Interactive comment on “The effects of global change upon United States air quality”

by R. Gonzalez-Abraham et al.

Response to Anonymous Referee #1

We appreciate the referee for his/her constructive and thorough review. We have revised the manuscript according to the referee’s comments. During the revision, we learned there were bugs in CMAQv4.7 in processing emissions, as described here: www.cmascenter.org/cmaq/documentation/4.7.1/RELEASE_NOTES.txt. Accordingly, we have re-run all simulations using CMAQv4.7.1, resulting in substantial changes to the PM2.5 section. Changes in ozone also occurred as a result of upgrading to CMAQ 4.7.1 due a combination of updates in emission, advection and plume rise emission modeling. Most notably, a larger increase in ozone in coastal urban areas is projected as a result of changes in US anthropogenic emissions. We believe the manuscript has been much improved in addressing comments by both referees and updating the manuscript to reflect the new simulations. Our detailed responses to Referee #1’s comments (in italics) appear below.

General Comments

This paper describes a climate model downscaling study to investigate the impact of future climate change (following the IPCC A1B scenario) on US air quality. The authors find that daily maximum 8 h average ozone (DM8O) will increase by 2-12 ppb in the US due to increased temperatures, enhanced biogenic emissions, and land use changes, which will overwhelm the reductions in DM8O that would have happened from reductions in US anthropogenic emissions in the absence of climate change. They also find that PM2.5 levels are expected to increase 2-4 μ g m-3 in the Southeast US and nearby regions due to enhanced biogenic emissions and land use changes. This is a well-written paper on a scientific question relevant to ACP. The methods are valid and clearly outlined, as are the modeling experiments performed. Substantial conclusions are reached that are generally supported by the model results. There are a few places where the discussion is confusing or not supported by the results presented, and the tables need some work, but overall I recommend publication after minor revisions to address by concerns below.

We appreciate the reviewer’s positive comments. We have revised the manuscript according to both referees’ comments to have more clarity in the text and updated figures and tables. Below we address Referee #1’s comments directly.

Minor Comments

1) *P31844, L13-14: Since you mention evaluating the impacts of Asian emissions as a goal of the study, you should also include your findings on their impacts on O3 and PM2.5 in the abstract.*

We have revised the abstract to include: "The model predicts an average increase of 1-6 ppb in DM8O due to projected increase in global emissions of ozone precursors."

2) *P31853, L8-11: You say MARKAL was used to get growth factors of NOx, SO2, and PM2.5, but then mention the use of CO2 factors as well. Should CO2 be on the initial list as well?*

The Referee is correct. We have revised the manuscript to include CO2.

3) *P31853, L10-11: I'm not sure that it is appropriate to use CO2 growth factors for CO, NH3, VOCs, HCl, and chlorine. I understand doing it in the absence of other data, but how realistic do you think it is that CO will increase proportionally with CO2 even with future control technologies being implemented to reduce NOx and SO2 emissions. This gives a 70% increase in CO and 20% increase in NMVOCs in the Midwest – how realistic is that? And how does this affect your results?*

We agree that using CO2 growth factors for CO and VOC is not the most appropriate. Historically CO emissions and concentrations have been decreasing while CO2 emissions are increasing (http://www.epa.gov/airtrends/carbon.html). Also, for the mobile sources, the CO and NMHC emissions will either level off or will continue decreasing (McDonald et al., 2013, DOI: 10.1021/es401034z). Therefore, increase in CO emission as presented in this manuscript means higher ozone (less reduction from the current decade) in urban (high-NOx) areas in comparison to a projection with CO-specific growth factor. We have added text to this section to note this caveat. We have added to section 3.3.1: "The smaller reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase in VOC and CO emissions from business-as-usual scenario of ESP v1.0 which uses CO2 as a surrogate for growth factors for CO (Loughlin et al. 2011)."

4) *P31857, L12-16: You are really stretching the words "majority" and "most" here – the results in Figure 6 don't look all that great. The claim that PM2.5 meets the guide- lines for 4 regions seems false to me –by my eye 5 of the PM2.5 results fall outside the weaker bias and error constraints. I would reword this section to be a little more accurate about the model performance.*

Note that due to the lack of speciated PM data for our period of simulation, we have removed the section on model performance for speciated PM (the observed data in the ACPD version were from 2003-2008 and we believed is not representative of our period of simulation). Also, the PM2.5 section changed substantially after the new simulations were performed.

5) *P31860, L15-22: This paragraph confused me on my first read-through, as you discus the increases in isoprene, monoterpenes, and overall BVOCs all in the first sentence. I'd try to separate out this discussion, and add a total BVOCs bar to Figure 3 as well. It is also not clear when you say "biggest increase" if you mean biggest percentage increase or biggest absolute increase.*

In Figure 3, having isoprene and monoterpene emissions as separate bars is useful to aid the discussion of biogenic SOA; however, we don't think it is necessary to include another bar in Figure 3 for total BVOC because the change in BVOC emissions is dominated by changes in isoprene emissions. For clarity, we have modified the text to read:

> The model projects bigger percentage increase in monoterpenes than isoprene across the domain; however, total isoprene emission is an order of magnitude higher and thus dominates the changes in total BVOC. The increase in total BVOC ranges between 17% and 45%. The only region that is projected to have reduced total BVOC emissions is the Northwest, where the model simulates a 7% reduction in isoprene emissions (Figure 4) that in absolute amount is greater than the 20% increase in simulated monoterpene emissions.

6) *P31861, L9-10: I think this sentence on monoterpenes belongs in the next paragraph. Also, you say "because of higher across the domain" – higher what?*

We have rearranged the text in Section 3.2 for better clarity, and the sentence referred to by the referee is no longer in the text. Also, please note that the section has changed drastically.

7) *P31863, L8-9: Cloud cover only increases in the Northwest and Central regions, correct? Can you make that clear here?*

The reviewer is correct that the increase in cloud cover is in the Northwest and parts of the central region (western Montana). We have modified the text to read: "The reductions in DM8O concentrations in the Northwest resulted from an increase in cloud cover and lower solar radiation reaching the ground, and resulting in a reduction in photochemistry."

8) *Comment from Referee. P31864, L1-6: I'd like to see more discussion here about how the emissions differ between this study and the previous ones and how the climate simulations differ. Some of this information is in section 2, but it would be nice to restate it here to make the discussion of the results clearer.*

As requested by the reviewer, we have added more discussion to the text. First, the reference was corrected to be Nolte et al. (2008) instead of Leung and Gustafson (2008). We now have "However, the difference in geographical features of DM8O changes with Nolte et al. (2008) and Tagaris et al. (2007) suggests that the source of

disparities resides in the simulated regional meteorological fields resulting from different global climate models, regional climate models and the methods used to estimate emissions from biogenic sources. We used the ECHAM5 global climate model results while both Nolte et al. (2008) and Tagaris et al. (2007) used results from the GISS global climate model. For regional climate simulations, both Nolte et al. (2008) and Tagaris et al. (2007) used MM5 while we used WRF here. In contrast with Nolte et al. (2008) and Tagaris et al. (2007) who use the BEIS/BELD3 (Hanna et al., 2005; http://www.epa.gov/ttn/chief/emch/biogenic/) tool to compute biogenic emissions, this investigation estimates the biogenic emissions with MEGAN v2.04. MEGAN v2.04 generally predicts higher isoprene emissions than BEIS (Hogrefe et al., 2011; Sakulyanontvittaya et al., 2012). Hogrefe et al. (2011) shows that for the Northeast, MEGAN leads to higher DM8O by upwards of 7 ppb using 2005 anthropogenic emissions; however, under a scenario by which anthropogenic NOx emissions were reduced by ~60%, difference in DM8O was generally 3 ppb because of greater sensitivity to NOx emissions when MEGAN was used."

9) *P31864, L11: Please be quantitative about the size of the decrease in DM8O you are discussing here.*

We have now included "2 to 4 ppb" in the text.

10) *P31864, L18: Please be quantitative about the size of the reduction in the VOC to NOx ratio and the depletion of DM8O you are discussing here*.

We have revised the text to explain the change in BVOC and NOx emissions under the land use change scenario instead of VOC to NOx ratio: “When land use changes are included along with biogenic emissions (Simulation 3), the increase in BVOC emissions is projected to be less, while NO emission is projected to increase in areas where natural vegetation is converted to cropland. This combination leads to higher DM8O in Simulation 3 than Simulation 2 (Simulation 3; Figure 12d).”

11) *P31864, L21: The reduction of BVOC emissions due to land use changes (discussed on P31861, L7-9) also plays a role here, right?*

Yes, there is a decrease in BVOC emissions and an increase in NO emissions where natural vegetation is converted to cropland. The text has been updated to reflect this. See response to comment above.

12) *P31864, L28: Instead of saying “mostly” can you be quantitative?*

We revised the sentence to be more quantitative. The sentence now reads: “The increase in DM8O is mostly due to an increase in global emissions of ozone precursors from the semi-hemispheric domain, which contributes to an increase of 2-6 ppb under current climate conditions (Fig. 13f).”

13) *P31865, L13-15: These two statements are not clearly supported by the results in Figure 12. In Figure 12f it looks like Asian emissions lead to a very slight increase in the*

southern half of the US and very slight increases in the northern half, with no reason to single out the western US as a homogenous group. The impact of climate change and biogenic emissions in Figure 12c seems to increase PM2.5 throughout the US rather than increases and decreases in different regions.

We had misplaced text on ozone in this section. This error has been corrected in the revised manuscript. The line now reads as follows: "Changes in global emissions do not have a significant impact on $PM_{2.5}$ concentrations, while changes in the climate and biogenic emissions can lead to both increases and decreases in $PM_{2.5}$ depending on the region."

14) *P31866, L1-2: I think you should explicitly state here that your results for sulfate are different than Avise et al. (2009).*

This section has been re-written to reflect the results of new CMAQv4.7.1 simulations after discovery of ammonia emission bugs in CMAQv4.7.

15) *P31866, L23: Why is there no discussion of aerosol ammonium here? The effect of the boundary conditions on ammonium is huge in Table 3 and should be addressed in the text.*

We have revised the PM2.5 section to reflect the new simulation results and include discussion of ammonium.

16) *P31867, L1: Can you be quantitative instead of saying "insignificant"?*

The line now reads as follows: "…the effect of climate change alone (with no change to biogenic emissions) on total $PM_{2.5}$ concentrations over land is a change of less than 1 $\mu g\ m^{-3}$"

17) *I think you need to discuss the increases in SOA in the Northwest region here –SOA increased with increases in BVOCs, but sulfate decreased, in contrast to the other regions that had negligible changes in sulfate with increased BVOCs.*

The decrease in sulfate with increasing BVOC and SOA is due to the competition between BVOC and SO2 for OH. The text now includes: "The smaller increase or absolute reduction in sulfate in comparison to the climate-only case is due to the competition between BVOC and SO2 for the availability of OH, which is an oxidant for both."

18) *P31869, L24: The "positive influence (reduced concentrations)" phrasing is confusing, consider rewording this to make what you mean clear.*

The text now reads: "Decreases in future US anthropogenic emissions of ozone precursors are the only consistently beneficial influence that improves the air quality in the US; updated assumptions to generate scenarios of future US anthropogenic emissions may show even more positive influence."

Sections

19) *Section 4: You should be as quantitative as possible about the magnitudes of the impacts here, as you are in the abstract.*

Our main aim for the conclusion section is to focus on the influence of each individual attribution within the context of one single future scenario (the combined changes), rather than summarizing the individual contributions quantitatively.

20) *P31870, L3-5: Here I'd stick to the regions you defined in Figure 3 and avoid less specific phrases like "East regions" and "regions with high biogenic emissions."*

Text has been edited to reflect referees comments: "…2) climate changes (namely, increased temperatures and solar radiation) which increase ozone concentrations in the Central, South, Midwest, Northeast and Southeast regions of the domain; and 3) increases in US BVOC emissions which also increase ozone concentrations in regions with high biogenic emissions such as the South, Midwest, Northeast and Southeast"

Figures and Tables

21) *Table 2: This is like Table 5 for PM2.5, but where is the equivalent of Table 3 for DM8O? I'd suggest adding a table with similar quantitate results for each region.*

We agree with referee's comment. A new Table 2 with the percentage change in DM8O from each scenario has replaced the old Table 2; and a new Table 5 with the percentage change in PM2.5 has replaced the old Table 5.

22) *Tables 2-5: It's not clear in these tables what the scenario names in the column headings mean. Does "BVOC" include climate impacts, so that it is Scenario 2 minus Scenario 0, or does it only look at the impact of BVOCs on top of climate, and so is Scenario 2 minus Scenario 1? The same question applies for the land use changes, which aren't listed in Tables 2 and 5 but are listed in Tables 3 and 4 as BVOC future land use". Is this Scenario 4 minus Scenario 3, 2, 1, or 0? Please clarify this in footnotes in Table 2 and then use consistent definitions for all other tables.*

The original Tables 2 and 5 have been replaced with tables showing percentage changes of DM8O and PM2.5. The new Tables 2 and 5 as well the original Tables 3 and 4 now have column headings listing the simulation numbers consistent with those of Table 1.

23) *Table 4: You have a row called "SOA" – does that mean these results are only for SOA and not primary organic carbon? Doesn't this contradict your caption? Are the POC results just missing?*

The caption should state only secondary organic aerosol. The table caption has been corrected.

24) *Figure 3: Add a bar for the percentage change in total BVOCs in each region as well.*

Because total BVOC is dominated by isoprene, percent change in total BVOC is very similar to percent change in isoprene. Therefore, we do not think it is necessary to add an additional bar for BVOC.

25) *Figure 5: Add a legend to the box and whisker plot as in Figure 4.*

The figure has been updated to include a legend. Note that the original Figure 5 is now Figure 6.

Typos and Technical Corrections

26) *P31856, L6: I think you mean Figure 11a, not the top of Figure 10. And I think this should be renumbered Figure 6, as it comes after you mention Figure 5 but before you mention Figure 6*.

The reviewer is correct about the typo. The number is now Fig 12a in the revised manuscript. We have kept the figure location because it is discussed most heavily in the results section later in the manuscript.

27) *P31856, L14: I think you mean Figure 12a, not the top of Figure 11. And I think this should be renumbered Figure 7*.

The reviewer is correct about the typo. The number is now Fig 13a in the revised manuscript. As noted in the above comment, we prefer keeping this figure in its current position because it is discussed most heavily in the results section later in the manuscript.

28) *P31858, L21: "the result of" instead of "resulted of"*

We have corrected the error.

28) *P31864, L11: "The decrease", not "this decrease"*

We have corrected the error.

29) *P31865, L24: I think you mean Table 3, not Table 2. And shouldn't the Southeast region also be in this list?*

Yes, Southeast has been added to the list

30) *P31867, L14-15: You can't say "in all regions" and then discuss an exception. Try "in nearly all regions" and "The lone exception."*

We have corrected the error.

31) *P31868, L8: Just reference Figure 12c here, and then reference Figure 12d in L11 below.*

We have added the figure references in the text. (The relevant figures are now numbered 13c and 13d.)

32) *P31868, L11: Shouldn't the Southeast region also be in this list?*

We have corrected the text.

33) *P31869, L2: Remove comma after "monoterpene"*

We have corrected the typo.

34) *P31870, L28: "intended to", not "intended so"*

We have corrected the typo.

35) *P31871, L1: Typo, remove the "7".*

This was an error introduced during typesetting.

36) *P31871, L3: "take" not "takes"*

We have corrected the typo.

37) *P31871, L3: The semicolon should go before the word "and" not after.*

We have corrected the typo.

"The effects of global change upon United States air quality"

by R. Gonzalez-Abraham et al,.

Anonymous Referee #2: Response

We appreciate the referee for his/her constructive and thorough review. We have revised the manuscript according to the referee's comments. During the revision, we learned there were bugs in CMAQv4.7 in processing emissions, as described here: www.cmascenter.org/cmaq/documentation/4.7.1/RELEASE_NOTES.txt. Accordingly, we have re-run all simulations using CMAQv4.7.1, resulting in substantial changes to the PM2.5 section. Changes in ozone also occurred as a result of upgrading to CMAQ 4.7.1 due a combination of updates in emission, advection and plume rise emission modeling. Most notably, a larger increase in ozone in coastal urban areas is projected as a result of changes in US anthropogenic emissions. We believe the manuscript has been much improved in addressing comments by both referees and updating the manuscript to reflect the new simulations. Our detailed responses to Referee #2's comments (in italics) appear below.

The authors present findings related to the application of the WRF and CMAQ models to downscale future climate predictions estimated with a global model simulation. They find that improvements in ozone and PM air quality due to decreases in anthropogenic emissions are somewhat offset due to climate change and increases in global emissions. The manuscript is generally well written and the approach is thoroughly described. However, certain aspects of the study need further explanation. The scope of this manuscript in its current form does not present particularly novel work in this area, especially for ACP.

