Response to comments of referee 2 on "Strong influence of 2000-2050 climate change on particulate matter in the United States: Results from a new statistical model"

We thank the referees for their careful reading of the manuscript and the valuable comments. This document is organized as follows: the Referee's comments are in *italic*, our responses are in plain text, and all the revisions in the manuscript are shown in blue. **Boldface blue** text denotes text written in direct response to the Referee's comments. The line numbers in this document refer to the updated manuscript.

Reviewer 2

I believe the study presents several analyses investigating projections of climate change impacts on $PM_{2.5}$ pollution that provide valuable insights to the air quality modeling community. The manuscript is well-written and clear. I appreciate the authors' effort to undertake a study that includes several layers of research: developing and describing a new statistical regression model, applying the model to the projections of a multi-model GCM ensemble, using these results to guide an investigation into $PM_{2.5}$ projections from CCMs, and using a CTM to identify factors contributing to the inconsistencies in simulations of $PM_{2.5}$ impacts. As it stands, the study presents several useful findings that make it worthy of publication. However, by addressing different research needs and different modeling approaches (including a statistical regression model, 17 different GCMs, 4 different CCMs, and 1 CTM), the analysis of results for each research question being investigated is at times limited. I encourage the authors' to go deeper in their discussion. I would also persuade the authors to further investigate the major findings of their work individually in follow-up research. Some specific comments are included below.

Response: We thank the reviewer for raising so many good points. This feedback has significantly improved the manuscript. In particular, we have tried to deepen the discussion section in response to this reviewer's concerns.

We also to draw the reviewer's attention to the fact that we have increased the number of CMIP5 models used in this study from 17 to 19. All the results in the new manuscript will be based on the results from this ensemble of 19 models. For a full list of these models, please see Table S1. We have also replaced mass of organic carbon (OC) with the inferred mass of organic aerosol (OA) in Figures 6 and S13. OC is the measured carbon component of OA.

- I felt there is some disconnection between different aspects of the study as it moves from the regression model to GEOS-Chem. The study could be broken down into separate analyses: (1) a PM2.5/meteorology linear regression model; (2) projection of PM2.5 climate impact from the CMIP5 GCM ensemble; (3) PM2.5/temperature relation in 4 ACCMIP CCMs; (4) GEOS-Chem sensitivity of PM2.5 to temperature. The connection between (1) and (2) is evident, while the connection between subsequent sections is not as clear. In moving from sections 4 to 5, the manuscript goes from statistical inference of PM2.5 changes from 20-yr present/midcentury simulations with 17 GCMs, to atmospheric chemistry simulations from 4 CCMs covering a different 15- yr present period and conditions, to a CTM simulations for a different 9 yr period. Is there truly a clear connection between these different types of models and the nature of these simulations, other than saying that the temp-PM25 relationship is important? The scope of the study limits the depth with which each finding is examined.

Response: The reviewer raises valid concerns. In an effort to make the connections between sections more clear, we have rewritten much of the first paragraph of Section 5.

Page 11, line 29-31. A key question is why previous model studies show no consistent sign in the in the change of future $PM_{2.5}$ relative to the present (Jacob and Winner, 2009). Such discrepancies among models no doubt arise in part because of differences in model projections of

future climate or in model speciation of $PM_{2.5}$. In this section, we investigate whether differences in model representation of the sensitivity of $PM_{2.5}$ to meteorological variability may also contribute to uncertainty in projections of future $PM_{2.5}$.

Page 12, line 6-8. This section consists of two parts. First, we test the capability of four ACCMIP models and GEOS-Chem in capturing the observed relationship between JJA monthly mean $PM_{2.5}$ and temperature. We find no model simulates this relationship well. Second, using GEOS-Chem as a testbed, we investigate the reasons of this failure in this particular model.

