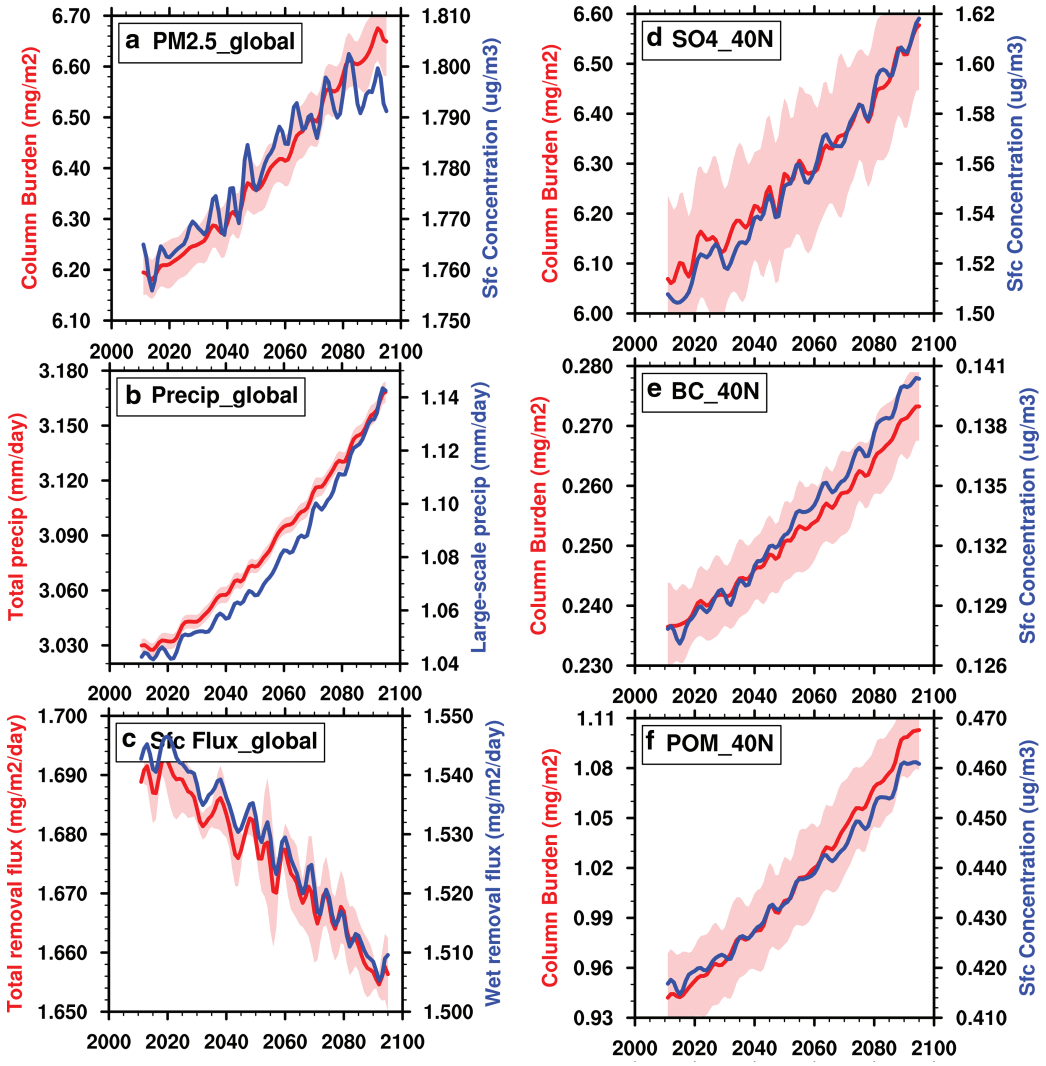

Figure 1. Results of RCP8.5_FixAerosol2005 model experiments. (a) Total PM2.5 (including sulfate, black carbon, primary organic matter, and secondary organic aerosols in the two fine modes) future changes in the surface concentration ($\mu g/m^3$, blue) and column burden (mg/m^2, red). Ensemble variance (1 sigma) for column burden is shown in red shadings. (b) Global mean total precipitation (mm/day, red) and the component of large-scale precipitation (mm/day, blue). (c) Global mean total removal flux of SO_4 (mg/m^2/day, red) and the component of wet removal flux (mg/m^2/day, blue). Out of the total wet scavenging flux, about 40%–50% is from below-cloud scavenging and the rest is from the in-cloud removal and the ratio does not change much in the 21st-century simulation. (d, e, f) Same as (a) but for SO_4, BC, and POM average in the 30°–50°N band centered around 40°N. The SO_4 global change and regional changes in three NH mid-latitude regions are shown in Figure 2.

21st-century simulations are only a result of GHG-induced climate change, rather than changes in aerosol emission.

3. Results

3.1. Changes of PM2.5 Under Global Warming Conditions

Figure 1a shows that the ensemble-mean PM2.5 loadings simulated by the model, in terms of column burden (mg/m^2, red lines) and surface concentration ($\mu g/m^3$, blue lines), are increasing throughout the 21st century, despite the emissions being constant throughout the experiment. The global increase in total PM2.5 is 8% for the column burden and 3% for the surface concentration that has a more direct effect on human health. Note that the total PM2.5 changes are due to sulfate (SO_4) (accounting for approximately 50% of the total PM2.5 mass), black carbon (BC), POM, and to a lesser extent secondary organic aerosols.