While the manuscript does not present a novel methodology, the set of simulations do present different elements than previous publications. The novelty is stated below.

a) The paper separates the effect of climate change on air quality from the effect of changes in biogenic emissions due to climate change. This approach, while unrealistic, situates the largely documented effect of climate change (with changes in biogenic emissions) on air quality on a different perspective. The approach followed in this manuscript shows that climate change alone has a dominant effect on PM2.5 and O3 concentrations over the increase in biogenic emissions in some regions of the US. Also, the regional changes in land use and the resulting effect in the emissions of BVOC with

respect to the emissions of NO offer a wider set of sensitivities of the model to changes in emissions.

b) Unlike previous research, the paper shows the sensitivity of Ozone and the inorganic fraction of PM2.5 to different regimes of VOC and NOx concentrations. The approach is followed in the form of a sensitivity analysis.

c) The manuscript presents a wider range in the levels of atmospheric oxidants and their effect on Ozone and PM2.5 due to climate, biogenic and anthropogenic emissions.

d) In the case of DM8O, our approach presents a diverse perspective of the reactions that involve the gas and the organic and inorganic particle phase. One example is the importance of sequestration versus recycling of isoprene nitrates in the SAPRC99 mechanism presented in section 3.3.3.

1) *The current year model performance is concerning. If these results are consistent with other downscaling efforts then maybe that is a notable addition to the literature and could be focused on in more detail as a potential confounding factor in interpreting these projected future year scenarios.*

As noted above, due to a bug in the emission processing in CMAQv4.7 we have re-ran all simulations using CMAQv4.7.1. As a result, the model performance for $PM_{2.5}$ has improved (Figure 7 of the revised manuscript). Also, The model performance has to be understood in terms of the climatological perspective of the investigation. Both current and future meteorological conditions are derived from downscaled climate simulations rather than meteorological reanalysis. In our particular study, ECHAM5 global climate model was used. Biases in the drivers of ozone and PM formation and removal are in both the current and future decade simulations. Previous studies that used downscaled climate realizations to drive air quality models also show overestimations of DM8O (Tagaris et al., 2007; Nolte et al., 2008) and underestimations of total PM2.5 mass (Tagaris et al., 2007).

2) *RCP scenarios are not always good indicators of future changes in air quality since they are focused on greenhouse gases rather than O3 and PM precursors. The conclusion that ozone reductions due to lower anthropogenic emissions being totally offset by increases in global ozone and climate change impacts seems inconsistent with other studies. Even more importantly, observed ozone concentrations continue to trend*

downward in many places in the United States suggesting US emissions reduction impacts are outpacing climate and international emissions increases. We are 10 years past the baseline for this model simulation so that reality should be recognized in some way.

This study is based on the SRES (used in IPCC's Fourth Assessment Report (AR4) rather than the RCPs (used in AR5). The Greenhouse gas emissions from the SRES scenarios are used to drive the global climate models. We agree that they are not always good indicators of future changes in air quality; but in many countries it is the only proxy for future scenarios. As presented in the results, we explicitly recognize that the scenarios of global and regional anthropogenic emissions are the reason for the inconsistencies between studies. We also explicitly acknowledge in the conclusions that updated emission inventories and future emission scenarios are needed to improve the assessment of the impacts of global change upon the US.

While the emission inventory, the assumptions for the regional and global emission projections and the meteorological simulations may be outdated, the results presented in this manuscript are still relevant evidence of the dependence of the US air quality of global emissions and the need of rigorous emission controls.

Nevertheless, additional information was added to the manuscript for clarity. A figure with the emissions from the semi-hemispheric domain was added. The figure shows the dramatic increase in NOx emissions across the domain and aims at the reason behind the increase in DM8O. A line addressing a possible higher reduction in ozone as a result of updated future regional emission projections was also added to the conclusion.

3) *How closely did the projections using MARKAL match those of the IPCC A1B scenario for areas where both are applied? Are the ozone changes aggregated and postprocessed similarly to other similar studies? More clarity is needed regarding methane. The authors state they do not consider methane but may have indirectly as impacts of increasing methane emissions globally may be part of the global 2050 simulation*

Emissions from the IPCC A1B scenario and results of the MARKAL methodology were never overlapped on the same simulation. However, CO_2 is underestimated in the A1B scenario in comparison to the US emission inventory and MARKAL projection.

The metric used (daily maximum 8 hour average ozone) is common to the studies based on US air quality. Methane emissions are not used in the chemical mechanism used in CMAQ. The impact of methane is in the global climate represented in the ECHAM5 simulations and passed onto the downscaled meteorological fields that drive CMAQ. However, impact of increased methane on chemistry is not considered in this study; in all our CMAQ simulations, methane concentration is fixed at 1.85 ppm.

Author's Change in the manuscript. Two changes have been made:

a) Changes in the text of the introduction to highlight the direct effect of methane. "Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the direct effect of emissions of methane in the air quality simulations"

b) Section 2.5. Added the line "Methane concentration is fixed at 1.85 ppm for all CMAQ simulations."

Introduction section:

4) *First paragraph; the authors have 2 references for negative health effects associated with O3 and PM. Typically journal articles focus on either O3 or PM so it is not clear that both of these references are relevant for both O3 and PM. End of second paragraph; it would be appropriate to also reference Cooper et al 2010 and Cooper et al 2012 here.*

For clarity, we added "and, in the case of PM," to the sentence. We also added Cooper et al. 2010 and Cooper et al. 2012, as requested.

5) *Page 31847 lines 1-5; It would be helpful for the reader if the authors add what time scale these increased ozone concentrations are seen. Annual average? Average of the highest days? Just the highest days or are low ozone days part of the average?*

The text was modified for clarity: "In summary, despite the differences in modeling elements, all studies found an increase in the summer average of the daily maximum 8-hour average ozone concentrations over large regions of the simulated CONUS domain on the order of 2 to 8 ppb (Weaver et al., 2009)"

6) *Page 31847 lines 6-20; this section should mention the time period over which the climate changed and whether the climate impact was isolated or US emission changes were also considered.*

The manuscript was modified for clarity and the time periods were also added to the paragraphs: "This downscaling approach has been used in a variety of studies in

Europe, Canada, and Asia at different time scales of climate change (e.g., Liao et al., 2006 [2000 to 2100]; Langner et al., 2005 [2000 to 2060]; Forkel and Knoche, 2006 [2990 to 2030]; Meleux et al., 2007 [1975 to 1985]; Kunkel et al., 2007 [1990 to 2090]; Lin et al., 2008 [2000 to 2100]; Spracklen et al., 2009 [2000 to 2050]; Kelly et al., 2012 [2000's to 2050's])." Later in the fifth paragraph of the introduction, we also added "projected to the 2050s" for the US studies funded by the EPA.

7) *The A1B scenario has aggressive methane emissions growth through 2040. Is that impact included in the semi-hemispheric model? That is not clear but important in interpretation of the global impacts offsetting decreases in US anthropogenic emissions.*

The air quality simulations in this study do not take into account of future changes in methane. This caveat was added in the introduction and the first paragraph of section 2.5:

> "Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the direct effect of emissions of methane on the air quality simulations"
>
> " Section 2.5. Added the line "Methane concentration is fixed at 1.85 ppm for all CMAQ simulations."

Methods section:

8) *Sections 2.3 and 2.4: A summary Table similar to Figure 3 showing the non-U.S. emissions would be extremely helpful with interpretation of the results. This is especially critical given the main conclusion that global emissions increases are offsetting reductions in US anthropogenic emissions. It is unclear what is driving the large increases in VOC, primary PM2.5, and ammonia. Primarily emitted PM2.5 is fairly cost effective for point sources and VOC would seemingly be decreasing due to vehicle fleet turnover and mobile source sector regulations. It is not clear why emissions from confined animal operations and/or fertilizer application would just continue to increase in the future. There is only so much land to farm and so much fertilizer than can be put in the soil. One of the reasons the authors see global emission increases compensating for decreases in US anthropogenic emissions is related to the choices made here for*

these species so the reasoning behind these VOC/PM/ammonia increases are important.

A new figure was added that shows the changes in global emissions (Figure 3 in the revised manuscript). For energy systems, CO_2 growth factors were used as a surrogate for CO, NH_3, and VOC. For mobile sources, NOx growth factors were used for CO, VOC, and NH_3. Non-combustion industrial emission growth factors were developed from projections of economic growth. Growth factors for non-combustion emissions from the residential and commercial sectors are linked to population growth. More text were added in paragraph 4 of Section 2.4 to make this clearer.

We agree that more discussion is required about the effect of increasing CO and VOCs. In the last paragraph of Section 2.4, we have added the following sentence: "The use of CO_2 as a surrogate for growth factors as described above means the projected CO and VOC emissions are likely too high." Text was also added in the last paragraph of Section 3.3.1 to address the effect of growing emissions of CO and VOC: "The smaller reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase in VOC and CO emissions from business-as-usual scenario of MARKAL, which uses surrogates as growth factors for CO and VOC (Loughlin et al. 2011)."

9) *Evaluation of model performance (2.6): A tighter scale for Figure 4 would make this Figure more useful for interepretation. More explanation and discussion is needed regarding the overestimated temperatures in the central and eastern U.S. These are rather large overpredictions and would seem to result in overestimated biogenic VOC. These ozone overestimates are extremely large. Mean MDAO3 in the Midwest in current conditions is 50 ppb but the modeling suggests 70 ppb! I appreciate that the authors have not tried to generate Tables and Figures to minimize our focus on the current conditions model performance but it is important to consider these issues when thinking about the results.*

The original Figure 4 is now Figure 5 and has been modified as suggested by the referee. We agree with the referee that that overprediction of temperatures and its impact on ozone should be noted. The revised manuscript includes the following statement: DM8O is be over-estimated in regions where temperature maxima are also over predicted, most noticeably in the Midwest, the South, and the Southeast but also in the Northeast."

10) *Given the large overestimation bias it would be good for the authors to show that rather than refer to a different manuscript. Also, it would be helpful to know why it is important to represent the correlation between ozone and temperature. If they are both grossly overpredicted is model performance really ok just because these variables correlate well?*

Evaluation of model performance was carried out comparing modeled values vs observations (Figure 5 which is 6 in the revised version of the manuscript). The correlation between ozone and temperature across different years is evidence that bias in temperature is the main cause of the bias in DM8O and that the air quality model is responding to the meteorological driver of ozone production and thus can predict the impact of climate change on DM8O. The text has been edited to have: “Despite the bias, results from the modeling framework presented here have been shown to accurately represent the correlation between ozone and temperature at rural CASTNET sites throughout the US (Avise et al., 2012), suggesting that the bias in temperature is a main cause of the bias in DM8O and that the chemical transport model is responding to the meteorological driver of ozone production and thus can predict the impact of climate change on DM8O.”

11) *These underpredictions for PM2.5 are very large and puzzling since ozone is grossly overestimated. Generally when a modeling system can predict a lot of ozone there would be a lot of sulfate, but the spatial plot of average current condition PM2.5 does not show the typical eastern US regional sulfate signature typical for model runs representing the early 2000s. The PM2.5 performance looks like there is something fundamentally wrong with either the CMAQ simulations or the way they were post processed.*

As mentioned previously, in the process of addressing this comment, we discovered a bug in CMAQv4.7 that resulted in NH_3 emission being read into the model incorrectly. We have re-run all the simulations using CMAQv4.7.1. Modeled to observed $PM_{2.5}$ comparison is show in Figure 7 in the revised manuscript. The $PM_{2.5}$ results are significantly improved, though PM2.5 is still underestimated in the western half of the US. We note in the revised manuscript that the causes of underprediction include the lack of wind blown dust and fire smoke emissions, underestimation of SOA formation, and overprediction of precipitation. Note that due to the lack of speciated PM data for our period of simulation, we have removed the section on model performance for

speciated PM (the observed data in the ACPD version were from 2003-2008 and we believed is not representative of our period of simulation).

12) *Comment from Referee. Despite what the authors suggest in 375-377 this type of underestimate is not typical of other regional CMAQ simulations for this time period and performance here is far worse than Foley et al and Appel et al (and probably the others but I don't have time to go back and check them all).*

Thanks to the referees' comment, we encountered a bug on CMAQ version 4.7. We re-ran the simulations with CMAQv4.7.1 and the results changed significantly. Please see comments above.

13) *There is really no value to discussing the fractional composition of PM2.5. How is that important for providing confidence in the model? If the model gets the composition right how does that make the large underestimates of all the major species ok? The fractional evaluation should be removed from the manuscript. Please just provide a comparison of speciated mass for the major species.*

We agree with the Referee. This section has been removed (see response to comment #11 above)

14) *It is very debatable the model is performing well for PM2.5. A five year summer average of 5.6 ug/m3 for total PM2.5 mass during the early 2000s is extremely small. Typically models do quite well capturing regional sulfate and since these simulations are under predicting so much it raises some questions about the model inputs, in particular the emissions.*

The text in this section has been changed after we re-ran the simulations using CMAQv4.7.1. After the simulations, PM concentrations did increase across the simulation domain with slight overprediction in the eastern half of the US. For the western US, however, $PM_{2.5}$ remain heavily underpredicted. The underprediction can be

explained by the overprediction of precipitation. The last paragraph of Section 2.6 has been edited to reflex the new simulation results.

Results section:

15) *The A1B scenario is a not a great projection of what will happen to global methane and NOX in the future so that caveat would be useful here and in other places where this is mentioned. Does the Hogrefe 2004 paper detail how much of this increase in related to methane emissions?*

In this study, methane concentration is fixed at 1.85 ppm. We have added this caveat to the manuscript; see response to comments #3 and #7. Hogrefe does not detail how much increase is due to methane.

It has been published that the emission projections considered from the SRES have been underestimated (Garnaut et al., 2008; Pozzer et al., 2012; Williams et al., 2014). But the level of uncertainty of the socio economic drivers behind each scenario was the motivation behind the SRES developers to consider each of the scenarios equally plausible. We would be thankful if the referee expands on the evidence behind his/her assertions on future emissions of NOx from the A1B scenario.

16) *Comment from Referee. Please provide more explanation about how biogenic VOCs are sequestering ozone precursors and which ozone precursors are being sequestered. NOX? Toluene? How do the authors separate the impacts of this sequestration and recycling of isoprene nitrates? It sounds like this information is available and since this might be a more novel aspect of this research I suggest the authors provide spatial plots or some way to present the relative contributions of these processes here.*

We have added Figure 14 and text in Section 3.3.3 regarding the sequestration of NOx:

> "This [DM8O] decrease is associated with a decrease in NOx concentrations (Fig. 14a). This decrease in NOx suggests that the effect of sequestration of NOx by the biogenic VOCs as organic nitrates (RNO_3) is predominant over the effect

of recycling of NOx considered in SAPRC-99, which lumps all non-PAN organic nitrates as one compound that has a NOx recycling efficiency of about 30%. This reduction in ozone is consistent with the results of Xie et al. (2013), who reported increases in NOx and ozone in the Southeast when sequestration by isoprene nitrates was reduced relative to the base SAPRC-07T mechanism that has the same RNO_3 treatment as SAPRC-99. Evidence of the predominant effect of sequestration over the recycling of NOx in the eastern US is seen in Figure 14, which shows an increase in RNO_3 and reduction in the NOx concentrations in most of the eastern US for Simulation 2 relative to Simulation 1

17) *Probably worth noting these increases are driven by global emissions changes.*

Reviewer didn't link the comment to any particular paragraph, line, table, figure or section in the manuscript.

Conclusions section:

18*) Are Asian and Mexican emissions impacts tracked separately from other countries? It would be nice to see the breakdown by country if that is available.*

We did not run additional sensitivity simulations to separate out the effects of Asian and Mexican emissions. It is something to consider for future work

19) *The biases in the model current conditions period should be mentioned here again*.

The text was added to reflect the referee's suggestion

The next line was added to the conclusion. "The evaluation of the model performance for the base case simulation shows an overestimation of DM8O and an underestimation of $PM_{2.5}$ across the projected domain"

20) Suggest changing the word “will” to “may” since this assessment is not singularly conclusive.

Referee is correct. The word “will” was changed to “may” throughout the conclusion section.

Figures:

21) Figure 1. The US only map is impossible to read with the current color scale.

We have changed the color scale of Figure 1 so that the US map is more visible.

22) *Figure 4. Please show number of stations similar to Figure 5.*

We have edited the figure according the referee’s suggestion.