- One topic I would encourage the authors to discuss further in their manuscript is the impacts of 2050 climate derived from the CMIP5 ensemble and the regression model. Only ensemble-mean results are presented. (a) I would be very interested in seeing the differences in the projections from individual GCMs. (b) Additionally, I would like to see the interannual variability across individual years in the 20-yr penalty estimates. How robust is the study's finding of an anthropogenic-induced climate change impact by 2050 under a stabilization scenario, given that natural variability has been shown to significantly influence U.S. temperature and precipitation projections on timescales as long as 50 years (Deser et al., 2014; doi: doi:10.1175/JCLI-D-13-00451.1.)? There is a great opportunity to explore climate-related uncertainty in PM2.5 projections at much greater depth within this data.

Response: These are two excellent questions.
In response, we have added several new figures in the supplement. The first shows the distributions of predicted changes of $PM_{2.5}$ concentrations for each CMIP5 models. We also refer to this Figure in the main text.

Page 10, line 8-10. In general, these models agree on the sign of the change of $PM_{2.5}$ across the East by 2050s, but the magnitude of the change varies among models (Figure S5).

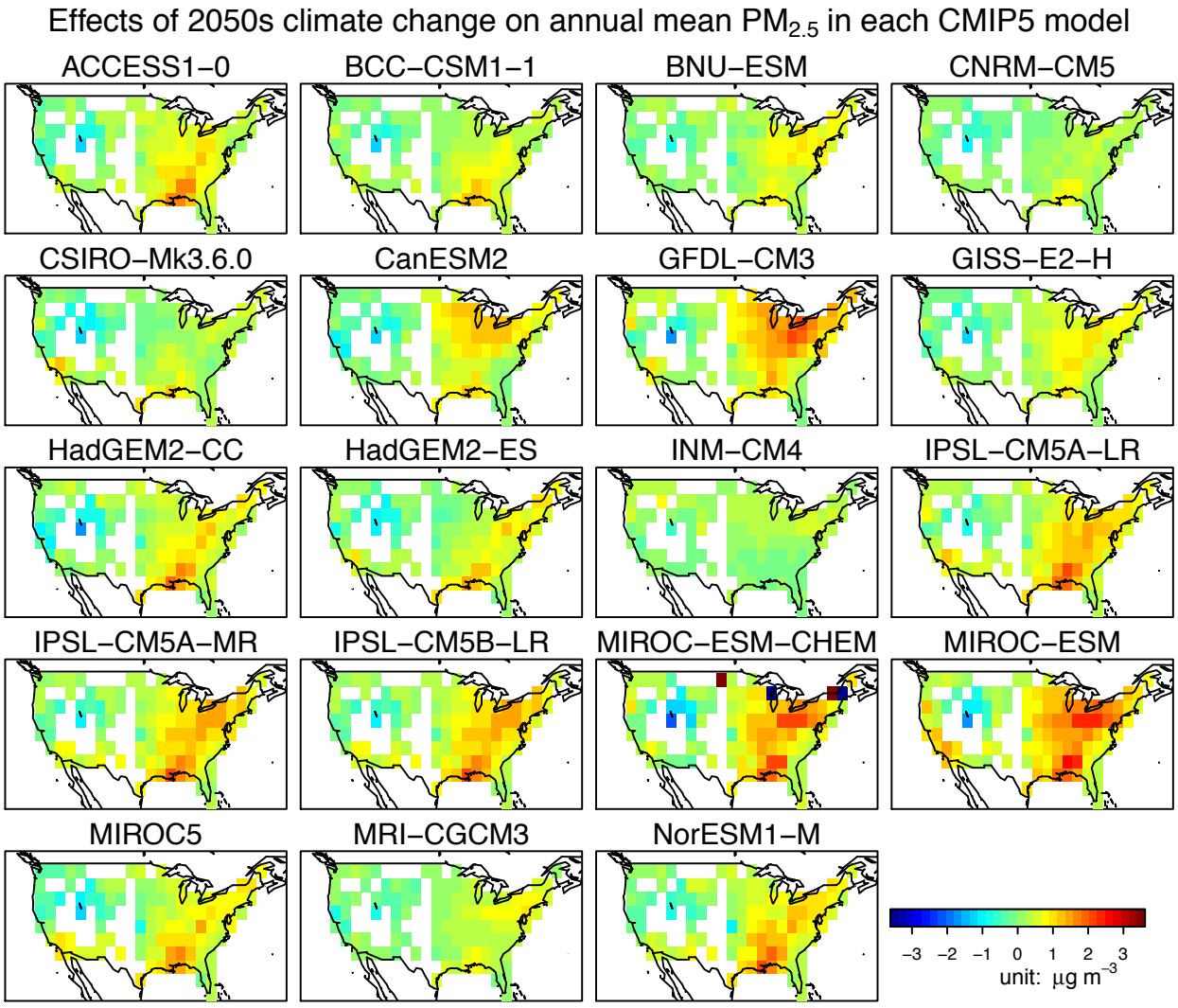

Figure S5. Effects of climate change from 2000-2019 to 2050-2069 on annual mean $PM_{2.5}$ concentrations, calculated with observed relationships of $PM_{2.5}$ and meteorology and with meteorology projected by each of the 19 CMIP5 models. White areas denote the regions with no $PM_{2.5}$ observations.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

We have also plotted the timeseries of $PM_{2.5}$ changes as annual, summertime, and wintertime means across eight regions from 2000 to 2069 (Figure S9-11). These figures have been added to the Supplement and are referred in the main text.

Page 10, line 14-19. We also examine the 2000-2069 timeseries of projected $PM_{2.5}$ concentrations as annual, summertime, and wintertime means, averaged over eight different U.S. (Figure S8-11). The spread in $PM_{2.5}$ trends is one measure of the uncertainty in our projections, arising in part from differences in model sensitivity to changing greenhouse gases and in part from internal variability of the climate system (e.g., Deser et al., 2013). Averaging results across

the CMIP5 ensemble reveals a robust response of $PM_{2.5}$ to increasing greenhouse gases, at least in some regions, giving us confidence in our approach.

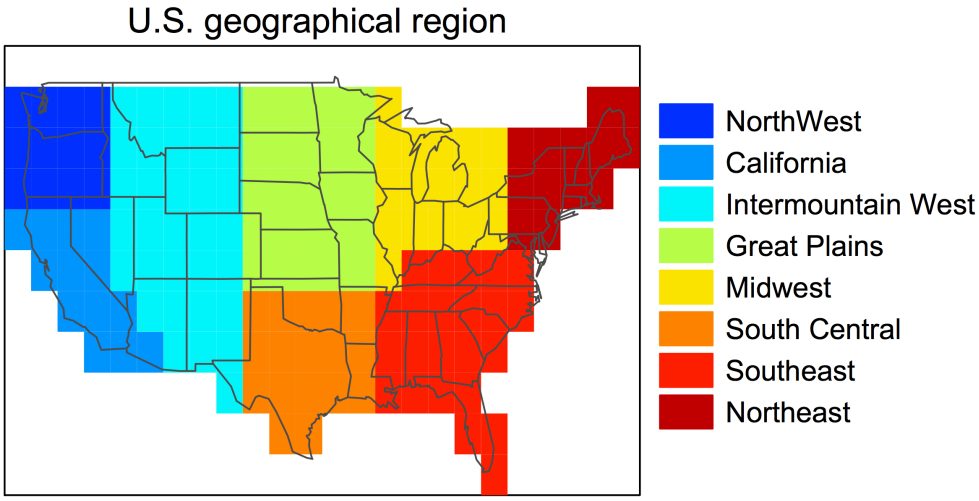

Figure S8. The eight U.S. geographical regions used for Figures S9-11.

