Anonymous Referee #4

The referee's comments are noted in italics below, followed by our responses.

This paper describes a multi decade air quality simulation over the contiguous US using a regional scale application of the model "CMAQ" with downscaled meteorology from global climate scenarios. The authors employ constant anthropogenic emissions and investigate changes to ambient concentrations of ozone and PM2.5 (mass and chemical composition) due to temperature/climate changes only. They find the largest changes in [PM2.5] come from reduction in [NO3] in winter and increased [OM] in warmer seasons, presumably due to higher biogenic VOC emissions. Though the authors provide no direct evidence for the biogenic emission/higher [OM] - but I do happen to agree. The authors find the largest changes in 8 hour max ozone occur at the higher end of the distribution. Their findings are consistent with many previous studies. The downscaling and model applications methods are done well. The figures in the manuscript and supplemental information are excellent. However the analysis is not as strong as the rest of this work. Below I list specific reasons why I think this.

This paper contributes to the body of knowledge indicating temperature and air quality relationships. The authors have a unique opportunity to evaluate chemical-temperature trends and better context is needed.

We thank the referee for the constructive comments on our manuscript.

Specific Comments: A powerful motivation for this study is that future PM2.5 is less well constrained than ozone. The authors use of CMAQ with detailed particle chemistry in long term simulations is an improvement over global scale models (with less particle chemistry) typically used in such research. The model simulations have been conducted for time periods for which changes in ambient values of O3 and PM2.5 have been recorded. Links of measurements to EPA policy and temperature change can be evaluated. Why is this not part of the model evaluation and work presented in this manuscript? If the authors expect confidence in the future relationships they present, evaluation of past trends and relationships for retrospective periods builds confidence for their assessment and is necessary. What are the current d[O3]/dT and d[PM2.5]/dT, for example in the regions outlined in Figure 1? Can they be replicated by the modeling system?

CMAQ has been extensively evaluated as a chemical transport model, including "dynamic evaluation" of changes in simulated ozone levels in response to changes in emissions and meteorology (e.g., Gilliland et al., 2008; Foley et al., 2015). The simulations conducted in this study used meteorology downscaled from global climate model (CESM) simulations of the historical period 1995-2005 and of the future period 2025-2035 under three scenarios of greenhouse gas trajectories and radiative forcing. Because the effect of air pollutant (principally NO_x and SO_2, but also VOC) emissions changes on air quality is much larger than the effect of climate change-driven changes in meteorology over this period, we used constant levels of anthropogenic emissions in all our CMAQ simulations. This enables us to estimate quantitatively the impact on air pollutant concentrations of climate change in isolation from other factors, such as changes in domestic and international emissions of air pollutants. However, it would be inappropriate to evaluate ozone and PM concentrations from the CMAQ simulations in this study against historical measurements, because the emissions used in this study represented projections of future conditions, and were much lower than actual historical emissions. The $d[O_3]/dT$ and $d[PM_{2.5}]/dT$ modeled under such a different emissions regime would not be directly comparable to $d[O_3]/dT$ and $d[PM_{2.5}]/dT$ based on historical observations. This point is discussed in the Conclusions section (bottom of p. 10):

> **Observational evidence (Bloomer et al., 2009) and modeling studies (Rasmussen et al., 2013) have argued that the O_3 climate penalty (ppb K^{-1}) is lower at reduced levels of NO_x emissions. It is important to recognize that the results presented here use a projected 2030 emission inventory with continued implementation of NO_x**

emissions controls. The increase in O_3 resulting from a given climate scenario would be expected to be greater if NO_x emissions are higher than projected here, particularly in NO_x-limited regions such as the eastern U.S.

Instead, we have taken an approach in which we first evaluated CMAQ using downscaled historical meteorology and historical emissions changes in comparison to measurements of air pollutant concentrations over the period 2000-2010 (Seltzer et al., 2016). That study demonstrated model performance using downscaled meteorology was comparable to that typically obtained in standard air quality applications, which provides confidence in the overall method employed. Of course, regional air quality model results obtained using this methodology depend critically on the global climate model simulation being downscaled.