23) *Figure 6. Please show absolute speciated PM2.5 predictions and observations. The fractional plot does not terribly useful. Since the authors are referencing EPA guidance I suggest showing prediction-observation pairs with shorter averaging times like either the 24-hr or monthly averages to be more consistent with that guidance and to provide more useful information.*

The plot has been added to the manuscript. See response to comment #11.

24) *Figure 11. Its not clear that any of these panels isolate the biogenic impacts. Is that on here already?*

Note that the original Figures 11 and 12 are now Figures 12 and 13. In Figures 12 and 13, panels c and d show the effects of biogenic emissions due to climate change and of biogenic emissions due to climate plus land use changes, respectively. We have edited the figure captions to be this clearer.

List of Most Relevant Changes

1. We have performed the small changes and edits in the text to reflect the suggestions posted in the interactive comments.

2. During the revision, we learned there were bugs in CMAQv4.7 in processing emissions, as described here: www.cmascenter.org/cmaq/documentation/4.7.1/RELEASE_NOTES.txt.
   Accordingly, we have re-run all simulations using CMAQv4.7.1, resulting in substantial changes to the PM2.5 section. Changes in ozone also occurred as a result of upgrading to CMAQ 4.7.1 due a combination of updates in emission, advection and plume rise emission modeling. The most notorious changes are on the modeling performance and the PM2.5 and O3 concentrations.

3. Due to the lack of speciated PM data for our period of simulation, we have removed the section on model performance for speciated PM (the observed data in the ACPD version were from 2003-2008 and we believed is not representative of our period of simulation). Figure 6 was removed and replaced for a regional performance plot of model vs observations PM2.5.

4. We have added Figure 14 and text in Section 3.3.3 regarding the sequestration of NOx:
   This [DM8O] decrease is associated with a decrease in NOx concentrations (Fig. 14a). This decrease in NOx suggests that the effect of sequestration of NOx by the biogenic VOCs as organic nitrates (RNO_3) is predominant over the effect of recycling of NOx considered in SAPRC-99, which lumps all non-PAN organic nitrates as one compound that has a NOx recycling efficiency of about 30%. This reduction in ozone is consistent with the results of Xie et al. (2013), who reported increases in NOx and ozone in the Southeast when sequestration by isoprene nitrates was reduced relative to the base SAPRC-07T mechanism that has the same RNO_3 treatment as SAPRC-99. Evidence of the predominant effect of sequestration over the recycling of NOx in the eastern US is seen in Figure 14, which shows an increase in RNO_3 and reduction in the NOx concentrations in most of the eastern US for Simulation 2 relative to Simulation 1

5. We have added to section 3.3.1: “The smaller reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase in VOC and CO emissions from business-as-usual scenario of ESP v1.0 which uses CO2 as a surrogate for growth factors for CO (Loughlin et al. 2011).”

6. We have rearranged the text in Section 3.2 for better clarity, and the sentence referred to by the referee is no longer in the text. Also, please note that the section has changed drastically.

7. As requested by the reviewer No1, we have added more discussion to the text in section 3.3.3. First, the reference was corrected to be Nolte et al. (2008) instead of Leung and Gustafson (2008). We now have "However, the difference in geographical features of DM8O changes with Nolte et al. (2008) and Tagaris et al. (2007) suggests that the source of disparities resides in the simulated regional meteorological fields resulting from different global climate models, regional climate models and the methods used to estimate emissions from biogenic sources. We used the ECHAM5 global climate model results while both Nolte et al. (2008) and Tagaris et al. (2007) used results from the GISS global climate model. For regional climate simulations, both Nolte et al. (2008) and Tagaris et al. (2007) used MM5 while we used WRF here. In contrast with Nolte et al. (2008) and Tagaris et al. (2007) who use the BEIS/BELD3 (Hanna et al., 2005; http://www.epa.gov/ttn/chief/emch/biogenic/) tool to compute biogenic emissions, this investigation estimates the biogenic emissions with MEGAN v2.04. MEGAN v2.04 generally predicts higher isoprene emissions than BEIS (Hogrefe et al., 2011; Sakulyanontvittaya et al., 2012). Hogrefe et al. (2011) shows that for the Northeast, MEGAN leads to higher DM8O by upwards of 7 ppb using 2005 anthropogenic emissions; however, under a scenario by which anthropogenic NOx emissions were reduced by ~60%, difference in DM8O was generally 3 ppb because of greater sensitivity to NOx emissions when MEGAN was used."

8. We have revised the text to explain the change in BVOC and NOx emissions under the land use change scenario instead of VOC to NOx ratio: “When land use changes are included along with biogenic emissions (Simulation 3), the increase in BVOC emissions is projected to be less, while NO emission is projected to increase in areas where natural vegetation is converted to cropland. This combination leads to higher DM8O in Simulation 3 than Simulation 2 (Simulation 3; Figure 12d).”

9. We had misplaced text on ozone in section 3.4 This error has been corrected in the revised manuscript. The line now reads as follows: “Changes in global emissions do not have a significant impact on $PM_{2.5}$ concentrations, while changes in the climate and biogenic emissions can lead to both increases and decreases in $PM_{2.5}$ depending on the region.”

10. The original Tables 2 and 5 have been replaced with tables showing percentage changes of DM8O and PM2.5. The new Tables 2 and 5 as well the original

Tables 3 and 4 now have column headings listing the simulation numbers consistent with those of Table 1.

11. A figure with the emissions from the semi-hemispheric domain was added (new Figure 3). The figure shows the dramatic increase in NOx emissions across the domain and aims at the reason behind the increase in DM8O. A line addressing a possible higher reduction in ozone as a result of updated future regional emission projections was also added to the conclusion.

12. Changes in the text of the introduction to highlight the direct effect of methane. "Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the direct effect of emissions of methane in the air quality simulations"

13. Added Cooper et al. 2010 and Cooper et al. 2012, and added references in the introduction and result section to reflect recent advances in the field.

14. Section 2.5. Added the line "Methane concentration is fixed at 1.85 ppm for all CMAQ simulations."

15. In the last paragraph of Section 2.4, we have added the following sentence: "The use of CO_2 as a surrogate for growth factors as described above means the projected CO and VOC emissions are likely too high." Text was also added in the last paragraph of Section 3.3.1 to address the effect of growing emissions of CO and VOC: "The smaller reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase in VOC and CO emissions from business-as-usual scenario of MARKAL, which uses surrogates as growth factors for CO and VOC (Loughlin et al. 2011)."

16. The original Figure 4 is now Figure 5 and has been modified as suggested by the referee. We agree with the referee that that overprediction of temperatures and its impact on ozone should be noted. The revised manuscript includes the following statement: DM8O is be over-estimated in regions where temperature maxima are also over predicted, most noticeably in the Midwest, the South, and the Southeast but also in the Northeast."

The Effects of Global Change upon United States Air Quality

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Abstract

To understand more fully the effects of global changes on ambient concentrations of ozone and particulate matter with aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) in the US, we conducted a comprehensive modeling effort to evaluate explicitly the effects of changes in climate, biogenic emissions, land use, and global/regional anthropogenic emissions on ozone and $PM_{2.5}$ concentrations and composition. Results from the ECHAM5 global climate model driven with the A1B emission scenario from the Intergovernmental Panel on Climate Change (IPCC) were downscaled using the Weather Research and Forecasting (WRF) model to provide regional meteorological fields. We developed air quality simulations using the Community Multiscale Air Quality Model (CMAQ) chemical transport model for two nested domains with 220 km and 36 km horizontal grid cell resolution for a semi-hemispheric domain and a continental United States (US) domain, respectively. The semi-hemispheric domain was used to evaluate the impact of projected ~~Asian~~ global emissions changes on US air quality. WRF meteorological fields were used to calculate current (2000s) and future (2050s) biogenic emissions using the Model of Emissions of Gases and Aerosols from Nature (MEGAN). For the semi-hemispheric domain CMAQ simulations, present-day global emissions inventories were used and projected to the 2050s based on the IPCC A1B scenario. Regional anthropogenic emissions were obtained from the US Environmental Protection Agency National Emission Inventory 2002 (EPA NEI2002) and projected to the future using the MARKet ALlocation (MARKAL) energy system model assuming a business as usual scenario that extends current decade emission regulations through 2050. Our

results suggest that daily maximum 8 hour average ozone (DM8O) concentrations will increase in a range between 2 to 12 parts per billion (ppb) across most of the continental US.~~, with t~~The highest increase occurs in the South, Central, and Midwest regions of the US~~,~~ due to increases in temperature, enhanced biogenic emissions, and changes in land use. T~~In the western US, t~~he model predicts an average increase of ~~2~~1-6 ppb in DM8O due to projected increase in global emissions ~~in Asia~~of ozone precursors~~, particularly from Asia~~. The effects of these factors are only partially offset by reductions in DM8O associated with decreasing US anthropogenic emissions. Increases in $PM_{2.5}$ levels between 4~~2~~ and 10~~4~~ μg m^{-3} in the Northeast, Southeast, Midwest and South regions are mostly a result of increase in primary anthropogenic particulate matter (PM), enhanced biogenic emissions and land use changes. ~~Little change in $PM_{2.5}$ in the Central, Northwest, and Southwest regions was found, even when PM precursors are reduced with regulatory curtailment.~~ Changes in ~~temperature, relative humidity, and~~ boundary conditions shift the composition but do not alter overall simulated $PM_{2.5}$ mass concentrations.

1. Introduction

Despite extensive efforts to reduce anthropogenic emissions, air pollution continues to be a public health issue in the United States (EPA, 2010). Elevated concentrations of pollutants in the troposphere, such as ozone (O_3) and particulate matter ~~(PM),~~, degrade air quality and have been associated with, among other things, increasing human respiratory diseases in urban areas (WHO, 2005), and in the case of PM, with low birth weights across the world (Dadvand et al., 2012).

High concentrations of tropospheric ~~ozone~~ O_3 and ~~particulate~~ PM matter with aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) are caused by a combination of adverse meteorological conditions and the atmospheric emissions of their primary precursors. While regulatory controls are expected to reduce emissions of many emitted pollutants in the United States (US) in the future, the negative effects of global climate change may offset the positive effects of such reductions. Furthermore, global emissions of greenhouse gases and other pollutant precursors are projected to increase (IPCC, 2007). Moreover, recent research has provided evidence of increasing long-range transport of ~~ozone~~ O_3 and $PM_{2.5}$ precursors from Asia and their influence over the western US. (Lelieveld and Dentener, 2000; Wuebbles et al., 2007; Cooper et al., 2010, Zhang et al., 2010; Ambrose et al., 2011; Cooper et al., 2012, WMO, 2012, Lin et al., 2012).

In the United States, regulations and technological changes in the transportation and energy sectors are projected to reduce regional atmospheric pollutants in the future (Loughlin et al., 2011). However, the interplay between climate change, increasing global

emissions, and intercontinental transport pose challenges that air quality managers will have to address in order to maintain regional air quality standards (Ravishankara et al., 2012). To provide a foundation for building effective management strategies and public policies in a changing global environment, modeling approaches that link global changes with regional air quality are required. The general approach has been to use output from general circulation models (GCMs) to drive regional climate models (RCMs) and regional or global chemical transport models (CTMs/GTMs; Giorgi and Meleux, 2007; Jacob and Winner, 2009).

This downscaling approach has been used in a variety of studies in Europe, Canada, and Asia at different time scales of climate change (e.g., Liao et al., 2006 [2000 to 2100]; Langner et al., 2005 [2000 to 2060]; Forkel and Knoche, 2006 [2990 to 2030]; Meleux et al., 2007 [1975 to 1985]; Kunkel et al., 2007 [1990 to 2090]; Lin et al., 2008 [2000 to 2100]; Spracklen et al., 2009 [2000 to 2050]; Kelly et al., 2012 [2000's to 2050's]). These investigations based the global emissions on future anthropogenic emissions scenarios developed from the Intergovernmental Panel on Climate Change (IPCC) assessment reports ~~projected to 2050's, 2080 and 2100~~. Despite the differences in emission scenarios, time scales, modeling frameworks and future climate realizations, increases in ozone concentrations on the order of 2 to 10 ppb in polluted regions were consistently predicted from these studies as a result of climate change alone. By contrast, there is little consistency among the model predictions of climate change effects on ~~particulate matter (PM)~~PM (Jacob et al, 2009; Dawson et al., 2013).

In the US, a combined effort between the Environmental Protection Agency and the academic community resulted in a set of modeling studies that adopted a variety of modeling methods (Hogrefe et al., 2004; Leung and Gustafson, 2005; Liang et al., 2006; Steiner et al., 2006; Tagaris et al., 2007; Liao et al., 2006; Racherla and Adams, 2006, 2008; Tao et al., 2007; Huang et al., 2007, 2008; Nolte et al., 2008; Wu et al, 2008a, 2008b; Chen et al, 2009b; Avise et al., 2009; Dawson et al., 2009). These US investigations based their current and future climate realizations on the results of GCMs using the various IPCC emissions scenarios (IPCC, 2007) projected to the 2050's. In some of the studies, the global climate realizations were subsequently downscaled to a higher resolution using the PSU (Pennsylvania State University)/NCAR (National Center for Atmospheric Research) Mesoscale Model version 5 (MM5; Grell et al., 1994) to horizontal resolutions that ranged from 90 km to 36 km. Many of these studies based their analysis on the effects of climate change on summer air quality in the Contiguous Continental US (CONUS). In summary, despite the differences in modeling elements, all studies found ~~an~~ increases in ~~daily~~ the summer average of the daily maximum ~~summer~~ 8-hour average ozone concentrations over large regions of ~~the simulated~~ CONUS ~~domain~~ on the order of 2 to 8 ppb ~~for the simulated CONUS domain~~ (Weaver et al., 2009)~~, but with regional variations~~. In contrast, PM concentrations showed changes between ± 0.1μg m^{-3} to ± 1μg m^{-3}, with little consistency between studies, including the sign of the differences (Day and Pandis, 2015; Trail et al., 2014; Jacob and Winner, 2009).

It is important to note that variations between modeling frameworks did result in very diverse regional patterns of key weather drivers for ozone and PM formation. Thus,

while most of the studies mentioned above ~~found~~ projected an average increase in ozone concentrations for the simulated domains, reductions or insignificant changes in certain regions of the domain were also simulated. Generally, temperature and solar radiation reaching the surface were the major meteorological drivers for regional ozone concentrations. For PM concentrations, most of the studies found a direct link between changes in precipitation and relative humidity and changes in PM concentrations (Liao et al., 2006; Unger et al., 2006; Racherla and Adams 2006, Tagaris et al., 2007; Avise et al., 2009; Chen et al., 2009b). Nevertheless, the direct impacts of changes in meteorological conditions are not the only factors of change for ozone and PM concentrations. Changes in emissions of biogenic volatile organic compounds (BVOCs), due to climate and land cover change, and the treatment of isoprene nitrates in the chemical mechanism were found to be a key factor in the regional variability of ozone and PM, particularly in areas of the southeastern US (Jacob and Winner, 2009; Weaver et al., 2009).

In this work, we present a continuation of the work described by Avise et al. (2009) and Chen et al. (2009a, b), who downscaled the Parallel Climate Model (PCM; Washington et al., 2000) and MOZART (Model for OZone And Related chemical Tracers; Horowitz, 2006) global model output for the A2 IPCC scenario using MM5 and the Community Multi-scale Air Quality Model (CMAQ; Byun and Schere, 2006) to simulate current and future air quality in the US. For this update, we implemented a semi-hemispheric domain for the Weather Research and Forecasting (WRF) mesoscale meteorological model (http://www.wrf-model.org) and CMAQ simulations in lieu of using

MOZART output for chemical boundary conditions for our CONUS CMAQ simulations. We used the ECHAM5 global climate model (Roeckner et al., 1999, 2003) output for the A1B scenario to drive these simulations for two decadal periods~~:~~, the current decade from 1995–2004 and the future decade 2045–2054. In presenting our results, we follow the attribution approach described in Avise et al. (2009), where the separate and combined effects of changes in climate, US anthropogenic emissions, global anthropogenic emissions and biogenic emissions due to changes in regional meteorology and land use are investigated. Ideally, this framework should include feedback from changes in atmospheric chemistry to the climate system (Raes et al., 2010). However, due to the computational requirements of an on-line approach, we did not incorporate feedback between the atmospheric chemistry and transport simulations from the CTM to the RCM. Furthermore, despite the observed sensitivity of tropospheric ozone to regional emissions and global burden of methane (Zhang et al., 2011; Fiore et al., 2008; Wu et al., 2008a; Nolte et al., 2008; Fiore et al., 2006), in this work, we do not address the ~~potential~~ direct ~~contribution~~ effect of emissions of methane o~~i~~n the air quality simulations.~~.~~

In Section 2, we provide an overview of the modeling framework and emissions scenarios. Evaluation of the model performance for the climate simulations and results of the changes in meteorological fields are presented in Section 3. Assessment of air quality changes and the individual and combined effects from changes in model components are presented in Section 4. Finally, we present a summary of the results and conclusions in Section 5.