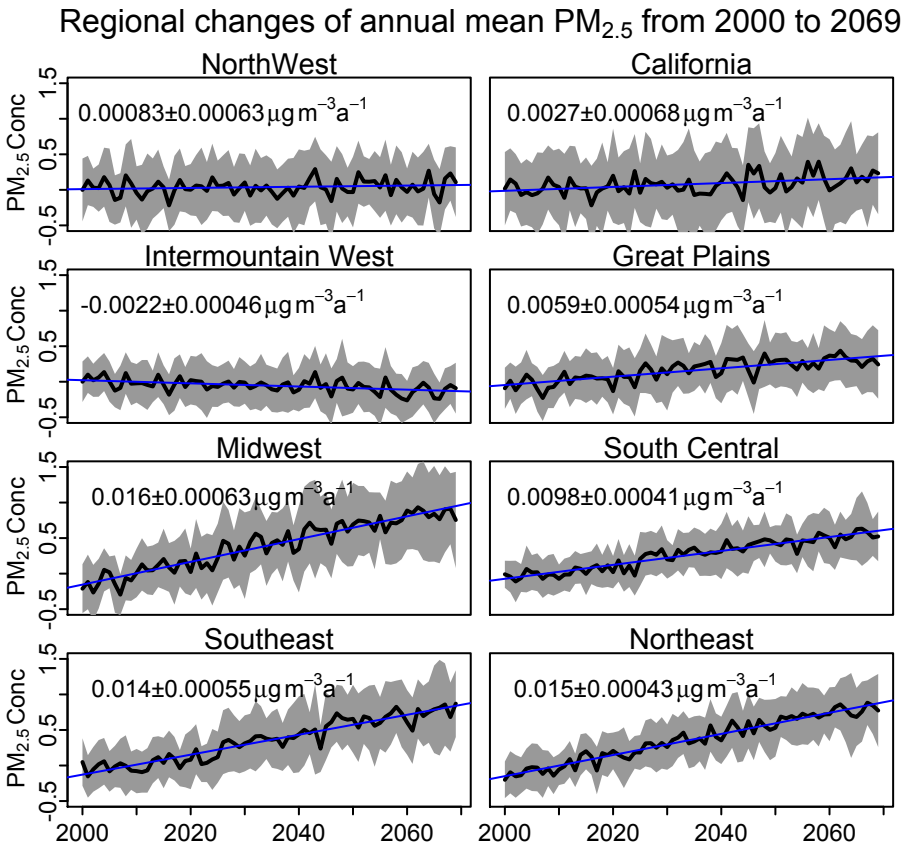

Figure S9. Regional trends in annual mean $PM_{2.5}$ concentrations from 2000 to 2069, calculated with observed relationships of $PM_{2.5}$ and meteorology and with meteorology projected by an

ensemble of 19 CMIP5 models. Shading denotes one standard deviation the mean change across models. The slopes of the timeseries over the 70-yr timeframe are shown inset. Figure S8 defines the eight regions.