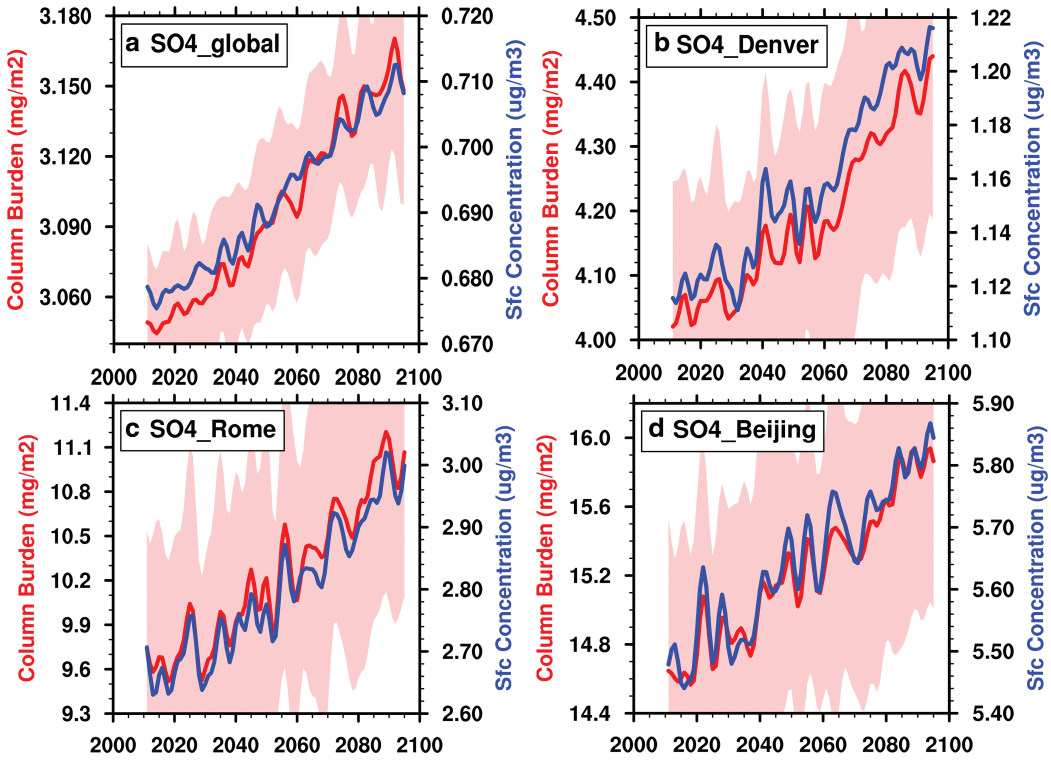

Figure 2. Similar to Figure 1d, but for SO_4 averaged globally and within the three representative boxes ("Denver", "Rome," and "Beijing").

The increases over the NH mid-latitude regions (30°–50°N) are stronger for all major PM2.5 species (BC: 11%–19%, POM: 11%–15%, and SO_4: 8%–10%) (Right column in Figure 1). Robust responses of the atmospheric burden over 30°–50°N can be expected because these regions have the largest present-day pollution levels (except that POM has its peak level over the tropics). The small ensemble variances (red shadings in Figure 1) indicate that the rising trend in the global mean loading of PM2.5 is robust across all ensemble members. This is, in particular, true for BC and POM (Figure 1), while SO_4 trends have larger ensemble variances than BC and POM.

Regarding the spatial distribution changes, Figure 3a shows the ensemble mean present-day (2005–2015) SO_4 column burden and Figure 3b shows the changes at the end of the 21st century (2090–2100 minus 2005–2015). The increase in the total column burden is significant over almost the entire NH land regions, with the largest increases by 16% found in the European, Mediterranean, and North African regions. Other regions with high PM background levels such as Eastern North America and East Asia also have substantial increases by 10% and 8%, respectively. The distribution of SO_4 surface concentration is similar to the total column burden but is more confined to local source regions (e.g., hotspots over China, Figure 4a). The increase in surface concentration over the entire North America and East Asia is not significant considering the large year-to-year variance and contrasting signs in the change. But significant increases are found over the Western U.S. and Western China by 9% and 11%, respectively (Figure 4b). Some parts of the regions with enhanced total column burden (e.g., Southeast U.S.) experience a slight reduction in surface concentration by about 5% (Figure 4b).

Over smaller geographical domains, such as the 5° by 5° boxes centered around the cities of Denver, Rome, and Beijing, ensemble variations of the increasing trends become much larger (red shadings in right columns of Figure 2). The larger variances versus trends are expected due to the natural variability of the climate system, which plays a significant role in determining regional climate changes (Deser et al., 2012), and consequently impacts to PM (Manders et al., 2012). Using a relatively large ensemble modeling approach, we can suppress a significant fraction of the contribution of natural variability to the estimates of decadal changes. Note that to derive the changes in Figure 3b we are effectively using 150 years of model