2. Methodology

2.1 General Framework

Results from the global climate model ECHAM5 under the IPCC Special Report on Emissions Scenarios (SRES) A1B scenario (Nakicenovic et al., 2000) were downscaled using the WRF model separately to a semi-hemispheric (S-HEM) 220 km domain and nested CONUS domains of 108 km (not shown) and 36 km (Figure 1). Although, it has been suggested that periods of 10 to 30 years are required to fully determine climatological conditions (Andersson and Engardt, 2010), the fact that emission inventories can substantially change from one decade to the next suggests that using five to ten year periods for air quality assessment is more appropriate. Thus, five representative summers (June-July-August; with May as a spin-up period) for the present (1995 to 2004) and the future (2045 to 2054) decades were selected. Ranked in terms of their CONUS-mean maximum temperature of the year, the summers of the warmest and coldest years, as well as the second, fifth and seventh warmest years in each decade were selected for CMAQ simulations. Comparison of the meteorological conditions of these five selected~~chosen~~ summers to those of the full decades is presented in section 3.1. These five representative summers ~~(June-July-August; with May as a spin-up period)~~ for the present and future periods were processed with the Meteorology-Chemistry Interface Processor v3.4.1 (MCIP; Otte and Pleim, 2010) for the S-HEM and 36 km CONUS domains. Meteorological fields generated from MCIP for both domains were used to estimate biogenic emissions using the Model of Emissions of Gases and Aerosols from Nature v2.04 (MEGANv2.04; Guenther et al., 2006) and to calculate the temporal profiles within the Sparse Matrix Operator Kernel Emissions (SMOKE) v2.7

(http://www.smoke-model.org). With the elements described above, a framework to perform air quality simulations using the Community Multiscale Air Quality Model (CMAQ v4.7; Foley et al., 2010) was created. The overall schematic for the modeling system is shown in Figure 2.

2.2 Climate and Meteorology

The regional weather model WRF includes advanced representations of land-surface dynamics and cloud microphysics to simulate complex interactions between atmospheric processes and the land surface characteristics. Detailed descriptions of WRF can be found at http://wrf-model.org and a discussion of its range of regional climate modeling applications ~~can be found in~~is detailed by Leung et al. (2006). In this experiment, WRF was used to downscale the ECHAM5 output for both the S-HEM and 108/36 km CONUS domains. The model was applied with 31 vertical levels and a vertical resolution of ~ 40 – 100 m throughout the boundary layer with the model top fixed at 50 mb. Details of the model setup and a discussion of the results are reported by Salathé et al. (2010), Zhang et al. (2009, 2012), and Duliére (2011, 2013).

2.3 Current and Future Biogenic Emissions and Land Use Changes

The MEGANv2.04 biogenic emission model (Guenther et al., 2006, Sakulyanontvittaya et al., 2008) was used to estimate current and future biogenic VOC and soil NO_X emissions based on the WRF meteorology with current and future estimates of land use and land cover. For the current decade, the default MEGANv2.04 land cover and emission factor data (Guenther et al., 2012) were used. For the future decade, cropland distributions were estimated by combining three datasets: the IMAGE 2100 global

cropland extent dataset, (Zuidema et al., 1994), the SAGE maximum cultivable land dataset (Ramankutty et al., 2002), and the MODIS-derived current cropland data (as used in MEGANv2.04 and described in Guenther et al., 2006). The IMAGE 2100 dataset was created from the output of a land cover model, which forms part of a sub-system of the IMAGE 2.0 model of global climate change (Alcamo, 1994). The SAGE cultivable dataset was created using a 1992 global cropland dataset (Ramankutty and Foley, 1998) modified by characterizing limitations to crop growth based on both climatic and soil properties. The future global cropland extent distribution was generated by analyzing predicted changes in agriculture on a continent-by-continent basis (using the IMAGE data). These changes were then applied to the MODIS based cropland map (used for present day MEGAN simulations) using the SAGE maximum cultivable dataset as an upper limit to cropland extent. The resulting land cover data has considerably lower cropland fraction than the original IMAGE data, which likely overestimates future cropland area by not considering whether a location is cultivable.

In addition to generating a future crop cover dataset to simulate potential biogenic VOC emissions using MEGAN, future datasets representing several other MEGAN driving variables were developed. These included geo-gridded potential future plant functional type (PFT)-specific emission factor (EF) maps for isoprene and terpene compounds, as well as future-extent maps of four non-crop PFTs: broadleaf trees, needle-leaf trees, shrubs, and grasses. For regions outside of the US, the non-crop PFT distributions were generated by reducing the current extent of each non-crop PFT map by an amount that would appropriately offset the predicted cropland expansion for a given

continent. For the US, future non-crop PFT maps were generated using the Mapped Atmosphere-Plant-Soil System (MAPSS) model output (http://www.fs.fed.us/pnw/corvallis/mdr/mapss/; Neilson, 1995), based on three GCM future scenarios. Present-day MAPSS physiognomic vegetation classes were associated with current PFT fractional coverage estimates by dividing the US into sub-regions and by averaging existing (MODIS-derived) geospatially explicit PFT data within each sub-region as a function of MAPSS class. Sub-regions were created based on Ecological Regions of North America (http://www.epa.gov/wed/pages/ecoregions.htm). After every current MAPSS class had been assigned PFT-specific fractional coverage estimates, future PFT cover was determined by re-classifying future distribution maps for the three MAPSS datasets using the fractional PFT cover estimates for each MAPSS class (within each ecological region), and averaging the three resultant future datasets into a single estimate of future cover for each PFT.

For the eastern US, future isoprene and monoterpene PFT-specific EF maps were constructed using changes in tree species composition predicted by the US Department of Agriculture A 'Climate Change Tree Atlas' (CCTA, http://nrs.fs.fed.us/atlas/tree/). The CCTA data arewas based on ecosystem changes driven by the average of three GCMs, which that represented the most conservative emissions scenarios available.

Using existing speciated EF data (Guenther, 2013), we applied anticipated changes in the average species composition of each PFT to generate species-weighted PFT-specific EF maps on a state-by-state basis (the CCTA data is organized by state).

As data was lacking on predicted species-level changes for areas outside the eastern US, we did not attempt to alter EF maps outside the eastern US.

2.4 Anthropogenic Emissions

For S-HEM domain CMAQ simulations, global emissions of ozone precursors from anthropogenic, natural, and biomass burning sources were estimated for the period 1990-2000 (applied to 1995-2004) using the POET emission inventory (Granier et al., 2005). Non-US anthropogenic emissions (containing 15 sectors) were projected based on national activity data and emission factors. Gridded maps (e.g. population maps) were applied to spatially distribute the emissions within a country. The global emission inventory for black and organic carbon (BC and OC respectively) was obtained from Bond et al. (2004), which ~~uses~~ applies emission factors ~~on the basis~~based of fuel type and economic sectors alone. The Bond et al. (2004) inventory includes emissions from fossil fuels, biofuels, open burning of biomass, and urban waste. Emissions are varied by combustion practices, which consider~~Considering~~ combinations of fuel, combustion type, and emission controls, as well as their prevalence on a regional basis ~~covers the dependence of emissions on combustion practices~~.

Global emissions for the year 2000 from the POET, MEGAN, and Bond et al. (2004) inventories were combined, and the 16 gas-phase POET and MEGAN species, along with the OC and BC species were adapted to the SAPRC99 (Carter 1990, 2000) chemical mechanism. Diurnal patterns were developed and applied to the gridded emission inventories and processed using SMOKE. For the future decade hemispheric domain simulations, current decade emissions were projected to the year 2050 based on

the IPCC A1B emission scenario. ~~After the emission inventories were combined and adapted to SAPRC99 and the diurnal patterns were applied, the differences percent change in emissions between the future and the current decade wereas estimated. To aid in understanding the effects of changes in global emissions upon the US, T~~the percent change in emissions was summarized according to the regions and countries in the S-HEM domain that surround the CONUS domain (Figure 3).

~~For the 36-km CONUS current decade CMAQ simulations,~~ US anthropogenic emissions for the 36-km CONUS current decade CMAQ simulations were developed using the 2002 National Emission Inventory. The Emission Scenario Projection (ESP) methodology, version 1.0 (Loughlin et al., 2011), was applied to project future decade US anthropogenic emissions. A primary component of ESP v1.0 is the MARKet Allocation (MARKAL) energy system model (Loulou et al., 2004). MARKAL is an energy system optimization model that characterizes scenarios of the evolution of an energy system over time. In this context, the energy system extends from obtaining primary energy sources, through their transformation to useful forms, to the variety of technologies (e.g., classes of light-duty personal vehicles, heat pumps, or gas furnaces) that meet "end-use" energy demands (e.g., projected vehicle miles traveled, space heating). Within ESP 1.0, ~~the~~ MARKAL is used to develop multiplicative factors that grow energy-related emissions from a base year to a future year. Surrogates, such as projected population growth or industrial growth, are used to develop non-energy-related growth factors. The resulting factors were ~~used~~ applied within SMOKE to develop a future decade inventory from the 2002 NEI inventory.

For the work presented here, the EPAUS9r06v1.3 database (Shay et al., 2006) was used with MARKAL to develop growth factors for CO_2, NOx, SO_2 and PM with aerodynamic diameter smaller than 10 μm (PM_{10}). The PM_{10} growth factors were also applied to $PM_{2.5}$; and the CO_2 growth factors were used as a surrogate for energy system CO, NH_3, VOC, HCl and chlorine. For mobile sources, NOx growth factors were used for CO, VOC, and NH_3. Non-combustion industrial emission growth factors were developed from projections of economic growth. Growth factors for non-combustion emissions from the residential and commercial sectors are linked to population growth. The resulting energy and non-energy factors were then used within SMOKE to multiply emissions from the 2002 National Emissions Inventory (NEI) to 2050.

EPAUS9r06v1.3 originally was calibrated to mimic the fuel use projections of the U.S. Energy Information Administration's 2006 Annual Energy Outlook (AEO06; U.S. DOE, 2008). Energy demands were adjusted to account for population growth consistent with the A1B storyline. The results reflect business as usual assumptions about future environmental and energy regulations as of 2006. Thus, while electric sector emissions are capped to capture the effects of the Clean Air Interstate Rule (CAIR; US EPA, 2005), the impacts of increases in natural gas availability, the 2007 economic downturn, and the relatively new 54.5 Corporate Average Vehicle Efficiency (CAFÉ) standard (US CFR, 2011) are not reflected. More recent versions of the MARKAL database reflect these factors with expanded pollutant growth coverage and refined emission factors (U.S. EPA, 2013). The ESP v1.0, including the MARKAL database EPAUS9rv1.3, was selected here to maintain compatibility with previous and ongoing activities.

After SMOKE was used to develop a 2050 inventory, The differences between the base year and future-year US inventories were summarized at the pollutant and regional level, as are shown in (Figure 4)3. Using the ESP v1.0 methodology, emissions of NO_X and SO_2 are projected to decrease between 16% in the South and Southwest to 35% in the Northeast and Northwest. On the other hand, emissions of pollutants that were not captured endogenously in by MARKAL, such as carbon monoxide (CO), non-methane volatile organic compounds (excluding methane; NMVOCsNMVOCs) and ammonia (NH_3) are projojectected to increase in nearly all regions across the CONUS domain. The use of surrogates for growth factors as described above means the projected changes in CO and VOC emissions are likely too high. The largest increase (70%) of in emissions of CO is projected in the Midwest; this is co-located with a 70% increase combined with an increase of about 20% of NMVOC. The smallest increase of CO is projected for the South; however, the same region was projected to increase NMVOC by about 12%. The largestr increase in $PM_{2.5}$ emissions is projected in the Northwest (<20%) and tThe smallest increase (3%) of $PM_{2.5}$ is projected in the central region, which also has a projected 34% increase in NMVOC.

2.5 Air Quality Simulations

The CMAQ model version 4.7.1 was employed to simulate the potential impact of climate change on surface ozone and $PM_{2.5}$ over the CONUS at 36 km horizontal grid spacing and covering 18 vertical layers from the surface up to 100 mb. The model configuration included the use of the SAPRC99 chemical mechanism and version 5 of the aerosol module, with Secondary Organic Aerosol parameters of Carlton et al. (2010).

Methane concentration is fixed at 1.85 parts per million (ppm) for all CMAQ simulations. Stratospheric intrusion (STE) was not included in the CMAQ simulations; ~~h~~H~~H~~igh STE events are mostly relevant during the spring season, thus, lack of STE in our summer simulations is not expected to have a significant effect in our results.

Using the framework components described above, a matrix of CMAQ simulations that included changes in predicted meteorological conditions and potential emission scenarios was constructed (Table 1). For each set of simulations shown in Table 1, five representative summers were modeled. Simulation 0 represents the base case simulation, where all model inputs are set to current decade conditions. Simulation 1 is used to investigate the impact of climate change alone; ~~where~~ all model inputs are set to current decade conditions except for meteorology (biogenic emissions are not allowed to change with the future climate for this case). Simulation 2 is the same as Simulation 1, except that biogenic emissions are allowed to change with the future climate; and in Simulation 3, future land use is also incorporated into the biogenic emission estimates. Simulation 4 is used to investigate the impact of future decade US anthropogenic emissions, where all inputs are set to current decade levels except for US anthropogenic emissions. The impact of future global emissions is investigated in Simulation 5. Finally, ~~and~~ Simulation 6 represents the combined impacts of Simulations 1-5.

2.6 Evaluation of Model Performance

To aid in summarizing model results, the 36 km domain was divided geographically into 7 regions (Figure 43, lower right). Since the WRF simulations used to drive CMAQ are based on a climate realization rather than reanalysis data, a direct comparison between the modeled output and observations cannot be made. Instead, the frequency distributions of simulated and observed values are compared. For the simulated meteorological fields, daily maximum temperature, and daily precipitation are compared against a decade of summer observations (1995 to 2004) from the United States Historical Climatological Network (US-HCN; http://cdiac.ornl.gov/ftp/ushcn_daily/; Karl et al., 1990) in Figure 54. The model distributions of temperature and precipitation agree reasonably well with the observations, and provide a good representation of the regional variability of precipitation and temperature. Except for the Northwest and Southwest regions, the observed mean and maximum temperatures are slightly over predicted, with the largest overprediction in the Midwest. which is likely to result in higher emissions of ozone and PM precursors from biogenic sources.. However, fFor all analyzed regions the model successfully simulates the seasonal trend of summer temperatures, showing the observed increase in mean temperature from June to July and subsequent decrease in mean temperature from July to August (not shown).

The modeled daily maximum 8 hr ozone concentrations (DM8O) from the five representative summers (Figure 65) from the current decade CMAQ simulations (Simulation 0 in Table 1) were compared to the range of observations from the AIRNow network (http://airnow.gov/). As seen in Figure 65, DM8O tends isto be over-estimated in

regions where temperature maxima ~~is~~ are also over predicted, ~~such as the South, Midwest, Southeast and Northeast~~most noticeably in the Midwest, the South, and the Southeast but also in the Northeast. Except for the less populated Central region, DM8O shows a bias that ranges between +10 ppb (+15%) and +25 ppb (+37%) across the domain. This is consistent with previous climate downscaled results by Tagaris et al. (2007), who found a bias of +15%, and with Avise et al. (2009), who found regional biases as high as +39%. Despite the bias, results from the modeling framework presented here have been shown to accurately represent the correlation between ozone and temperature at rural Clean Air Status and Trends Network (CASTNET) ~~-~~sites throughout the US (Avise et al., 2012), suggesting that the bias in temperature is the main cause of the bias in DM8O. This implies ~~and~~ that the chemical transport model is responding to the meteorological driver of ozone production and thus can predict the impact of climate change on DM8O.~~-~~

Simulations for the current decade show a domain ~~mean~~average DM8O of 66 ± 20 ppb (standard deviation between simulated DM8O for the five summers), while the observed average at the AIRNow sites was 5~~6~~3 ± 1~~8~~9 ppb. The s~~S~~imulations successfully captured the enhanced DM8O concentrations over the major urban areas and regions with high biogenic sources (Figure 1~~0~~23~~1~~a~~, top~~). Interannual ~~V~~variability of the simulated summer~~time~~ DM8O concentrations ~~between summers~~ is on the order of 10% (not shown) in highly populated areas and ~~down to~~as little as 1% in less populated areas, with the greatest variability found in the Northeast region.