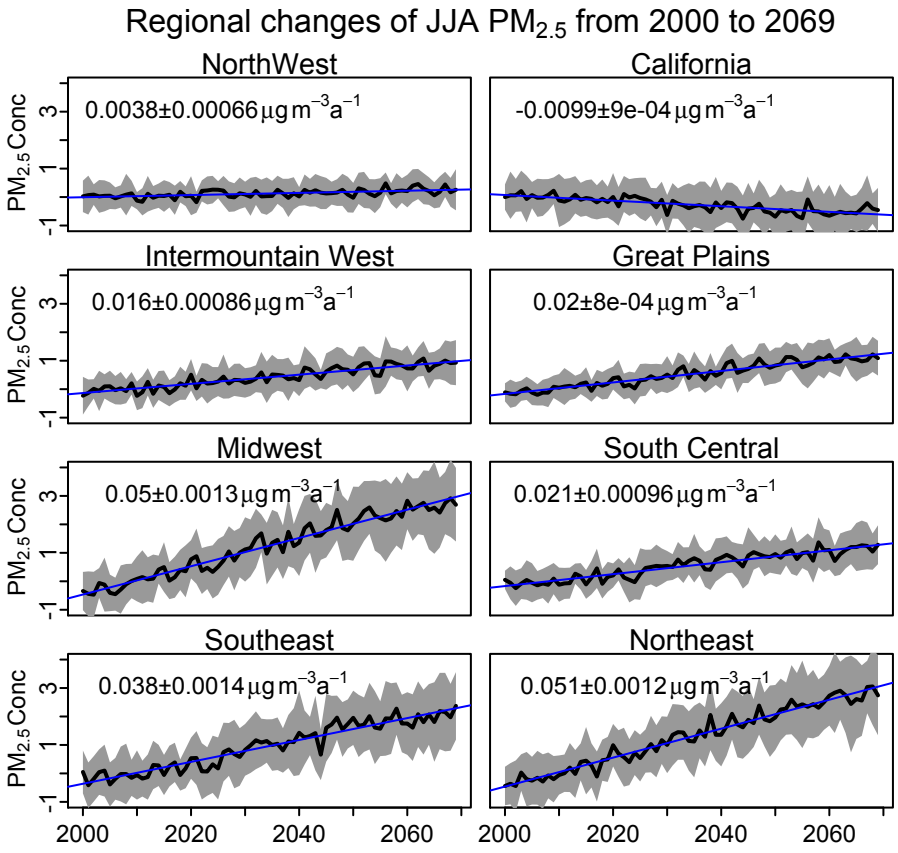

Figure S10. Similar as Figure S9, but for trend in JJA $PM_{2.5}$ concentrations.

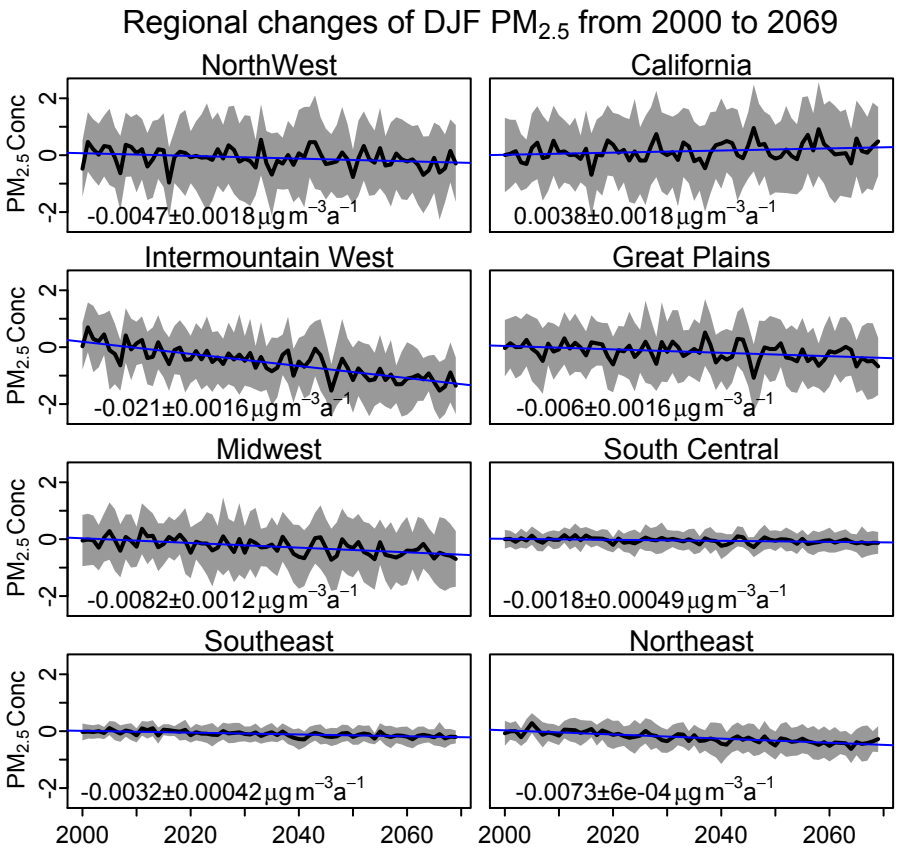

Figure S11. Similar as Figure S9, but for trends in DJF $PM_{2.5}$ concentrations.

To elucidate the influence from the internal variability of climate system, we need to analyze numerous simulations with one single model, as inferred from Deser et al. (2013). But here we only use the simulated meteorology from the first ensemble run for each model, so the sources of the uncertainty shown in Figure S9-11 consist of two parts, including the internal variability of climate system as well as the spread of physical parameterization among different models. Since the investigation into the influence of natural variability takes a lot of efforts and it is outside the scope of this study, we decide to briefly discuss this subject and leave the more detailed analysis to the follow-up study. Thanks for making such a good suggestion.