References:
Gilliland, A.B., C. Hogrefe, R.W. Pinder, J.M. Godowitch, K.L. Foley, S.T. Rao (2008): Dynamic evaluation of regional air quality models: Assessing changes in O3 stemming from changes in emissions and meteorology. *Atmos. Environ.* 42, 5110-5123.

Foley, K.M., C. Hogrefe, G. Pouliot, N. Possiel, S.J. Roselle, H. Simon, B. Timon (2015): Dynamic evaluation of CMAQ part I: Separating the effects of changing emissions and changing meteorology on ozone levels between 2002 and 2005 in the eastern US. *Atmos. Environ.* 103, 247-255.

There are temperature dependent anthropogenic emissions. Electricity sector emissions, in particular in the United States (e.g., California: Miller et al., 2008; Farkas et al., 2016), change with increasing temperature and this is not captured in this work. The absence of such relationships suggest changes at peak O3 and PM2.5 pollution is under represented here. The authors should note this and explain the uncertainty, the complications this introduces, in particular when they describe changes at the peak end of pollutant distributions.

We have added this point to the paragraph in the Conclusions (p. 11) discussing limitations of the present study:

To isolate the effect of climate change on air quality, we kept anthropogenic emissions constant across all modeled years. However, electric sector emissions increase during peak temperature events due to increased demand for air conditioning, and emissions from electric generating units used to provide power during peak periods are less strictly regulated (Farkas et al., 2016). The increased emissions associated with increased electricity demand during heat waves is not represented in our analysis, potentially underestimating the impact on upper percentile and annual 4th-highest O_3 levels.

Starting at Line 22, page 1: The authors state that due partly to Tier 3 emission standards for motor vehicles, anthropogenic emissions are expected to decrease through 2030. Is this still true? How does the Ozone Standards Implementation Act of 2017 affect/not affect expected trends in emissions and ambient air quality?

Our statement and analysis is based on existing legislation and regulations. The Ozone Standards Implementation Act of 2017 is a bill that has passed the U.S. House of Representatives but has not yet been acted upon by the Senate. Accordingly, it does not yet have the force of law.

The references for AERO6 (Simon and Bhave, Nolte et al.) are insufficient to describe the AERO6 module. Further, the authors discuss that some of the largest [PM] prediction changes are due to temperature induced changes on BVOC emissions that affect [OM] predictions in some portions of the the US. The chosen references do not explain why this would be the case in the model at all. Please provide better reference(s) that help readers understand the relationship between biogenic VOC emissions and the connection it PM2.5 OM (presumably biogenic secondary organic aerosol) in CMAQ.

We have added a reference to Carlton et al. (2010), which describes the secondary organic aerosol model in this version of CMAQ. Additionally, we now cite Carlton and Baker (2011) for the BEIS biogenic emissions module.

Line 25, Page 5: Can the authors explain what "wet bias" means and the implications? Does this mean excess precipitation? Does this mean the implication is there is more wash out/cleaning of the atmosphere?

We have modified the text to read "while CFSR precipitation is positively biased." As noted in the Discussion section (p. 9), "Scavenging of soluble aerosols by precipitation is an important removal process for atmospheric particulate matter." Overestimated precipitation (especially too-frequent precipitation) would overestimate removal of particulate matter from the atmosphere due to scavenging and wet deposition, and therefore result in underestimated $PM_{2.5}$ concentrations.

Line 12, Page 9: "This supports the conclusion that warmer temperatures in a future climate results in increased partitioning of aerosol NO3 to HNO3" Presumably, the authors can test this idea/hypothesis in their model output?

This sentence has been deleted. We examined changes in seasonal mean concentrations of gas-phase HNO_3 as well as the fraction of total nitrate in the gas phase ($[HNO_3]/[TNO_3]$). Both showed some increases in areas where aerosol NO_3 decreased during winter, but the changes in HNO_3 and HNO_3/TNO_3 were smaller and less widespread than the change in NO_3.

Editorial
Line 22: "Pope III", is that formatting correct?

We have modified the text so that the parenthetical citation is (Pope, 2007) and the full reference reads Pope, III, C.A.