Simulated concentrations of current decade $PM_{2.5}$ ($PM_{2.5}$ with no water content, unless otherwise specified) show a five summer average of 12.056.9 ± 10.87 μg m^{-3}, compared to 14.3 ± 9.2 μg m^{-3} observed at the Speciation Trends Network (STN; US EPA, 2000). ~~Simulated $PM_{2.5}$ show the highest concentrations occurring inland of coastal regions and throughout the Northeast and Southeast (Figure 1421, top).~~

In general, the model slightly overestimates $PM_{2.5}$ in the Midwest, the Southeast, and the Northeast and significantly underestimates $PM_{2.5}$ in the western half of the US (Figure 7). Several factors contribute to the underestimation of $PM_{2.5}$ in the western US, including a lack of windblown dust and fire smoke emissions, and an underestimation of secondary organic aerosol (SOA) formation (Carlton et al., 2010; Foley et al., 2010; Appel et al., 2012; Luo et al., 2011~~). In our study, when comparing to the STN data, we found an underestimation of all species, including SO_4^{2-} and total carbon (Organic Carbon + Elemental [Black] Carbon), except for the un-speciated $PM_{2.5}$ species (also known as PM "other"). Nevertheless, when comparing the average fractional composition we found a slight overestimation of the SO_4^{2-} fraction for most regions (Figure 86, top panel). Most regions were also found to underestimate the NO_3^- and NH_4^+ fractions. Low concentrations of NH_4^+ relative to SO_4^{2-} result in a sulfate-rich system, where aerosols are dominated by aqueous phase HSO_4^- and SO_4^{2-} and have lower concentrations of $(NH_4)_2SO_4$ and NH_4NO_3 (Fountoukis and Nenes, 2007; Kim et al., 1993; Sienfield and Pandis, 2006). Further discussion of the response of the inorganic aerosol system to global changes is provided in Section 3.4.~~).~~ Another important factor that influenced the underestimation of $PM_{2.5}$ is the over-prediction of precipitation as shown in Figure 5

When compared to STN data (Figure 86, top panel), we found a large underestimation of the fraction of organic carbon in all regions, while the unspecified fraction was over-predicted. The unspecified fraction in CMAQ is composed of all the non-carbon atoms associated with the OC fraction, unspecified direct $PM_{2.5}$ emissions, and other trace species (Foley et al., 2010). The underprediction in OC reflects the uncertainties in precursor sources and the SOA formation mechanisms which have been previously documented (e.g., Carlton et al., 2010; Foley et al., 2010).

Speciated $PM_{2.5}$ model performance evaluation using mean fractional error (MFE) and mean fractional bias (MFB) statistics for the major $PM_{2.5}$ components as suggested by Boylan and Russell (2006) was performed (Figure 86, middle and bottom panels). Overall, the model has a large underestimation of organic carbon due to underestimation of SOA as dcoumented in previous studies (see compilation by Simon et al., 2012). To a less degree the model also underesimates EC, SO_4^{2-}, NO_3^-, and NH_4^+; this most likely is due to overpredicition of precipitation (Fig. 4b), but errors in other meteorological variables and emissions also contribute to the underesimation. For the unspecified component, the model meets the performance goal for 5 of the 7 regions. The majority of the speciated components show MFE and MFB within the criteria threshold for most regions. Furthermore, tThe model performance was within the criteria thresholdsthese guidelines for $PM_{2.5}$ in four of the seven regions, and only in the Central region did the model not meet these guidelines. Similarly, SO_4^{2-}, NO_3^-, NH_4^+ and unspecified fractions meet the benchmark thresholds for model performance in most regions. In terms of the unspecified fraction, the better model performance in most regions is due to the heavy contribution to

~~the total mass of the $PM_{2.5}$. For the SO_4^{2-}-NO_3^--NH_4^+ system, the values for the MFE and MFB indicate that the model performed sufficiently well in responding to the conditions that drive inorganic aerosol formation. These values increase the confidence about the response to global changes in the system. In the case of OC and EC, poor model performance was found, with concentrations largely underpredicted for all regions.~~

3. Results and Discussion

3.1 Projected Changes in Meteorology

For these types of climate change simulations, it is important to consider whether the five selected summers represent the climatological conditions for the 1995-2004 and 2045-2054 periods. To address this, we compared the regional mean temperature and total precipitation (Figure 9~~7~~8)~~)~~ as well as maximum daily insolation and mean relative humidity (not shown) for all ten summers versus the five selected summers. Based on the two sample *t*-test, except for the Northwest region, we found no statistical difference in the overall regional average conditions between the five and ten summer samples (p >0.01). ~~The effect of selecting five summers instead of ten summers for the Northwest region is explained below.~~ For the purposes of this air quality assessment, this comparison of the meteorological conditions for the five selected summers to the full ten summer set of data suggest that the five summers provide a reasonable representation of decadal summer meteorological conditions. While no statistical difference was found between the five and ten summer samples, some distinct features should~~need to~~ be highlighted: 1) For the current decade, except for~~t~~ the Southeast, t~~T~~he ~~former~~ chosen set of five summers on average ~~represents an average warmer current decade~~is slightly

warmer than the average of the ten summers ~~sample~~~~latter~~; 2) The five summers chosen for current and future decades ~~Fiver year summer~~led to a projection of cooling in the Northwest ~~average represent a decrease in future temperature in the Northwest region~~. The effects of the higher average temperature as result of the five summer sample, and the projected decrease in future temperature in the Northwest ~~region are~~~~explained~~ discussed below.

Similar to the 30 year meteorological variability assessment carried out by Andersson and Engardt (2010), the differences between current and future summer meteorological conditions, based on the five representative periods, were found to be significant at the 99% confidence level for all regions except for the Northwest. This further supports the use of five representative summers as the basis for the air quality assessment of current and future conditions.

Projected changes in selected meteorological parameters are shown in Figure 9~~10~~8. Except for some minor cooling along the Pacific coast~~, the resulted of selecting five versus ten summers~~, mean summer temperature across the continental US is projected to increase between 0.5°C and 4°C (Figure 9~~10~~8a). This increase falls within the lower bound of the warming predicted by the ensemble of 20 GCM's under the A1B emission scenario described by Christensen et al., (2007), but differs in the regional variability due to the higher resolution of our simulations. When compared to similar studies of equal resolution using a GCM (e.g. Goddard Institute for Space Studies (~~,~~GISS) GCM II') driven by the A1B IPCC emission scenario and downscaled with MM5 to 36-km resolution, our simulated temperatures show higher temperature differences between future and current

decades (Leung and Gustafson, 2005; Tagaris et al., 2007). ~~Furthermore,~~ Tagaris et al. (2007) and Leung and Gustafson (2005) predicted an average increase of between 1 and 3 ^{o}C for most of ~~the domain~~CONUS, and temperature reductions in the border states of the Central and South regions. Nevertheless, despite the differences in physical parameterizations contained in the GCMs and the driving IPCC emission scenarios that were used, similar temperature differences (2 to 4 ^{o}C) between our study and previous investigations were simulated for the Northeast and Southeast regions (Leung and Gustaffson, 2005; Tagaris et al., 2007; Avise et al., 2009).

Projected increases in solar radiation reaching the ground vary by region. A decrease in solar radiation in the Northwest that extends to the northern boundaries of the Central regions is simulated. Small changes in the Southwest, South and Midwest are also predicted, with the largest increase experienced in the Northeast and Southeast regions (Figure 9~~10~~~~8~~b). Similar results for the Northeast regions are reported ~~for~~ by previous investigations (Leung and Gustafson, 2005; Tagaris et al., 2007, and Avise et al., 2009)~~:~~. h~~H~~owever, these same investigations had higher reductions in solar radiation at the border states between the Central and South regions.

Projected changes in precipitation across the US also vary depending on the region. With the exception of the Northwest and the northern boundary of the Central region, summertime precipitation is projected to decrease between -10% and -80%. The largest decrease is projected in the Southwest region. Our results show greater precipitation reductions than those presented in Christensen et al., (2007), who ~~predicted~~ projected between a ~~-~~5 to ~~-~~15% decreases in the South and Southwest regions. Also,

previous investigations agreed with our ~~predicted~~ projected mean precipitation reductions across the domain (Figure 9~~10~~8c). In the Northwest, the modeled increase in precipitation is ~~also~~ consistent with Leung and Gustafson (2005), who projected an increase in precipitation throughout the Northwest region. In contrast, the Southeast and Northeast regions show disparities in the magnitude and the sign of the change in precipitation. While our simulations ~~predict~~ show a reduction in precipitation between ~~-~~10 to ~~-~~20%, the ensemble of 20 GCM~~'~~s in Christensen et al., (2007) ~~predicted~~ resulted in an increase between 5 to 10% across these ~~same~~ regions. The disparities~~y~~ may be a result of the differences in resolution and parameterization schemes between our study and those used for the 20 GCM~~'~~s.

Changes in relative humidity are shown in Figure 9~~10~~8d. Relative humidity is ~~predicted~~ shown to decrease in most of the domain except for the regions where decreases in solar radiation were projected. The greater decrease in relative humidity occurs in the Southwest and Central regions of the domain, and the largest increase is observed in the Northwest region.

3.2 Changes in Biogenic Emissions

Average summertime isoprene emissions over five summers of simulation for each decade are shown in Figure 10a. ~~As expected, i~~Isoprene emissions occur at relatively high rates (>50 metric tons/day) in the eastern US and at much lower rates in the western US (<10 metric tons/day). Under future climate conditions and current land use, isoprene and monoterpene emissions are projected to increase in all regions except for the Northwest (Figs. 4 and 10b); this follows the spatial pattern of projected temperature

changes (Fig. 9a).– The most noticeable increases occur in the Northeast and Southeast regions. The model projects bigger percentage increase in monoterpenes than isoprene across the domain; however, total isoprene emission is an order of magnitude higher and thus dominates the changes in total BVOC. The increase in total BVOC ranges between 17% and 45%. The only region that is projected to have reduced total BVOC emissions is the Northwest, where~~, despite the increase in monoterpenes,~~ the model simulates a 7% reduction in isoprene emissions (Figure ~~4~~3)~~ that in absolute amount is greater than the 20% increase in simulated monoterpene emissionsis simulated~~. The reduction in isoprene emissions in the Northwest is a result of the decrease in temperatures in ~~areas~~ the coastal area where the higher isoprene emissions are encountered (Figure 9~~10~~8a).

~~Furthermore, despite~~projects ~~having t~~the ~~biggerhe percentage biggest increases in monoterpenes than isoprene across the USdomain, s arethe latter still drives the absolute change in the Central and South regions, the larger increase in isoprene for the Midwest, followed by the Northeast, Southeast, South, Central and Southwest regions, drives the increase in total BVOC in emissions of BVOC. The increase in BVOC ranges between 17% and 45%.~~ Previous investigations (Liao et al., 2006, Nolte et al., 2008) show the greatest increase in BVOC emissions in the Southeast region (10-50%). Similarly, Leung and Gustafson (2005) predicted the greatest increase in BVOC in the Southeast, but did not show any significant changes in the Northwest region.

~~Average summertime isoprene emissions over five summers of simulation for each decade are shown in Figure 1019a. As expected, isoprene emissions occur at relatively high rates (>50 metric tons/day) in the eastern US and at much lower rates in the western~~

US (<10 metric tons/day). When the emissions are projected to future climate conditions with current land use distributions, isoprene emissions are projected to increase across the domain (average increase of about 30%; Figure 1019b) with the most noticeable increases occurring in the Northeast and Southeast regions. However, wWhen future climate is combined with future land use to project biogenic emissions, there are still increases in the eastern US, but the spatial extent of isoprene emission the increase is reduced, reflecting the expansion of low isoprene-emitting croplands into regions of high isoprene-emitting deciduous forests. In this case, the domain-average increase was approximately 12% of current decade emissions, compared with a 25% increase when changes in land use are not included (Figure 1120a). Thus, future expansion of cropland and subsequent reduction of broadleaf forested lands are projected to lessen the overall increase in US isoprene emissions that result from a warmer climate. When the future decade meteorology is combined with future land use, an increase of over 100% of current decade monoterpene emissions is predicted (Figureg. 11b). The growth is most noticeable in the Central, South and Midwest regions. Also, an overall increase between 25% and 50% for the western and eastern regions is predictedsimulated. This limited increase is primarily driven by the projected changes in land use predicted for those regions.

Future monoterpene emission estimates increase because of higher temperatures across the domain. Since the version of MEGAN used in this work does not include the suppression of isoprene emissions due to elevated concentrations of CO_2 (Rosenstiel et al., 2003; Heald et al., 2009), the future estimates in this study are likely to be an upper

bound on isoprene emissions, and it is likely that future isoprene emissions will be lower than predicted by this work. Monoterpene emissions from US landscapes are not expected to be suppressed by increasing CO_2 and so are not impacted by omitting this process.

~~When the future decade meteorology is combined with future land use (Figure 1120b), an increase of over 100% of current decade monoterpene emissions is predicted. The growth is most noticeable in the Central, South and Midwest regions. Also, an overall increase between 25% and 50% for the Western and Eastern regions is predicted. This limited increase is primarily driven by the projected changes in land use predicted for those regions.~~

3.3 Effects of Global Changes upon Ozone Concentrations

Results for how the various global changes affect summertime DM8O are summarized in Table 2 ~~3~~2 and Figure 123~~1~~. Simulations for the future decade (Simulation 6) show a domain average of 48 ± 11 ppb with higher DM8O ~~across the domain~~in the Northwest, Central and South regions than the current decade simulation (Simulation 0) ~~with a domain average of 51 ± 10 ppb~~. In general, increases in DM8O are due to growing global anthropogenic emissions and climate change, while decreasing US emissions reduce DM8O. Changes in biogenic emissions as a result of a changing climate and land use have less of an influence on DM8O than ~~intercontinental transport~~increase of global anthropogenic emissions~~; the influence can be either positive or negative depending on the region~~. ~~These various~~ Ffactors that influence future DMO3 are discussed in the following sections.

3.3.1 Contributions from Changes in Global and ~~Regional~~ US Anthropogenic Emissions

The effects of increased long-range transport of global emissions ~~from Asia and Mexico~~ are shown in Figure 1~~23~~1f. The changes in chemical boundary conditions (the difference between Simulations 0 and 5 ~~in Table 1~~) increase DM8O between 2~~1~~2 to 6~~5~~6 ppb across the CONUS domain. The general west-to-east ~~-~~and south-to-north gradient of the change in DM8O reflects intercontinental and regional transport of ozone and its precursors from the west and from Mexico at the south. The greatest impact ~~is predicted~~occurs in the South (6ppb) ~~(6 ppb)~~ and Southwest (5 ppb) ~~(4 ppb)~~ regions. These results show a smaller influence in DM8O from the intercontinental transport than the simulations presented in ~~are consistent with~~ Avise et al., (2009), who ~~show~~reported ~~s~~ increases between 3 and 6 ppb of DM8O across the domain, with the greatest increase in the Southwest and South regions. The higher effect from intercontinental transport presented in Avise et al., (2009) is due to ~~higher~~ larger increases ~~in~~NOx emissions ~~of NOx~~ from global anthropogenic sources under the SRES A2 emission scenario. The effects of future global emissions and intercontinental transport of ozone precursors in the continental US have also been investigated by Hogrefe et al. (2004), who predicted an increase of 5 ppb in the Northeast region under the SRES A2 ~~IPCC~~ emission scenario.

Changes in regional US emissions of ozone precursors (difference between Simulations 0 and 4) reduce DM8O concentrations between 2 and 15 ppb ~~across the domain~~in most of eastern US, most of western US, and Texas. Projected increases in ozone in urban areas near the coasts are mainly due to the limited representation of the heavy duty, shipping and rail sectors on the ESPv1.0 (Loughlin et al., ~~–~~ 2012) by which

local steady or increase in emissions of NOx and VOCs in ports are the main cause of increase in ozone in those urban areas. Regionally, l~~L~~arger reductions are observed in the Southeast (-3%) and Southwest (-3.5%) regions with a reduction of 5 ppb and the Northeast (-2.3%) and South (-0.9%) regions with a reduction of 3~~.~~ ppb (Figure 12e, Table 2). Similar results are shown in Nolte et al., (2008) and Tagaris et al., (2007) despite a difference in the magnitude of projected emissions reductions. Tagaris et al., (2007) simulated similar ozone reductions (about 9%), with a higher nationwide reduction of 51% in NO_X emissions and a slight increase (about 2%) in VOC emissions from ~~A1B~~ projections based on the Clean Air Interstate Rule (CAIR) emission inventory. Nolte et al., (2008) showed a decrease in ozone across the domain (-12 to -16 ppb) as a result of projected reductions of 45% for NO_X and 21% for VOC emissions from the NEI 2002, following the SRES A1B ~~IPCC~~ emission scenario. In contrast, our future simulations included a 38~~21~~% reduction in NO_X emissions and a slight increase (about 2%) in VOC emissions. Avise et al., (2009) predicted an average contribution of +3 ppb across the domain as a result of projecting the NEI 1999 (NEI-1999) with the Economic Growth Analysis System (EGAS) and the SRES A2 ~~IPCC~~ emission scenario; increasing emissions by 5% for NO_X and 50% for VOCs in the future. The smaller~~lower~~ reduction in ozone concentrations between the future and the current decade in comparison to Nolte et al., (2008) is likely to be a consequence of the increase in VOC and CO emissions from the ~~resulted of the current version of the MARKAL database~~business-as-usual scenario of MARKAL, which, as explained in section 2.4, uses ~~CO_2 as a~~ diverse surrogates for growth factors for CO and VOC (Loughlin et al. 2011).