We now bring up the issue of internal variability of the climate system in the Discussion section.

Page 15, line 9-12. Drawbacks of this study include its assumption of constant anthropogenic emissions and its dependence on a relative short history (~15 years) of $PM_{2.5}$ observations. We also do not explicitly consider the role of interannual variability in the climate system and how that might influence our results (Deser et al., 2013).

- I suggest combining figures 4 and 5 and showing additional details of the penalty projections within the manuscript.

Response: We have combined Figure 4 and 5.

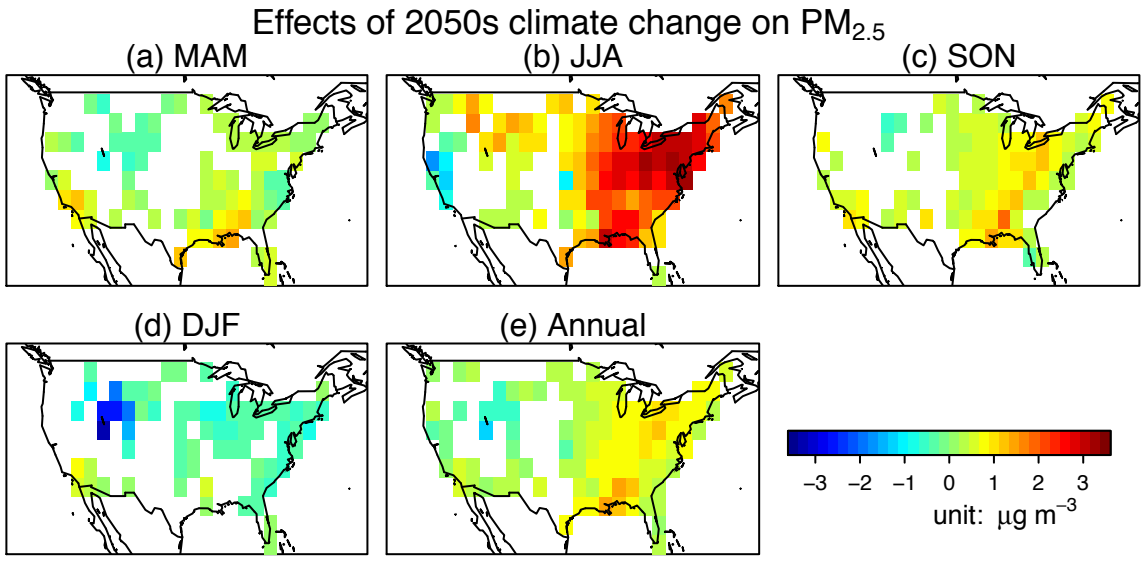

Figure 4. Effects of climate change from 2000-2019 to 2050-2069 on (a-d) seasonal and (e) annual mean $PM_{2.5}$ concentrations, calculated with observed relationships of $PM_{2.5}$ and meteorology and with meteorology projected by an ensemble of 19 CMIP5 models. The panels show the mean change in surface $PM_{2.5}$, averaged across the projections. White areas refer to the regions with no $PM_{2.5}$ observations or with fewer than 14 models yielding the same sign of changes.

- One analysis that is absent but would greatly benefit the study is a comparison of a climate penalty projection generated by the regression model to that generated with a CTM for the same GCM meteorological fields. Comparing the midcentury climate penalty estimated with the regression model, to a projection generated by driving a CTM with the same weather fields (e.g. a GEOS-Chem simulation driven by the present/future met fields from one of the CMIP5 models) would provide great insight into the potential to replace computationally expensive CTMs with a statistical model, and limitations associated with either approach. I encourage the authors to undertake this analysis in future work.

Response: This is an excellent suggestion for future work. We have added this idea to the Discussion section.

Page 15, line 13-15. Within these limitations, this study ….. It also demonstrates the utility of a computationally efficient model whose projections of the climate penalty on air quality can be readily compared to those from more traditional dynamic models.