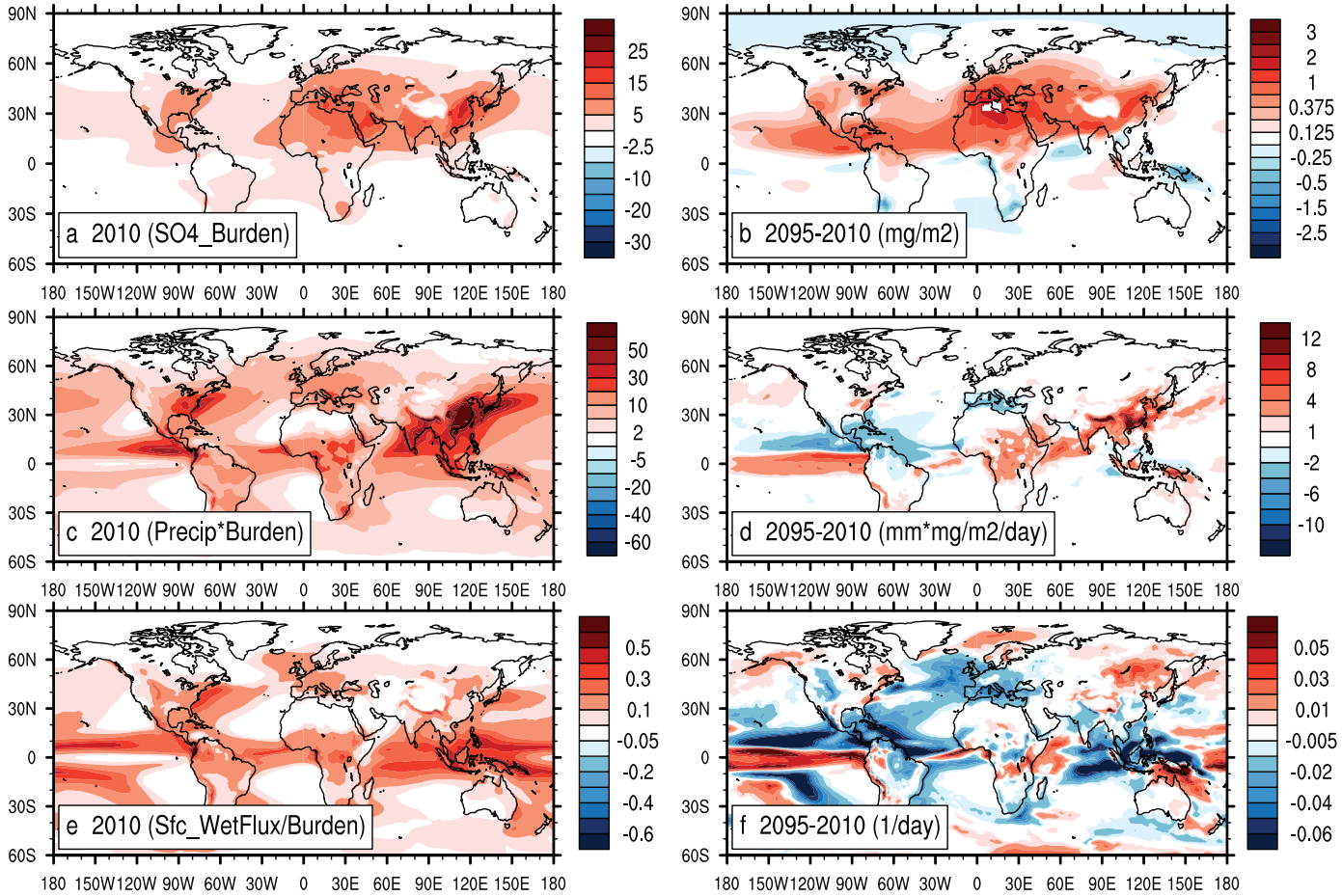

Figure 3. (a,b) SO_4 atmospheric column burden (mg/m^2) during the 10-year period centered at 2010 (2005–2015), and its changes over the 10-year period centered at 2095 (2090–2100). Surface concentration is shown in Figure 4 (a,b). (c,d) Derived wet removal flux as the product of precipitation and SO_4 column burden. (e,f) Wet removal flux divided by SO_4 column burden, as an indication of wet removal *efficiency* (unit: 1/day).

output, as opposed to the 5 or 10 years used in the previous analysis (e.g., Allen et al., 2016; Dawson et al., 2009).

3.2. Trends in Wet Removal Flux and Precipitation

At steady state, atmospheric burden of PM2.5 at the global scale can be simply expressed as the product of emission *times* lifetime. The chemical lifetime of simple tracers can be explicitly set to a certain value (e.g., Fang et al., 2011). However, in this coupled Earth system model, the lifetime of SO_4 is variable and approximately 3.7 days (Liu et al., 2012), determined by the overall removal efficiency. Since the production terms of PM2.5 in the model are fixed to their 2005 levels (with fixed emissions and oxidants and only a small dependence on temperature slightly affecting the production term), the increase in the atmospheric burden of PM (Figure 1a) can only be expected if the lifetime increases. Indeed, the PM lifetime is prolonged over much of the globe (calculated as the 1/lifetime, or loss frequency, in Figure 3f with the blue color meaning longer lifetime). A notable increase in local lifetime relevant to our discussion here is found over the Western U.S. (0.3 days increase from the 3.1 days at present) and Europe (1.2 days increase from the 4.9 days at present).