3.3.2 Contributions from Changes in Meteorological Fields

Figure 1231b shows the difference between simulations that include changes in meteorological conditions (without the effect of biogenic emissions or land use) and the current decade base case (Simulations 0 and 1). The local greater reductions in DM8O concentrations in the Northwest and Southwest regions resulted from an increase in cloud cover and lower solar radiation reaching the ground, and which causeresulting in aa reduction in photochemistry due to lower solar radiation reaching the ground (Figure 9108b), similar to the results of Jacob and Winner, 2009. Nevertheless,For other regions, increases in DM8O concentrations were projected (+5 ppb) because of increases in temperature had a greater impact on the ozone chemistryand solar insolation; this is particularly evident in the Central, Midwest, Northeast and Southeast regionseastern half of the US.

3.3.3 Contributions from Changes in Biogenic Emissions and Future Land Use

When biogenic emissions are allowed to change with the future meteorology, an average increase of DM8O with respect to the current decade base case simulations is predicted (Simulations 0 and 32). Increases of as much as 7 ppb in DM8O concentrations are mainly predicted in areas with substantial biogenic sources (Figure 1231c). Similar results are shown by Leung and Gustafson Nolte et al. (20085) and Tagaris et al. (2007),); both predicted an increase of DM8O above 5 ppb in the east coast. Simulated reductions between 2 to 4 ppb of DM8O in the coastal areas of the western regions are probably due to cooler temperatures and reduced solar insolation (Fig.ure 9a, b)increased cloud cover. Minor changes in DM8O concentrations are shown over the Southwest and Northwest regions. This is in agreement with Avise et al. (2009) and Nolte et al., Leung

and Gustafson (20085) who predicted reductions in DM8O concentrations from 1 to 4 ppb in the western regions, while Tagaris et al. (2007) also predicted similar reductions in ozone in the Central and Midwest regions. The disparities between this investigation and Avise et al. (2009) are reasonable due to the different climate realizations used (A2 vs. A1B; Storyline in scenario A2 consider higher emissions of CO_2 by 2050 than the scenario A1B). However, , the difference in geographical features of ozone DM8O changes with Leung and GustafsonNolte et al. (20085) and Tagaris et al. (2007) suggests that the source of disparities resides in both: the simulated regional meteorological fields resulted of the different global climate models, modeling systems, as discussed by Weaver et al. (2009);regional climate models both the climate realization and the methods used to estimate emissions from biogenic sources. We used the ECHAM5 global climate model results while both Nolte et al. (2008) and Tagaris et al. (2007) used results from the GISS global climate model. For regional climate simulations, we used WRF; while both Nolte et al. (2008) and Tagaris et al. (2007) used MM5 while we used WRF here. In contrast with Nolte et al. (2008) and Tagaris et al. (2007) who use the BEIS/BELD3 (Hanna et al., 2005; http://www.epa.gov/chief/emch/biogenic/) tool to compute biogenic emissions, this investigation estimates the biogenic emissions with MEGAN v2.04., whichMEGAN v2.04, which generally predicts higher isoprene emissonsemissions than BEIS (Hogrefe et al., 2011; Sakulyanontvittaya et al., 2012). is known to produceHogrefe et al. (2011) showed that for the Northeast using MEGAN leads to higher DM8O in the Northeast by upwards of 7 ppb for under the scenario of 2005 anthropogenic emissions; however, underfor a scenario by which anthropogenic NOx emissions were reduced by ~60%, difference in

DM8O was generally 3 ppb ~~because of~~due to greater sensitivity to NOx emissions when MEGAN was used. ~~higher concentrations of ozone than BEIS (Hogrefe et al., 2011)~~

When the results from Simulation 2 (Fig. 12c) are compared to the climate-only simulations (Simulation 1, Fig. 12~~1~~b), our results suggest that changes in the meteorological fields are the main driver of DM8O enhancement in Simulations 2 and 3 (Fig. 12~~1~~ c and d) across the domain. Even though BVOC emissions are higher in Simulation 2 relative to Simulation 1, Simulation 2 resulted in 2 to 4 ppb lower DM8O in the Southeast. This decrease is associated with a ~~decrease~~reduction in NOx concentrations (Fig. 14a)~~.~~. This decrease in NOx~~The change in biogenic emissions leads to an increase in the VOC concentrations to NOx ratio relative to the climate-only (Simulation 1). The~~is decrease between the Simulation 2 and Simulation 1 in our simulated DM8O~~ suggests that the effect of sequestration of ~~ozone precursors~~NOx by the biogenic VOCs as organic nitrates (RNO_3) ~~-~~is predominant over the effect of recycling of NOx ~~organic~~isoprene nitrates (RNO_3)~~-~~considered in SAPRC-99, which lumps all non-PAN organic nitrates as one compound that has a NOx recycling efficiency of about 30%.~~-~~; ~~A similar effect was reported by~~The simulated~~is~~ reduction in ozone is consistent with the results of Xie~~,~~ et al. (~~2012~~2013), who ~~simulated~~ reported ~~an~~ increases in NOx and ~~of 2 ppb of~~ ozone in the Southeast when sequestration by isoprene nitrates was reduced ~~in the~~relative to the ~~chemical mechanism~~base SAPRC-07T mechanism that has the same RNO_3 treatment as SAPRC-99. Evidence of the predominant ~~-~~effect of sequestration over the recycling of ~~organic nitrates~~NOx in the ~~Southeast~~eastern US is ~~shown~~seen in Figure 1~~4~~5, ~~where~~which shows~~a~~ an increase in RNO_3 and reduction in the NO_X concentrations

(Figure 145a) and the consequent decrease in the NOx to RNO3 ratio (RNO3; Figure 145b) in most of the eastern US in thefor sSimulation 2 relative to sSimulation 1 is observed. Furthermore, wWhen land use changes are included along with biogenic emissions (Simulation 3; Figure 11d), the increase in BVOC emissions is projected to be less whileto NO_x emission is projected to increase in areas where natural vegetation is converted to cropland. This combination leads to ratio is reduced and less depletion inhigher DM8O is simulatedin Simulation 3 than, thus, higher concentrations of DM8O than the Simulation 2 (Simulation 3; Figure 1231d) are also observed. This lower VOC to NO_x ratio is due to the increase in soil NO and the reduction of BVOC emissions associated with the land use change from natural vegetation to cropland.

3.3.4 Contributions from Combined Global Change to Future Changes in DM8O Concentrations

When the combined global changes are considered (Simulation 6), DM8O is projected to increase in all regions, except with local reductions along the western coastlinesin in the Northwest, Central and Southnearly all regions except along the western and eastern coastlines and inland areas of those regions. Increases of DM8O between 14 to 3712 ppb in the NorthwestSouth, Southwest and Central and MidwesNortheast regions are shown along with a local reductions increase of 1 to 634 ppb in parts of the South, Midwest and Centralwest and Northwest Midwest regions (Figure 1231g). The increase in DM8O is mostly due to an increase in global emissions of ozone precursors from the semi-hemispheric domain, which contributes to an increase of 2-6 ppb under current climate conditions (Figure 1231f). The other contributing factors to increasing DM8O are a combination of changed meteorological changesy (Figure

1231b) and higher BVOC emissions (with current and future land use; Figure 1231c, d). Reductions in DM8O in the urban areas resulted generally from reductions in ozone precursors from regional anthropogenic sources (Figure 1231e). However, in the western regions, lower DM8O are the result of a combination of favorable meteorological conditions (e.g. reduction in temperature and solar radiation reaching the ground) and reductions in regional ozone precursors.

3.4 Effects of Global Changes upon $PM_{2.5}$ Concentrations

Results for how the various global changes affect $PM_{2.5}$ concentrations and composition are summarized in Tables 34-56 and Figure 134. Overall, projected increase in US anthropogenic emissions have the largest impact on $PM_{2.5}$, with leading to an increase in concentrations in all regions. Changes in global emissions do not have a significant impact on $PM_{2.5}$ concentrations, while changes in the climate and biogenic emissions can lead to both increases and decreases in $PM_{2.5}$ depending on the region.

3.4.1 Contribution to $PM_{2.5}$ Concentrations from Changes in Global and Regional Anthropogenic Emissions

The results from this study are similar to those reported bySimilar results are shown in Avise et al. (2009), who predicted a change in PM2.5 of less than 1 μg m^{-3} as a result of changes in future chemical boundary conditions. In our simulation, the highest increase in $PM_{2.5}$ concentrations is found in the South region (< 1%). This increase in the South region is indicative of the effects of increased emissions from Mexico (Figure 13f). Due to the relatively short atmospheric lifetime of PM, the effects from long-range transport and increasing global emissions on US $PM_{2.5}$ concentrations are relatively small in comparison. to the current decade $PM_{2.5}$ concentrations$PM_{2.5}$ concentrations increase

~~by 8-10% in the South, Southwest, Central, and Northwest regions, with a south to north gradient indicative of the effects of increased emissions from Mexico (Figure 12f). Similar results are shown in Avise et al. (2009), who predicted a change of less than 1 μg m^{-3} as a result of changes in future chemical boundary conditions. However, w~~When the chemical composition is analyzed, Table 3~~simulations~~ shows an increase in aerosol nitrate (NO_3^-) in nearly all regions except for the South ~~(Table 33)~~; these increases are less than 0.1 μg m^{-3}, ~~as a result of increased global NH_3 emissions. In contrast,~~similar to the results of Avise et al. (2009) ~~predicted no change in NO_3^- for the same regions~~. ~~Furthermore,~~In our simulation, increases between 3% and 8% in SO_4^2 and NH_4^+ in the Southwest, Central and South regions are mostly a result of increase in emissions of SO_2 and NH_3 from Mexico~~,~~. Similarly, Avise et al. (2009) showed higher future concentrations (by 7% to 25%) of SO_4^{2-} for the same regions resulting from higher global SO_2 emissions. In our simulations, c~~C~~hanges in global anthropogenic emissions cause reductions in SOA in the Southwest, Central, South, regions and ~~an~~ increases in the Northwest, Midwest, Southeast and Northeast Regions (Table ~~4~~4); ~~However,~~ the simulated changes in SOA are very small ~~–~~ (< 1.3% and < 0.05 μg m^{-3}) and the variation ~~is probably~~may be due to small differences in modeled OH radical concentrations.

In the US, reductions in regional SO_2 and NO_X emissions from regulatory curtailment on electricity generation are offset by the projected increase ~~and further speciation of primary~~in emissions of $PM_{2.5}$ and NH_3 from other sources~~,~~ thus, resulting in an overall increase in $PM_{2.5}$ concentrations between 1 and 4 μg m^{-3} across ~~the nation~~CONUS. Similarly, Avise et al. (2009) predicted an average increase of 3 μg m^{-3}

780 across the domain but as a result of increasing NO_X and SO_2 from anthropogenic sources.
781 The greatest increase, between 2 to 4 μg m^{-3}, is found in the urban areas across the
782 Northwest, Northeast, Midwest and Southeast region (Figure 132e) as a result of increase
783 in primary emissions of $PM_{2.5}$. Similarly, Trail et al., (2015) find an increase in $PM_{2.5}$
784 concentrations between 1 and 2 μg m^{-3} as a result of an scenario that consider changes
785 in fuel use. In contrast, Tagaris et al. (2007), predicted a decrease of 23% as a result of
786 decreasing emissions. Increase in SOA concentrations is also resulted from higher
787 emissions of NMVOC and an increase in primary organic aerosol from anthropogenic
788 sources in the US. (Table 4).

789 In terms of the inorganic $PM_{2.5}$ components, reductions in SO_2 and NO_X emissions
790 in the US are offset by higher emissions of primary SO_4^{2-} sulfate and nitrate and ammonia
791 NO_3^- and NH_3, leading to an increase in both sulfate and ammoniumaerosol in the form
792 of ammonium sulfate and ammonium bisulfate. When cCompared to Tagaris et al. (2007),
793 our investigation shows no reduction in SO_4^{2-} sulfate concentrations as a result of smaller
794 reduction in SO_2 emissions from anthropogenic sources. Furthermore, similar to
795 Shimadera et al., (2013) the increase in NO_3^- nitrate concentrations in the form of
796 ammonium nitrate is highly dependent on the increase in NH_3 emissions and insensitive
797 less sensitive to changes in emissions of NO_X.

798 **3.4.2 Contribution to PM2.5 concentrations from global climate change alone**

799 Despite the effect of precipitation on PM loading, as it washes out the precursors
800 and the existing PM from the atmosphere (Seinfeld and Pandis, 2006), the effect of
801 climate change alone (with no change to biogenic emissions) on total $PM_{2.5}$

concentrations over land is a change of less than 1 μg m^{-3} ~~-~~(Figure 1~~2~~3b). However, the change in $PM_{2.5}$ composition due to climate change is highly variable and depends on changes in temperature, relative humidity and precipitation. Increases in reaction rate constants of SO_2 and higher oxidant concentrations from increased temperature and solar insolation lead to an increase in aerosol sulfate formed ~~and thus are correlated with changes in SO_4^{2-} concentrations~~ (Dawson et al., 2007). ~~—~~ Relative humidity and temperature affect the thermodynamic equilibrium of SO_4^{2-}-NH_4^+-NO_3^-, especially the partitioning of HNO_3 between the gas and particulate phases.

For all regions, sulfate concentrations are predicted to increase by 3-10%. Except for the Northwest regions, this change in concentrations is consistent with decreased precipitation, which reduces wet deposition, and~~;~~ increases in temperature and solar insolation, which increase radical production rates and increases the oxidation of SO_2 to produce aerosol sulfate. The same increase in temperature leads to nitrate being more volatile and thus decreases aerosol nitrate concentrations in regions where sulfate concentrations are predicted to increase. For the same regions where ~~SO_4^{2-}~~sulfate is projected to increase, higher concentrations of radicals also lead to higher oxidation of VOC, thus increasing SOA concentrations in the same regions.

While increasing precipitation is generally associated with decreasing $PM_{2.5}$, results here for the Northwest region showed an increase in $PM_{2.5}$ despite an increase in precipitation (Figure 1~~3~~2b). This suggests the effects of slightly colder temperature and higher relative humidity in this region, leading to an enhanced formation of ~~$(NH_4)NO_3$~~ammonium nitrate (Table 3). Furthermore, the increase in relative humidity in

the Northwest and the coastal areas of the Southwest regions leads to the increase in production of sulfate aerosol via aqueous reaction (Luo et al., 2011). Higher concentrations of ammonium nitrate~~(NH4)NO3~~ and~~,~~ ~~in addition to~~ higher concentrations of SOA (Table ~~4~~4) indicate increased aerosol formation~~, appear~~ ~~to~~ dominate over the effect of precipitation.

3.4.3 Contribution to $PM_{2.5}$ concentrations from changes in biogenic emissions and future land use

Simulations that consider projected climate change as well as the associated change in biogenic emissions (Simulation 2) show an increase in $PM_{2.5}$ between 0.5 and 3 μg m^{-3}~~.~~~~;~~ ~~These~~ these changes are mainly reflected in areas with high biogenic sources (Figure 1~~3~~2c). When the effects of future land use are considered (Simulation 3), an increase in the geographical extent of $PM_{2.5}$ is observed in comparison to the climate and biogenic emissions case, and higher increases (up to 6 μg m^{-3}) of $PM_{2.5}$ are predicted in parts of the South, ~~Southwest~~Southeast, Midwest and Northeast regions (Figure 1~~2~~3d). This is primarily due to the increase in emissions of sesquiterpenes (not shown) and monoterpenes (Figure 1~~0~~b), leading to more SOA being formed.

In terms of the inorganic components of $PM_{2.5}$, the effect of climate change is still the predominant factor for the change in ~~SO_4^{2-}~~ sulfate concentrations for the Central, South, Midwest and Southeast regions (Table 3). The ~~lessen~~ smaller increase or absolute reduction in ~~SO_4^{2-}~~ sulfate in comparison to the climate-only case is due to the competition between BVOC and SO_2 for the availability of OH, which is an oxidant for both. Additionally, a ~~lessen~~smaller decrease in NO_3^- in most of the domain and increase in the Northwest is predicted due to changes in the availability of OH as a result of the changes

in emissions of BVOC and soil NO. The increase in availability of OH and increase in soil NO emissions lead to higher formation of aAmmonium nNitrate in Ssimulations 2 and 3 than in Ssimulation 1.