- The penalty projections generated using the regression model assume that observed relationships between PM2.5 and meteorology remain valid at midcentury under significantly

different meteorological conditions and emissions levels. Is this an adequate assumption? An interesting analysis would be to compare the penalty projections of regression models generated under different levels of emissions within the 15-yr observational record.

Response: We have added one figure in the supplement to show the slopes of JJA $PM_{2.5}$ as well as its components with temperature for 1999-2006 and 2007-2013. We also discuss the influence of changing emissions on the sensitivity of $PM_{2.5}$ to climate change.

Page 11, line 14-24. One weakness of this study is that when estimating the sensitivity of $PM_{2.5}$ to meteorological variables, we do not consider the impact of changing anthropogenic emissions on this sensitivity. Figure S13 compares the slopes of monthly mean $PM_{2.5}$ and its components with temperature for two time periods: 1999-2006 summers with high anthropogenic emissions and 1997-2013 summers with low anthropogenic emissions. Using the monthly data, we find that the changes of sensitivity of $PM_{2.5}$ to temperature vary across different locations and species. As the anthropogenic emissions decrease, the slopes of $PM_{2.5}$ and temperature decrease over the Great Plains and Midwest, but increase slightly in the south Atlantic States. Sulfate exhibits decreased sensitivity across the eastern United States, and OC shows no significant pattern of change. Reasons for such inconsistencies may be related to the shorter time periods and therefore less robust sensitivity. In this study, we have thus chosen not to investigate the influence of changing emissions on the sensitivity of $PM_{2.5}$ to climate change using this statistical model.

