

Responses to reviewer

The authors have put in considerable effort for their revisions, and for the most part I am satisfied with their changes. The one point on which I still feel additional work is necessary is on the issue of PM_{2.5} speciation.

I understand the problem of data availability, and in my original review I was not suggesting that the authors create robust longterm speciated measurements where none exist. What I do (still) expect to see, however, is some significant discussion of this issue and how spatiotemporal patterns of PM_{2.5} speciation may affect the analyses and projections performed here. While we do lack much of the data necessary for a fully robust speciated analysis as performed in this manuscript, we are not completely blind with respect to how different types of PM_{2.5} precursors vary regionally, and much work has already explored differences in their expected behavior under varying ambient conditions. As just one example, Tai et al., 2010 (already cited in the manuscript) explore ways in which PM_{2.5} from sulfate and nitrate differ both in spatial distributions as well as their respective response to meteorology. Considering the significance of these differences, and the availability of previous studies like this one that have addressed them, I find it hard to accept the lack of meaningful discussion on the topic in this manuscript.

Furthermore, modern climate and chemical transport models (including CESM and the CAM4 atmospheric component) provide this speciation as gridded output, further supporting analysis on the model side. While full validation against observations may be outside the scope of this work, at the very least this output could aid in the interpretation of the model results themselves. Why has this not been done with the model output used here?

With this point addressed, I would feel comfortable giving full support for publication in ACP.

Response:

We would like to thank the reviewer for the constructive comments about the manuscript and we agree, and add the following text to the paper in lines 379-390 (at the end of Discussion section):

PM_{2.5} generally consists of multiple different aerosols each with different sources and variability; for example, the most important in the US are sulfate, organic matter, elemental carbon, nitrate, ammonium and desert dust. The different PM_{2.5} components respond to meteorological variables differently. The sulfate fraction of PM_{2.5} is predicted to be higher due to faster SO₂ oxidation under a warmer climate while the nitrate and organic fraction lower due to volatility (Dawson et al., 2007; Kleeman, 2008; Tai et al., 2010). Increased temperatures can lead up to higher biogenic emissions of PM_{2.5} precursors including agricultural ammonia, soil NO_x, and volatile organic compounds (Pinder et al., 2004; Bertram et al., 2005; Guenther et al., 2006; Riddick et al., 2016). Aqueous-phase sulfate and ammonium nitrate production increase with higher relative humidity (Liao et al., 2006; Dawson et al., 2007). Wildfires are an important source of black and organic carbon and they can increase or decrease depending on the local changes in climate and land use (Park et al., 2007; Spracklen et al., 2009; Kloster et al., 2012). Future exploration of the different components of aerosols and how each

responds to climate could provide more information about the effect on each type, but for these simulations, only PM_{2.5} was output and thus is not available for this study.

References:

- Bertram, T. H., Heckel, A., Richter, A., Burrows, J. P., and Cohen, R. C.: Satellite measurements of daily variations in soil NO_x emissions, *Geophys. Res. Lett.*, 32, L24812, <https://doi.org/10.1029/2005gl024640>, 2005.
- Dawson, J. P., Adams, P. J., and Pandis, S. N.: Sensitivity of PM_{2.5} to climate in the Eastern US: a modeling case study, *Atmos. Chem. Phys.*, 7, 4295–4309, <https://doi.org/10.5194/acp-7-4295-2007>, 2007.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181–3210, <https://doi.org/10.5194/acp-6-3181-2006>, 2006.
- Kleeman, M. J.: A preliminary assessment of the sensitivity of air quality in California to global change, *Clim. Change*, 87, S273–S292, <https://doi.org/10.1007/S10584-007-9351-3>, 2008.
- Kloster, S., Mahowald, N. M., Randerson, J. T., and Lawrence P. J.: The impacts of climate, land use, and demography on fires during the 21st century simulated by CLM-CN, *Biogeosciences*, 9(1), 509–525, 2012.
- Liao, H., Chen, W. T., and Seinfeld, J. H.: Role of climate change in global predictions of future tropospheric ozone and aerosols, *J. Geophys. Res.-Atmos.*, 111, D12304, <https://doi.org/10.1029/2005jd006852>, 2006.
- Park, R. J., Jacob, D. J., and Logan, J. A.: Fire and biofuel contributions to annual mean aerosol mass concentrations in the United States, *Atmos. Environ.*, 41, 7389–7400, 2007.
- Pinder, R. W., Pekney, N. J., Davidson, C. I., and Adams, P. J.: A process-based model of ammonia emissions from dairy cows: Improved temporal and spatial resolution, *Atmos. Environ.*, 38, 1357–1365, 2004.
- Riddick, S., Ward, D., Hess P., Mahowald, N., Massad R., and Holland E.: Estimate of changes in agricultural terrestrial nitrogen pathways and ammonia emissions from 1850 to present in the Community Earth System Model, *Biogeosciences*, 13(11), 3397–3426, 2016.
- Spracklen, D. V., Mickley, L. J., Logan, J. A., Hudman, R. C., Yevich, R., Flannigan, M. D., and Westerling, A. L.: Impacts of climate change from 2000 to 2050 on wildfire activity and carbonaceous aerosol concentrations in the western United States, *J. Geophys. Res.-Atmos.*, 114, D20301, <https://doi.org/10.1029/2008jd010966>, 2009.
- Tai, A. P. K., Mickley, L. J., and Jacob, D. J.: Correlations between fine particulate matter (PM_{2.5}) and meteorological variables in the United States: Implications for the sensitivity of PM_{2.5} to climate change, *Atmos. Environ.*, 44, 3976–3984, 2010.