SOA concentrations are predicted to increase as a result of higher emissions of BVOC across the domain (Table 4). Furthermore, when climate change and biogenic emissions are combined with future land use, concentrations of SOA are predicted to increase up to 121% in the Central region and up to 188% in the Southeast due to increased biogenic monoterpene and sesquiterpene emissions (not shown).

3.4.4 Changes in Precursors and $PM_{2.5}$ Concentrations from the Combined Global Changes

Table 5 shows the summary of changes to $PM_{2.5}$ ~~as a result~~for all Simulations ~~of the individual and combined global changes presented above~~. The differences in $PM_{2.5}$ between the future decade and current decade base case are greater in the eastern half of the US compared to the western half. In the eastern half of the US, the largest increases in $PM_{2.5}$ occur in the Southeast. Our results show that the 2 to 10 μg m^{-3} increase in $PM_{2.5}$ in the Southeast region is dominated by higher concentrations of SOA due to increased biogenic emissions as a result of climate change (Figure 1~~2~~3c), changes in land use (Figure 1~~3~~2d; Table 4) and increase in anthropogenic emissions (Figure 1~~3~~2e). Table 5 indicates that the combination of climate change, biogenic emissions and land use, and increase in anthropogenic emissions increases the concentrations of $PM_{2.5}$ between 27 and 78 % depending on the region.

4. Conclusions

We have investigated the individual and combined contributions of factors that impact US air quality by dynamically downscaling future climate projections using the WRF model and using the regional chemical transport model CMAQ version 4.7.1. Decreases in future US anthropogenic ozone precursor emissions are the only consistently ~~positive~~ beneficial influence that improves the ~~ozone concentrations~~air quality in the US and updated assumptions to generate scenarios of future US anthropogenic emissions may ~~improve such influence~~show even more positive influence. However that positive influence is offset by 1) ~~changes in long range transport and~~ increasing global emissions and changes in long range transport, which have a negative impact on air quality across the domain; 2) climate changes (namely, increased temperatures and solar radiation), which increase ozone concentrations in the Central, South, Midwest, Northeast and Southeast regions of the domain; and 3) increases in US BVOC emissions, which also increase ozone concentrations in regions with high biogenic emissions such as the South, Midwest, Northeast and Southeast.

In the case of the overall concentrations of $PM_{2.5}$, our results indicate that the effects of increasing biogenic emissions in addition to increase primary PM from anthropogenic sources have an overall negative impact on air quality by increasing $PM_{2.5}$ concentrations between 27 to 78%. In terms of the $PM_{2.5}$ composition, we show a regionally dependent mixture of inorganic aerosols and SOA. For the case of the Southeast, our findings indicate that increases in BVOC may result in higher concentrations of $PM_{2.5}$. This effect extends to the Midwest and Northeast regions due to

changes in land use. Furthermore, meteorological changes or regulatory curtailment, as incorporated in these simulations do not offset the increasing concentrations of primary PM and BVOC. In addition, synergistic effects of changes in meteorological parameters and changes in emission may shift the composition of the inorganic fraction of $PM_{2.5}$ in the ~~western~~ US. The synergistic effects of increase of ~~SO_4^-~~ sulfate and SOA in the urban areas of the coastal regions of the Northwest and Southwest lead~~s~~ to an increase in $PM_{2.5}$ in those regions, off-setting decreases due to increased precipitation and temperature, and reduced primary anthropogenic emissions of SO_2 and NO_X.

In conclusion, the results of this study suggest that the efforts to improve air quality through low emission technologies and public policy directed to the electricity generation sector may not have a major effect if future emissions ~~of primary PM~~from other sectors ~~are~~ are allowed to increase. In addition, higher global anthropogenic emissions, a warmer future world and the effects of these changes on emissions from biogenic sources may increasingly undermine all regulatory efforts. Consequently, additional measures may be necessary to improve air quality in the US.

Much of the modeling components used for this research carry different levels of complexity and have reached diverse stages of development, thus, subsequent research intended to assess the effect of climate change and future regional emissions upon air quality would benefit from newer versions of the emission inventories (e.g. 2011); updated assumptions on the US emission projections (e.g. New versions of MARKAL with the use of the ESP 2.0 methodology); newer versions of MEGAN that take into account the isoprene emission suppression due to CO_2 concentrations and more realistic estimates

of land use change; and the inclusion of emissions from wildfires and the consequent effect upon air quality.

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Table 1. List of simulations to assess the effect of global climate changes upon air quality in the United States

	Climate	Biogenic Emissions		Anthropogenic Emissions	
		Climate	Land Use	US	Global
0	Current	Current	Current	Current	Current
1	**Future**	Current	Current	Current	Current
2	**Future**	**Future**	Current	Current	Current
3	**Future**	**Future**	**Future**	Current	Current
4	Current	Current	Current	**Future**	Current
5	Current	Current	Current	Current	**Future**
6	**Future**	**Future**	**Future**	**Future**	**Future**

Table 2. Percent change in DM8O between each future scenario and the current decade base case.

Region	Climate (1)	BVOC (2)	BVOC Future Land Use (3)	US emissions (4)	Boundary Conditions (5)	Combined (6)
DM8O						
Northwest	0.4	-1.0	-0.8	-0.6	8.1	4.5
Southwest	2.0	0.4	0.0	-3.5	9.1	4.2
Central	5.6	4.5	4.9	-0.1	8.9	12.3
South	6.2	4.3	6.1	-0.9	9.6	13.0
Midwest	7.6	7.2	8.5	0.0	2.6	10.0
Northeast	8.2	6.6	7.6	-2.3	1.4	5.3
Southeast	8.6	6.1	7.7	-3.0	3.3	6.1

Table ~~3~~34. Percent change in the aerosol NH_4^+, SO_4^{2-} and NO_3^- between each future scenario and the current decade base case. The corresponding simulation number for each sensitivity simulation is shown i~~s~~n parenthesis

Region	Climate (1)	Climate & BVOC (2)	Climate, BVOC, Land Use (3)	US Anthropogenic Emissions (4)	Boundary Conditions (5)	Combined (6)
			NH_4^+			
Northwest	15.7	-0.6	-0.9	12.8	-0.2	12.2
Southwest	3.4	-8.8	-7.9	4.2	8.2	4.8
Central	12.5	2.1	2.7	6.9	3.3	14.8
South	9.1	4.3	5.8	7.5	4.8	22.9
Midwest	5.1	0.6	3.3	12.2	0.4	18.1
Northeast	1.8	-5.0	-4.2	17.5	-0.3	12.7
Southeast	10.0	5.0	4.8	12.4	0.5	21.3
			SO_4^{2-}			
Northwest	10.0	-5.4	-5.3	6.3	0.9	1.6
Southwest	5.4	-4.6	-4.0	0.7	6.2	2.8
Central	10.9	1.5	2.0	3.4	3.2	10.1
South	7.3	3.7	4.7	1.5	4.8	14.5
Midwest	7.2	1.8	4.1	2.7	0.4	10.9
Northeast	3.5	-4.0	-3.2	3.2	-0.2	2.3
Southeast	8.8	3.5	2.9	1.9	0.8	9.3
			NO_3^-			
Northwest	-0.3	2.3	0.9	20.3	6.4	27.4
Southwest	-10.1	-8.0	-7.3	11.8	8.2	12.7
Central	-34.4	-17.1	-12.0	87.6	2.6	68.4
South	-7.0	-18.7	-11.5	38.5	-2.0	17.0
Midwest	-38.4	-31.1	-23.6	96.6	2.6	56.4
Northeast	-43.9	-43.2	-42.1	74.0	2.0	4.8
Southeast	-29.4	-28.7	-28.7	54.6	7.5	19.6

Table 4 ~~45~~ Percent change of ~~secondary organic aerosol~~SOA ~~and primary organic carbon~~ between each future scenario and the current decade base case. The corresponding simulation number for each sensitivity simulation is shown i~~s~~n parenthesis

Region	Climate (1)	Climate & BVOC (2)	Climate BVOC & Land Use (3)	US Anthropogenic Emissions (4)	Boundary Conditions (5)	Combined (6)
SOA						
Northwest	11.6	17.5	40.7	17.4	1.3	61.1
Southwest	2.2	20.3	31.9	10.2	-0.2	41.0
Central	16.2	43.9	118.6	7.1	-0.2	126.4
South	4.7	57.0	113.2	7.4	-0.4	121.3
Midwest	16.0	48.6	121.2	7.9	0.1	131.0
Northeast	17.9	59.5	108.8	9.8	0.2	119.1
Southeast	14.2	73.2	135.1	8.1	0.3	143.5

Table 5 Percent change of $PM_{2.5}$ between each future scenario and the current decade base case. The corresponding simulation number is shown is parenthesis

Region	Climate (1)	BVOC (2)	BVOC Future Land Use (3)	US emissions (4)	Boundary Conditions (5)	Combined (6)
PM2.5						
Northwest	7.0	2.1	7.3	43.2	-0.8	51.7
Southwest	3.3	3.3	7.1	20.7	0.7	27.8
Central	10.5	12.6	31.0	14.5	0.0	46.5
South	5.4	21.3	40.5	17.6	1.0	60.8
Midwest	7.8	15.2	37.6	22.4	0.1	61.2
Northeast	7.8	16.0	30.4	28.5	0.0	58.3
Southeast	10.6	29.8	52.4	24.3	0.4	78.5

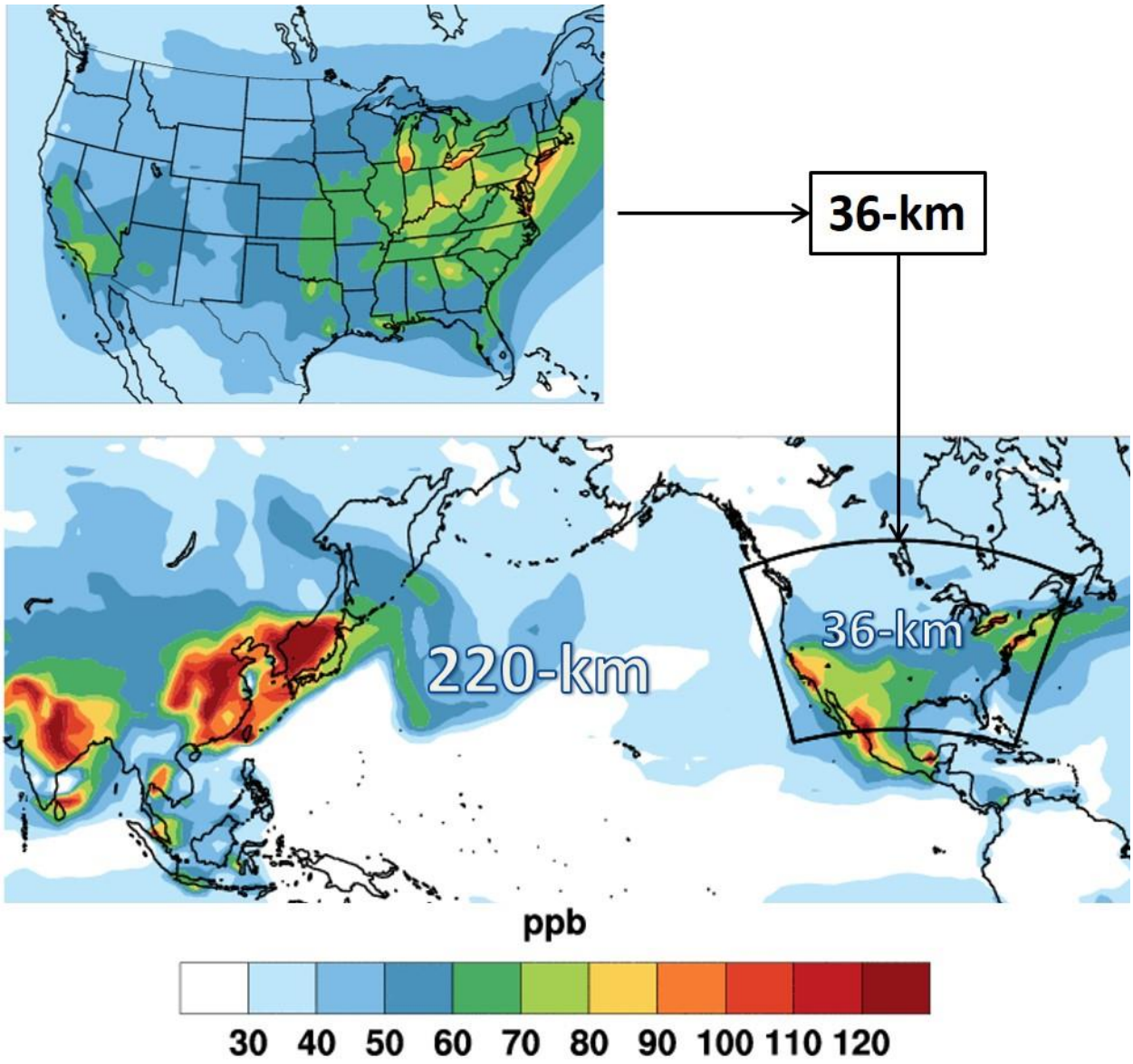

Figure 1. Projected future DM8O concentrations with future anthropogenic and biogenic emissions ~~used to show the~~for the 220-km and 36-km CMAQ modeling domains~~ at 36 and 220 km resolutions~~. ~~The 36 km modeling domain was nested inside the 220 km domain.~~

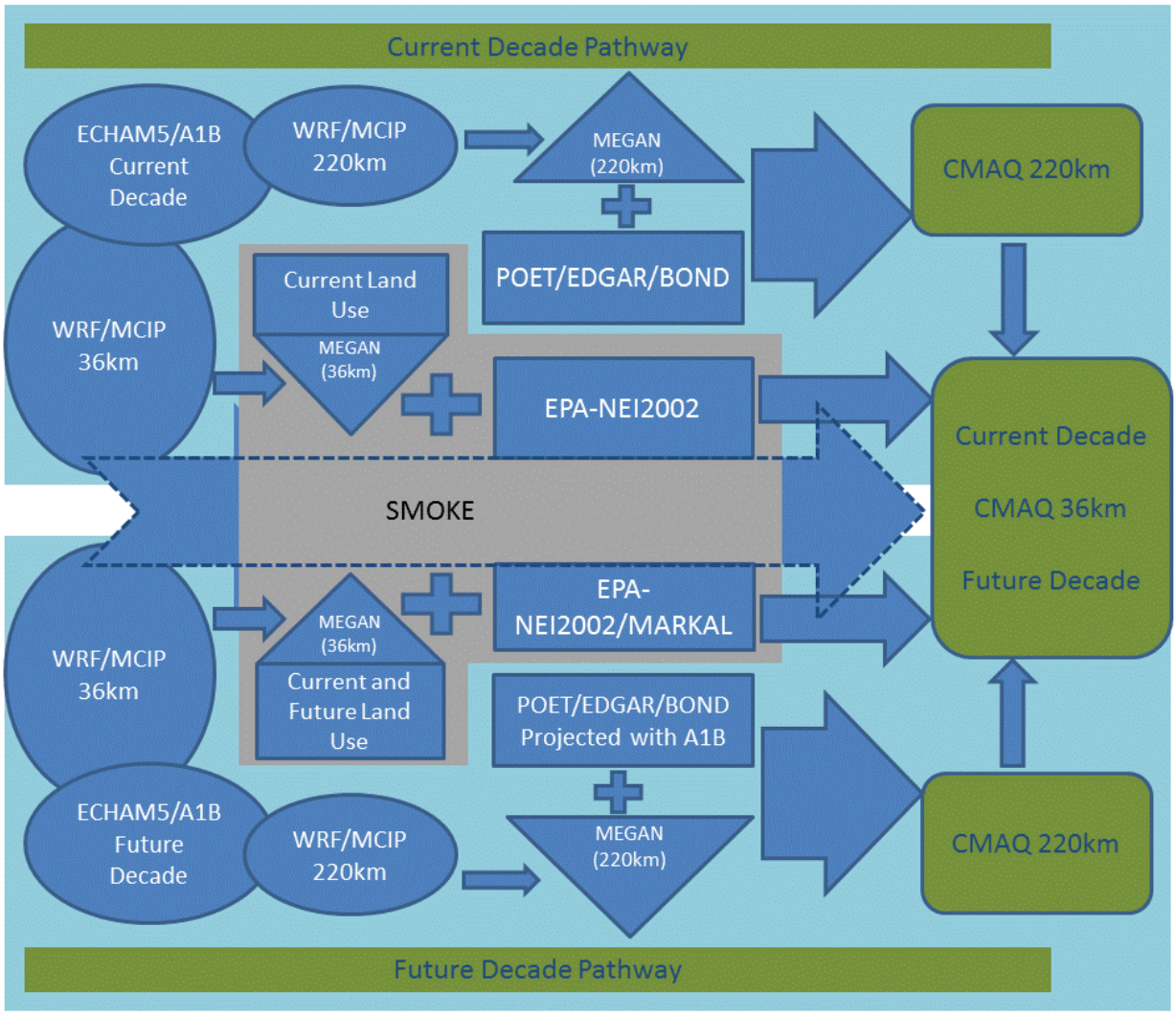

Figure 2. Schematic of the modeling framework.