One may ask: why does the lifetime of pollutants increase? We argue that it must be due to a temporary reduction of sink term (removal flux in the unit of kg/s). We note that the sink (i.e., total removal flux) decreases by 2%–4% (Figure 1c), which is largely contributed by the wet removal (blue line). The reduction of removal flux is not as strong as the increase in column burden (about 8% for total PM2.5 in Figure 1a and 4% for SO_4) because the increase in PM will enhance deposition, competing with the meteorology induced changes. An exact partitioning of the two opposing processes (smaller deposition rate due to

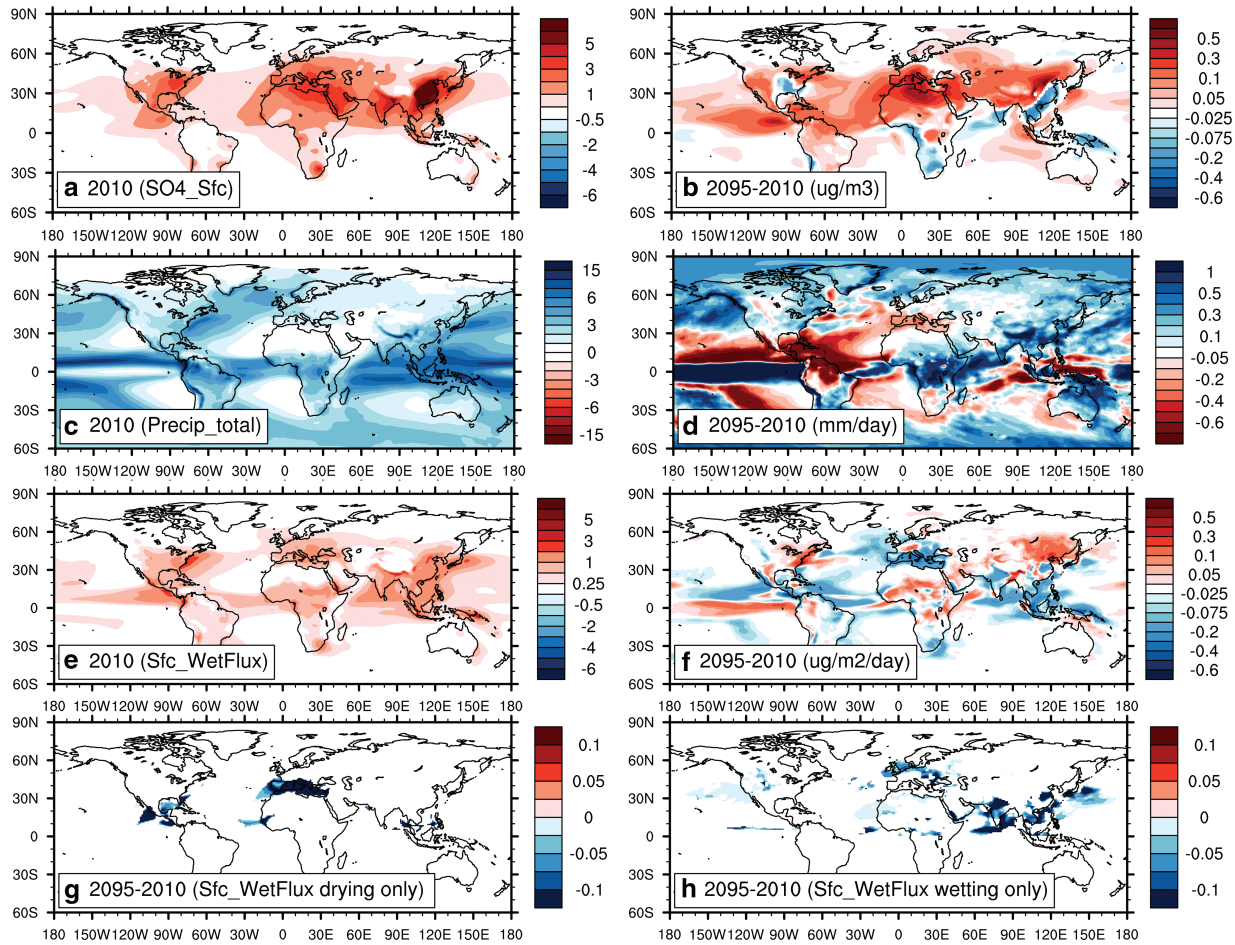

Figure 4. Similar to Figure 3, but for (a) SO_4 surface concentration, (c) total precipitation, (e) surface wet removal flux of SO_4, and their end-of-century changes in (b, d, f). (g) Change of SO_4 surface wet removal flux as in (f), but only showing the grid cells with reduced wet removal flux and decreasing precipitation ("drying"). These areas are predominately located in Mediterranean regions, as represented by the "Rome" box analysis in Figures 5 and 6. Panel (h) is same as (g), but only showing the grid cells with reduced wet removal flux and increasing precipitation ("wetting") or with insignificant changes. These areas are widely spread in tropics and mid-latitudes, as represented in the "Denver" and "Beijing" box analysis in Figures 5 and 6.

meteorological changes in a warmer climate versus larger deposition due to enhanced background pollution) is difficult in this dynamic model with coupled chemistry and meteorology, unless we conduct specially designed experiments such as fixing the certain quantity of metrological field. The decrease in removal flux also appears to be model dependent according to the three-model assessment by Allen et al. (2016).

What is interesting here is why the reduction in removal flux, especially the wet deposition part, runs counterintuitive to the fact that both large-scale and total rainfall is increasing under global warming by 3%–7% (Figure 1b). Note that the rainfall increase associated with warming is a robust feature across different climate models (Allen & Ingram, 2002). The opposing trends of global precipitation and wet deposition flux could be simply related to the mismatch in the location of precipitation change and background pollution levels (Allen et al., 2016). In our model simulations, the rainfall increases occur over large areas in the tropical oceans and Southern Hemisphere mid-latitudes (Figure 4d), where the climatological aerosol pollution loading is small. Therefore, wet removal flux does not change much in those regions (Figure 4f, insignificant changes are masked white). We also note that the seasonality of rainfall changes is unlikely to contribute to the inconsistency because maximum rainfall changes in the NH mid-latitude regions are found in boreal summer (June, July, and August), simultaneously with large aerosol background loading.