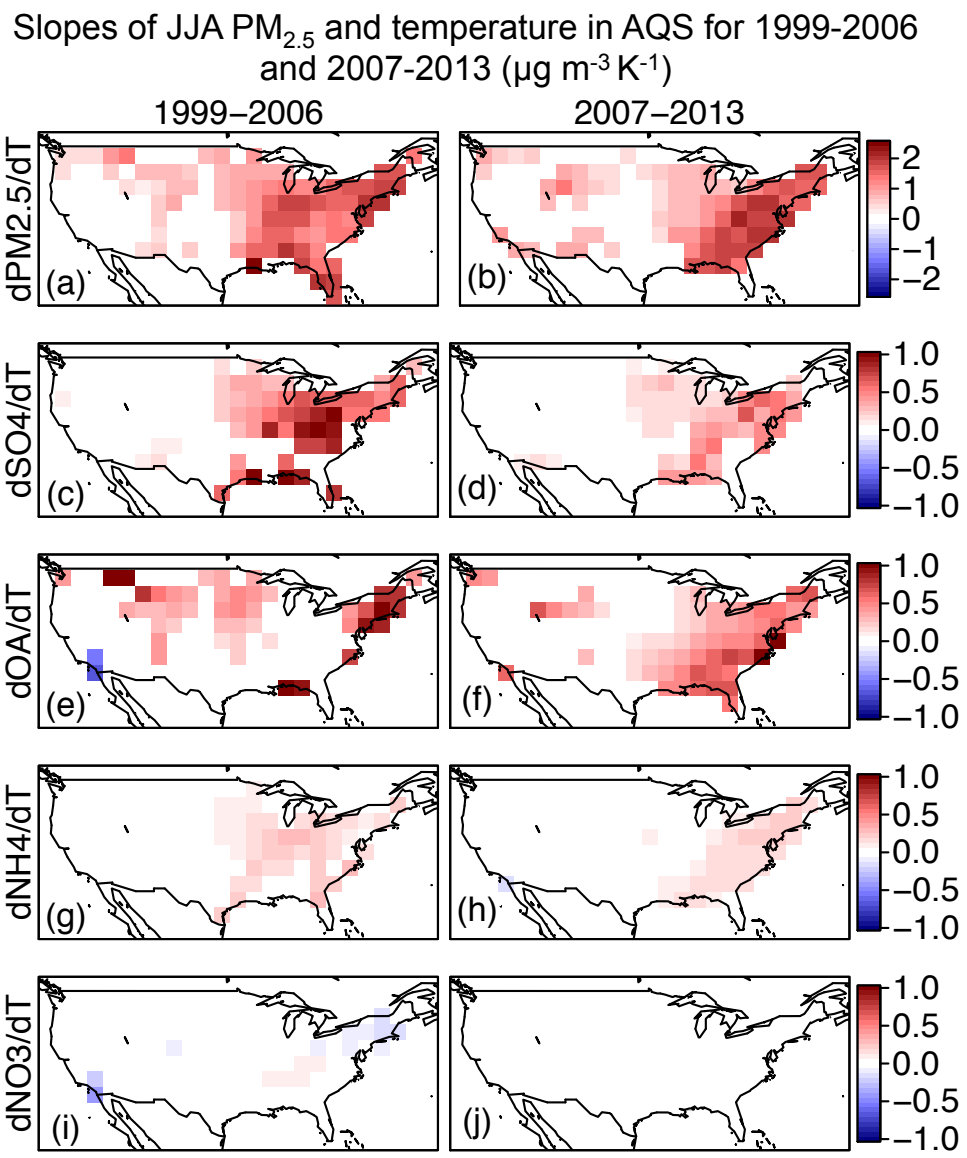

Figure S13. The slopes of detrended (a-b) monthly mean $PM_{2.5}$ and (c-j) different $PM_{2.5}$ components with surface air temperature for 1999-2013 summer months. Left column shows slopes for 1999-2006 with relatively high NOx emissions, and right column shows slopes for 2007-2013 with relatively low NOx emissions. Organic aerosol (OA) in Panel (e-f) is inferred from the measured organic carbon (OC) component using an OA/OC mass ratio of 1.8 (Canagaratna et al., 2015). White areas indicate either missing data or grid boxes where the slope is not significant at the 0.10 level. We note that the observation network has fewer sites in 1999 and 2000 than more recent years.

- The study explores PM2.5 response to surface temp in 4 CCMs, and then further investigates the dependency in GEOS-Chem. Given the differences between GEOSChem (a CTM) and CCMs, what insights from the GEOS-Chem analysis may useful to identify the causes for the discrepancies between observed and simulated sensitivities in CCMs? Would the authors expect to see any similarities?

Response: The reviewer raises an interesting question: is it possible that a CTM with specified meteorology calculates a different $PM_{2.5}$ sensitivity to temperature than CCMs with interactive chemistry and climate? Answering that question is beyond the scope of this paper as it would require a suite of sensitivity studies with CCMs. Below we iterate our response to a similar question from Reviewer 1.

Page 14, line 1-3. With regard to the ACCMIP results, understanding the failure of these models to capture the observed slopes of monthly mean total $PM_{2.5}$ and temperature is beyond the scope of this paper. Key diagnostics, such as the production rates of sulfate through different oxidation pathways, are not available.

- *PM2.5 concentrations and meteorology could be mapped on a finer resolution grid, and CMIP5 fields interpolated onto that grid. Would there be a benefit or significant change if the statistical regression model were built at higher resolution, rather than the coarse 2.5◦x2.5◦?*

Response: This is an interesting suggestion. For this study, we use 2.5°×2.5° horizontal resolution because this is the resolution in the NCEP Reanalysis 1, and most of the CMIP5 models have comparable resolution. We agree that mapping onto a finer resolution grid could be useful, especially for projections of air quality in urban areas. However, simple interpolation of CMIP5 meteorological fields could lead to large uncertainty and statistically downscaled fields are currently available for only a few CMIP5 variables (temperature and precipitation).

For now we choose not to speculate on the outcome of using finer spatial resolution in our regression model.

- *When listing the range of reported projections for climate change impacts on PM2.5 (e.g. pg. 3, line 7), I recommend using the updated range from the reviews published by Fiore et al. (doi: 10.1080/10962247.2015.1040526.)*

Response: We checked the range reported in Fiore et al. (2015) and have updated the text.

Page 3, Line 7. Reviewing earlier studies, Jacob and Winner (2009) and Fiore et al. (2015) concluded that the most of the projected effects of 21st century climate changes on $PM_{2.5}$ concentrations are in the range of ±0.1-1 μg m^{-3}, with changes up to ±2 μg m^{-3} in certain seasons or regions.