The model output of wet removal flux archived in the century-long simulations might not always be available from other chemistry–climate model simulations. A simple diagnostic to substitute the model generated wet removal flux is the product of monthly precipitation and monthly atmospheric column burden

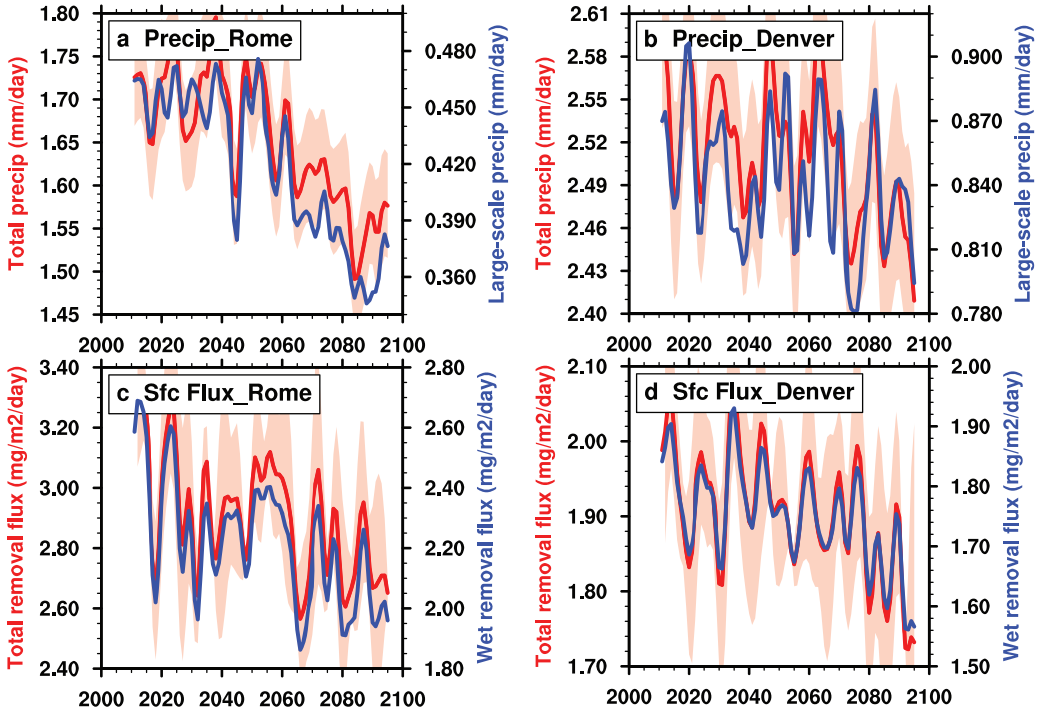

Figure 5. Total precipitation (red) and large-scale precipitation (blue) in the "Rome" box (a) and the "Denver" box (b). Total removal flux of SO_4 (red) and wet removal flux of SO_4 (blue) at the surface in the "Rome" box (c) and the "Denver" box (d). The results for the "Beijing" box are similar to the "Denver" box (not shown).

for each grid point (Figure 3c), which gives qualitatively the same results as the changes of wet removal flux (Figure 4). This metric, however, may over-predict the changes over South Asia and East Asia, where the background pollution levels are high.

Alternatively, we also plot the wet removal *efficiency* (local wet removal flux/local column PM2.5 burden) in the bottom panels of Figure 3. This is equivalent to the inverse of the local PM2.5 lifetime due to wet deposition only (unit: 1/day). The maximum values of wet removal efficiency (Figure 3e) are in the intertropical converge zone and storm track regions in NH mid-latitudes. We find that the wet removal efficiency becomes significantly smaller at the end of 21st century over land regions such as Europe, North America, and to a lesser extent, some parts of East Asia, which explains much of increase in the PM column burden over these regions. Over the ocean, the strong reduction of removal flux over North Atlantic and northern tropical Pacific also correspond to the increase in SO_4 column burden.

The "Rome" box (representing Southern Europe and Mediterranean regions) appears to have the largest reduction in removal flux and a robust increase in column burden as well as surface concentration. Figure 5a shows that the mean precipitation over the Mediterranean region decreases by 15%, leading to the decrease in removal flux by 25%. This drying trend directly leads to a corresponding increase in column burden (Figure 3b). We also note that the overall decrease in rainfall over subtropical land regions including the "Rome" regions turns out to be a robust feature of the CMIP5 models (IPCC, 2014). The declining precipitation trends that directly contribute to the wet removal flux reduction are also found in the entire Mediterranean region and Central America regions (Figure 4g).

Surprisingly, over other mid-latitude regions where the surface wet removal flux decreases, the total precipitation amount increases or has an insignificant trend (Figure 4h). These regions are represented here by the other two boxes "Beijing" and "Denver." For example, the "Denver" box (Figure 5, right columns) shows that the declining trend of precipitation (about 5% across the century) is insignificant considering the variability shown in the red shade. This is not sufficient to explain the large reduction in surface removal flux (about 20%) that leads to the PM2.5 concentration increase. The key message here is that metrics beyond

the mean precipitation amount, such as the rainfall intensity spectrum, may be crucial in governing local wet removal flux. We will examine this aspect in the next section.

Note that these boxes (about 500-km by 500-km in size) are selected to represent regions in the heavily polluted NH mid-latitudes (such as central United States, Mediterranean, and eastern China), rather than any single observational sites/points. For each representative region, the aerosol mass transport moving into and out of respective regions are not significantly different between the two climate states (present-day and end-of-century) (see Figure S3). Therefore, the change in the removal flux is the dominant factor explaining the loading change for each region.

3.3. Precipitation Intensity Changes

We next examine the rainfall changes as a function of intensity for two representative regions ("Rome" and "Beijing"). As we discussed earlier, the "Rome" region features robust mean precipitation reduction, while the declining trend of precipitation over "Beijing" or "Denver" is insignificant (Figure 5b).

Figures 6a and 6b show the probability density function of precipitation as a function of daily precipitation intensity (mm/day in logarithmic scale, blue) and its changes by the end of the century (red). Despite the slight increase in probability at very heavy rainfall days (larger than 10 mm/day), for the bulk of intensity spectrum, the occurrences of precipitation over the "Rome" box (Figure 6a) decrease, contributing to the total rainfall reduction (Figure 5a). The reduction in the mean rainfall can also be seen from the increased occurrences of low rainfall days (smaller than 0.1 mm/day). On the other hand, over the "Beijing" box (Figure 6b), the rainfall frequency decreases in those days with small to moderate rain rate (less than 1 mm/day), while it also *increases* at those larger rainfall days (more than 1 mm/day), in particular very heavy rainfall days (larger than 10 mm/day).

The precipitation intensity changes can also be depicted in a different way. We first segregate all rainfall daily data into 100 percentile bins following the present-day rainfall distribution, so that for the 365 daily rainfall data points in any given year, approximately every bin contains 3 days, with the 99%–100% bin consisting of the heaviest 3 days of rainfall. We then calculate how the number of days falling into each bin would change by the end of the century (Figures 6c and 6d). For the "Rome" box, by the end of the century, more days are falling into the small rainfall bins as measured by present-day distribution (10%–40% percentiles), while fewer days are considered as the moderate to high rainfall days (60%–100% percentiles), consistent with the overall drying trend. For the "Beijing" box, we again show that the number of days with small to moderate rainfall (10%–50% percentiles) decreases as the climate warms, which is then compensated by the increase in those heavy rainfall days (85%–100% percentiles). The shift of precipitation into the heavy end of rainfall intensity spectrum is also similarly found for the "Denver" box, despite a weak total precipitation decrease.

Although the increase in heavy rainfall events can balance the decrease in moderate rainfall events, thus leading to an insignificant trend of total precipitation, more heavy precipitation days appears to be insufficient to further enhance wet removal flux. This is readily shown in Figure 6f that heavy precipitation in certain days (10–50 mm/day) does not necessarily lead to larger surface removal flux. In fact, for days with very heavy rainfall intensity (>50 mm/day), there is even a weak anticorrelation (correlation coefficient amounting at −0.3 to −0.5) between rainfall intensity and surface removal flux (Figures 6e and 6f). Observational studies have provided ample evidence for this. For example, Seymour and Stout (1983) conducted the sequential sampling of rainwater during single rain events and found the concentrations of ions decreased rapidly after first few mm of rainfall. Similar results are obtained in other studies analyzing the relationship between raindrop size and chemical concentration (Wai et al., 2005 and references within). On the contrary, a reduction in small-to-moderate rainfall days (Fang et al., 2011) or frequency of rainfall events (Mahowald et al., 2011) leads to a decrease in wet removal flux.

We have mainly presented the analysis over the "Beijing" box simulations, but the feature that the rainfall change shifted toward heavy rain rate in a warmer world has been documented in observational records (Fischer & Knutti, 2016; Min et al., 2011) and other model simulations (e.g., figure 19 of Peacock, 2012 for the United States, and Kooperman et al., 2016 in a superparameterized global climate model). The shift of intensity rainfall is backed by robust physical understanding, which relates to the fact that total rainfall increase always falls behind the water vapor increase in the atmosphere (e.g., Lu & Cai, 2009) due to energy

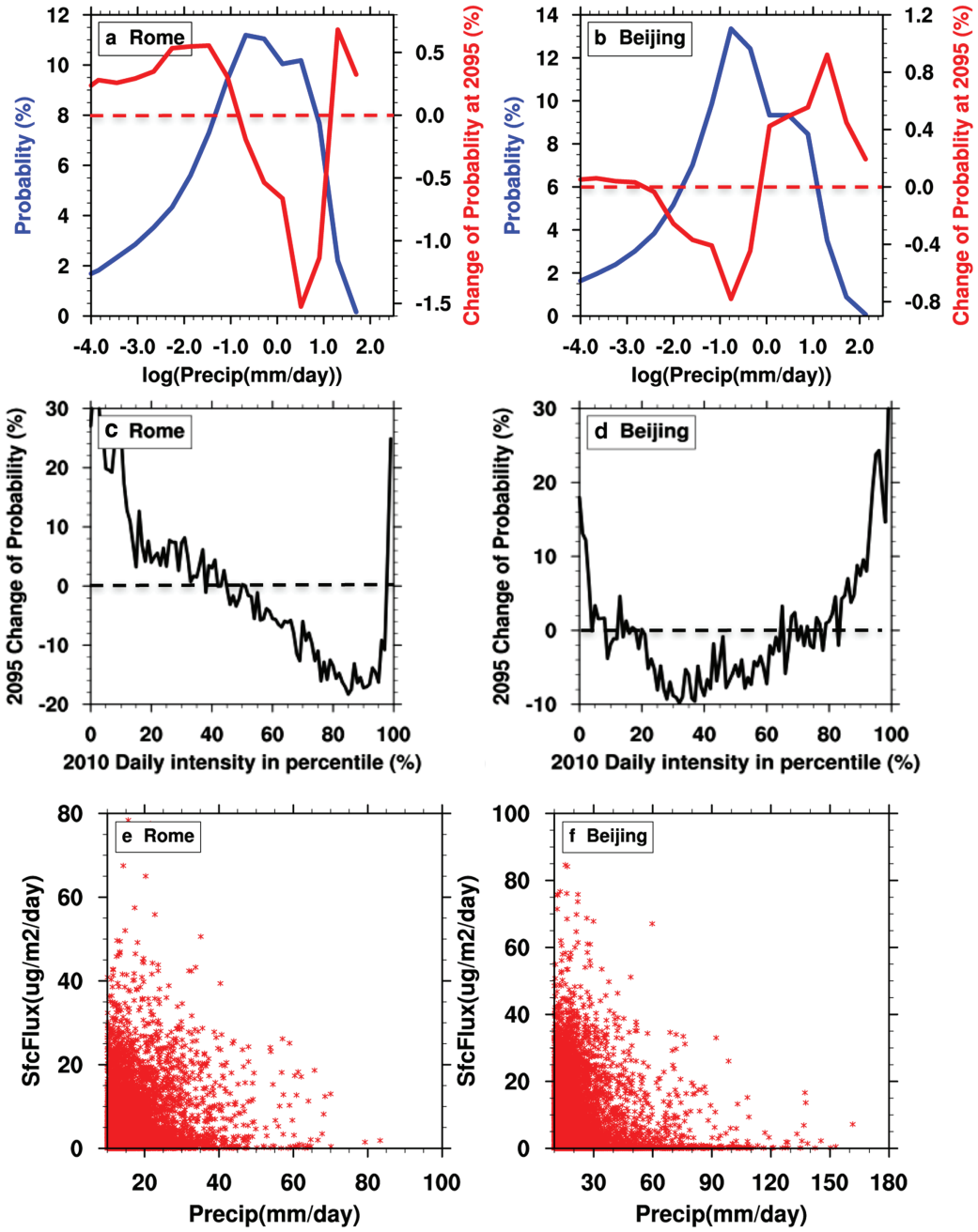

Figure 6. (a,b): Probability density function (%) of rainfall (log (mm/day)) at present-day (2005–2015, blue lines) and its changes at the end of the century (2090-2100, red line) in the "Rome" (a) and "Beijing" (b) boxes. (c,d): Change in precipitation occurrence (%) at 2090–2100 relative to 2005–2015, as a function of present-day precipitation intensity measured in percentile bins (%) in the "Rome" (c) and "Beijing" (d) boxes. The results for the "Denver" box are similar to the "Beijing" box (not shown). (e) The scatter plot of the daily precipitation intensity (mm/day) and daily SO_4 surface wet removal flux over all grid points in the "Rome" box, during the heavy rainfall days (>10 mm/day) in the 2006–2010 model simulation. (f) Same as (e) but for the "Beijing" box.

budget constraints, while water vapor increase scales more closely with extreme precipitation (Pendergrass & Hartmann, 2014). That is because, the extreme precipitation, which occurs over a limited time frame, is less subject to energy budget constraints. The end result is that extreme rainfall increases faster than mean rainfall, which represents a shift into heavy rainfall events in intensity spectrum (Pendergrass et al., 2015; Trenberth, 2011). The impact of the precipitation shift toward heavy rainfall events has been related to flooding hazards, and what we have demonstrated here is the chemical consequence of this physical feature

of climate change. The novel implication of our study is that the precipitation intensity spectrum and the parameterizations of wet removal process are key aspects worth close examination in chemistry–climate models.

In summary, the controlling factor for the monthly mean pollution levels in this Earth system model is that the wet removal flux decrease is associated with the rainfall change, not only in terms of total amount but more importantly the shift in rainfall intensity spectrum over some regions. The moderate rainfall occurs much more often than heavy rainfall, so a reduction in moderate rainfall is more important for wet removal changes than an increase in much fewer heavy precipitation events. The shift in the balance of PM2.5 source and sink terms due to smaller removal flux consequently leads to the increase in the PM2.5 column burden. Our results indicate that to meet the same air quality standard as of today, regional air pollution emission regulations might need to be further strengthened.

4. Discussions and Concluding Remarks

The 21st-century global warming, primarily driven by GHG increases, has many consequences for the Earth system. The research question we aim to address here is how GHG global warming alone would affect the atmospheric lifetime and the distribution of PM2.5 pollution. We find that PM2.5 pollution increases under the global warming conditions even with fixed present-day emissions, thus posing greater health threats. The decrease in wet removal flux, although not large (1%–2% during the 21st century), is sufficient to lead to the PM2.5 column burden increase (see figure 1 and table S2 of Allen et al., 2016). The imbalance of PM2.5 production and removal, due to a smaller wet removal flux, is likely to be restored in a hazier world beyond 2100, with the sink term increasing to match with the emission flux (i.e., the source term) again.

This paper complements earlier studies (e.g., Allen et al., 2016; Fang et al., 2011) and provides valuable results in that it applies a different model, a different simulation set-up and a longer simulation time, and examines such impact down to regional scale. This study also utilizes model output from a larger (single-model) ensemble (15) than previous studies (e.g., Allen et al., 2016 had a maximum of three ensemble members for each model). The general features (e.g., reduction of wet removal flux in the global average and zonal mean) are very similar to previous studies, in particular, those models examined by Allen et al. (2016). At the regional scale (e.g., East Asia), there seems to be more discrepancies regarding the magnitude and even the sign of the pollution loading changes. These discrepancies are also obviously seen in the three models analyzed by Allen et al. (2016). Model parameterization of wet scavenging is likely a major source of uncertainty. But we also note that the results reported here are based on 15 runs (i.e., 15×10 years for each mean climate state), and is likely to be different from the results obtained from a smaller sample size (e.g., 30 years) due to the precipitation variability in the model.

In this study, we purposely did not aim to examine air pollution change due to anthropogenic emission changes (e.g., Hogrefe et al., 2004; Lam et al., 2011; Pye et al., 2009). Aerosol emissions in many source regions have gone down markedly since 2005 already and are expected to go down further in the future (RCP8.5; Lamarque et al., 2011. Aerosol pollution level (surface concentration or column burden) is, to the first degree, governed by the emission intensity. For example, figure 3d of Xu et al. (2015) shows the aerosol reduction in 2060–2080 following the emission decline in RCP8.5 simulations. Also the meteorology conditions, as examined in this study, only play a secondary role. Therefore, the results presented using a fixed emission simulation here should not be viewed as a forecast of future PM levels and the 2005–2015 increasing trends simulated here (Figure 1) is already inconsistent with the observed declining trend of SO_4 over the United States. To provide a full assessment of future PM levels, certain future emission scenarios (most likely declining as in RCP database) should be used as input to chemistry–climate models. Yet, the goal of this study based on fixed emission simulation is to isolate the role of future climate in governing the pollution level and should not be misinterpreted as a realistic projection of future PM.

We comment on a few limitations of the current work that may point to future research directions.

1. Changes in natural aerosols such as soil dust (Pu & Ginoux, 2016; Yuan et al., 2016) and sea salt (Gettelman et al., 2016) are not included in the analysis, as they contribute little to the mass in fine PM modes (Aitken and accumulation) in this model. Also, it is difficult to separate the climate effect on transport and removal, because the emission of dust and sea salt is online predicted based on

meteorological field and is not easily fixed as we have done to the four chemical species here. There are also reports that biomass burning emission (Yue et al., 2013) and biogenic emissions from vegetation (Wiedinmyer et al., 2006) could change under a warmer climate. This effect is also intentionally omitted as we have fixed these emission sources along with other anthrophonic sources for all species considered here.

2. Ozone change was not considered in the model here (Weaver et al., 2009), because ozone concentration is prescribed from a set of offline CAM-Chem simulations (Lamarque et al., 2011), rather than predicted from more complex online CAM-Chem simulations (Tilmes et al., 2015). Utilizing the full chemistry package rather than the simple chemistry and three-mode aerosol model as used here, would be much more computationally expensive, thus limiting the approach of large ensemble and century-long transient simulations.
3. One may also wonder whether the PM production (i.e., source terms of pollution species) contributes to changes in atmospheric loading. In the real atmosphere, the formation of sulfate passes through a complicated oxidation process, which might depend on many climatic conditions (e.g., temperature, solar insolation, and humidity for aqua-phase reactions). Previous models have reported that increased sulfate concentrations can result from an increase in hydrogen peroxide that leads to enhanced in-cloud production (Liao et al., 2006; Racherla & Adams, 2006). Other studies such as Rae et al. (2007), however, suggested that contribution from this temperature-dependent production is small. Such a production mechanism impacting future changes is intentionally eliminated in our simulations, as we have kept the atmospheric oxidant level as a constant. The purpose of fixing both aerosols/precursors and oxidants is to isolate the meteorology impact on transport and removal terms of PM.
4. This paper aims to isolate climate impact on PM, but the resultant PM2.5 level change will further perturb atmospheric energy budget and climate (through direct radiative effect or interactions with clouds), thus creating a complicated feedback loop. To the first order, the precipitation changes here can be seen as a response to GHG warming because of the larger radiative forcing. Note that GHGs cause more than 3°C warming at 2100 in this simulation, and the aerosol reduction by more than 60% (due to emission cut) can cause a global warming of about 0.5°C only (Xu et al., 2015). While in the present study, the aerosols increase is at the level of 5%–15%, which leads to a negligible radiative forcing compared to the GHG increase (Lin et al., 2016). Regionally, aerosol impact on cloud and precipitation may play a competing role with GHGs. For example, it has been shown that NH aerosols can change the tropical circulation (Rotstayn et al., 2014; Xu & Xie, 2015) to affect the heat extreme frequency (Xu et al., 2015). But this climate–aerosol feedback is difficult to separate in current experiment configuration. To cleanly separate the effect and cut off the feedback loop, one can perform a simulation in which the aerosol hygroscopicity is set to zero (as water-insoluble tracer).
5. Future research with multiple models to account for diversity especially in precipitation field (Xie et al., 2015) is needed. Based on the analysis in Allen et al. (2016) on three models, CESM1 appears to provide a middle-range estimate of climate change impact on air pollutions. On the other hand, although the decadal climatic trend on air pollution is hard to quantify in the observations due to the simultaneous changes of pollution sources, observational records may still provide useful constraints on the relationship between air pollution and climate variability at seasonal (monsoon) to interannual time scales (El Niño–Southern Oscillation).

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