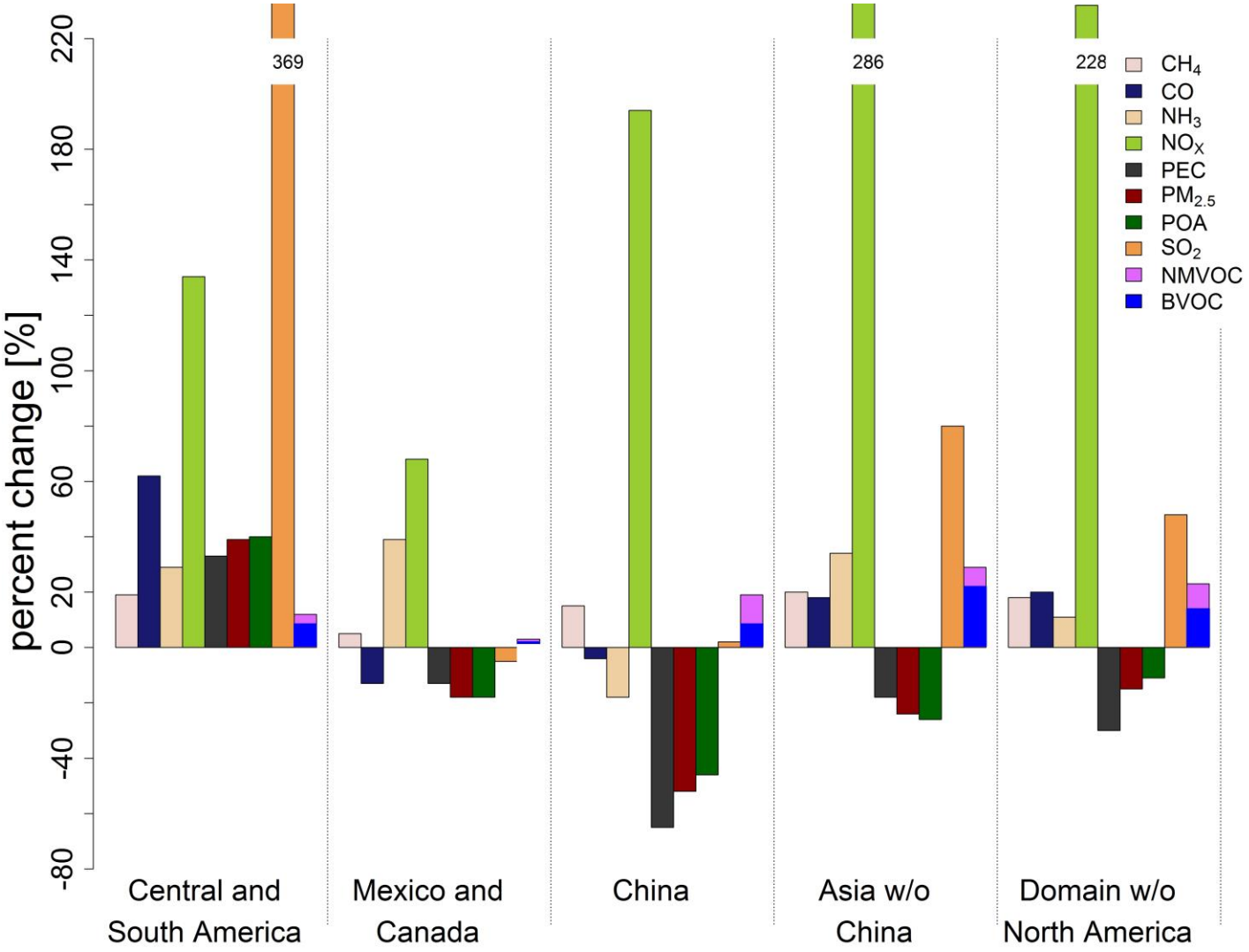

Figure 3. Summary of regional changes in global anthropogenic and biogenic emissions.

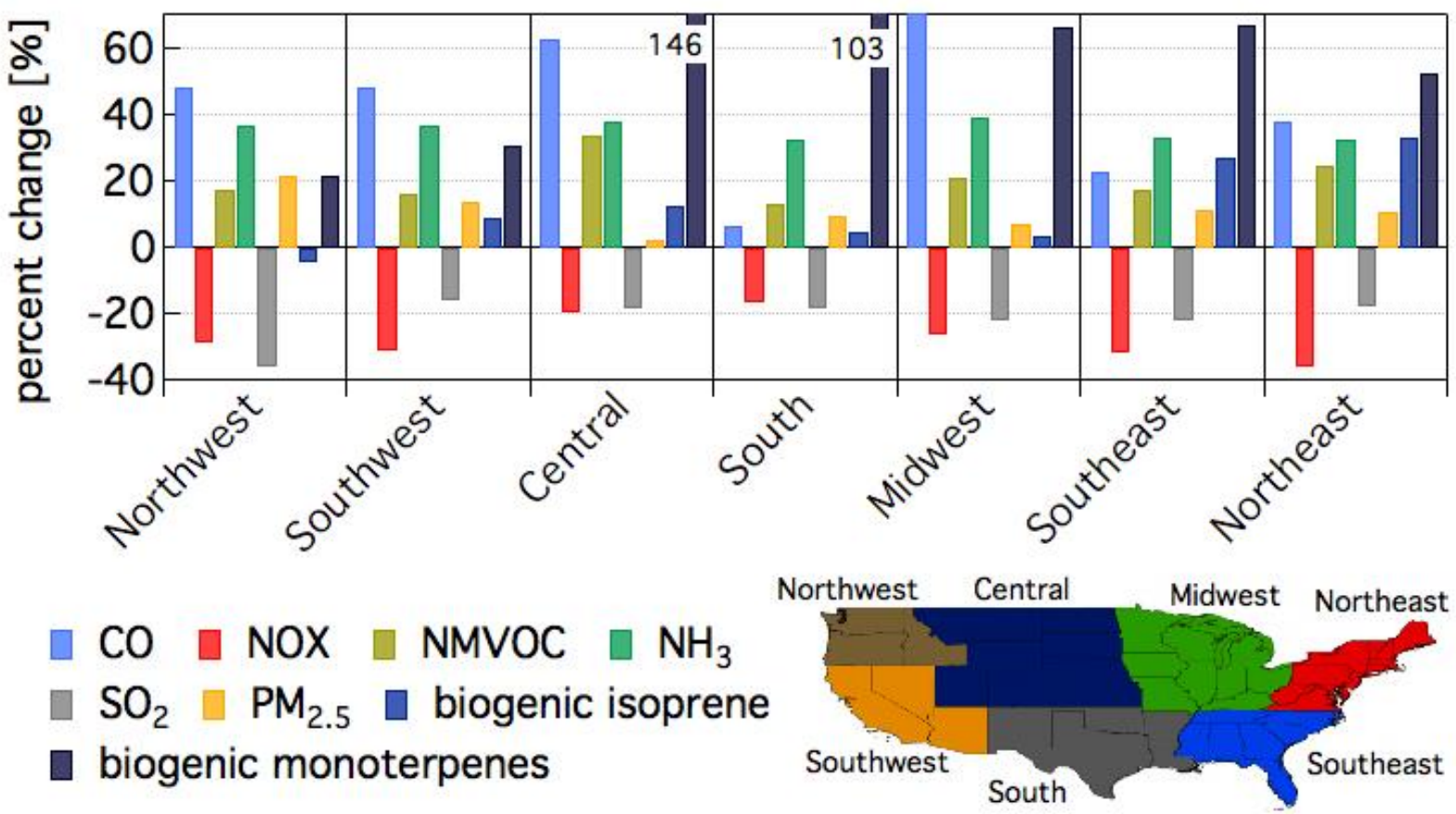

Figure 4~~3~~. Summary of regional changes in US anthropogenic and biogenic emissions from future decade land use.

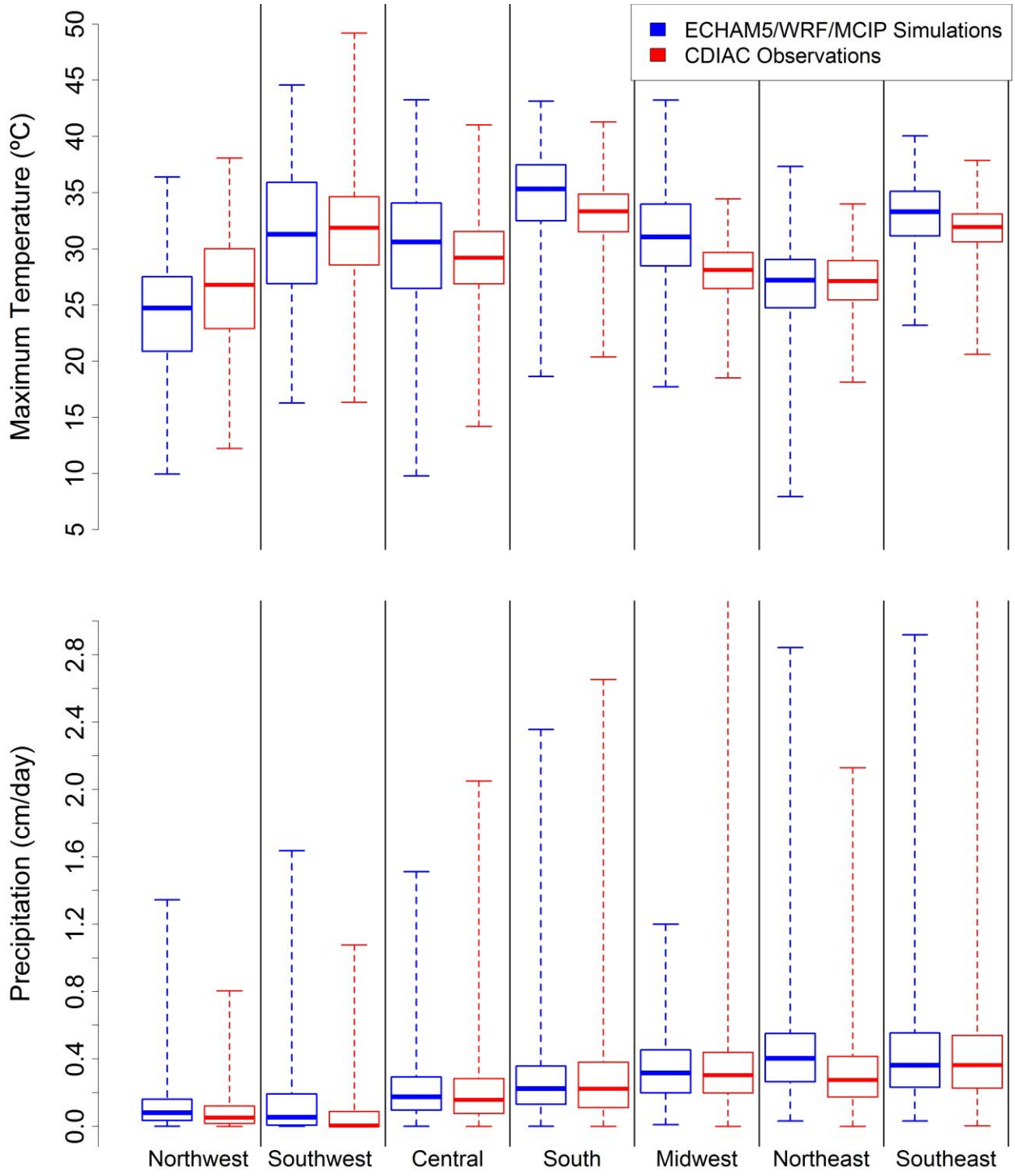

Figure 45. Comparison of modeled and observed seasonal-mean meteorological variables by region: maximum daily temperatures (top); and precipitation rates (bottom). Each box-and-whisker indicates median, 25% and 75% quartiles, maximums and minimums of the values across all sites within each region.

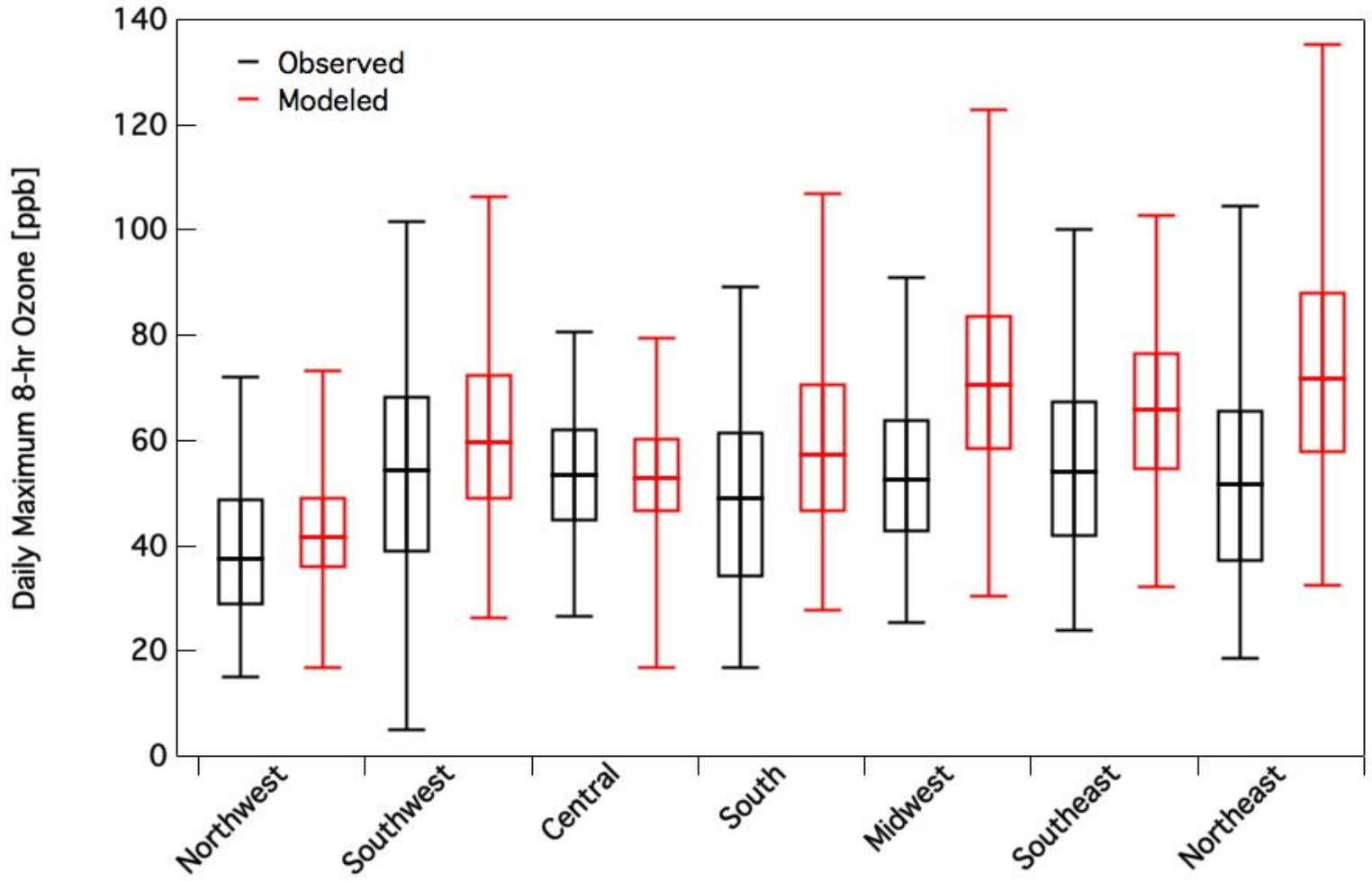

Figure ~~5~~6. 2nd, 25th, 50th, 75th, 98th percentiles of observed ~~(black)~~ vs modeled ~~(red)~~ values of DM8O for each region. ~~The number of monitoring stations per region is shown in parenthesis.~~

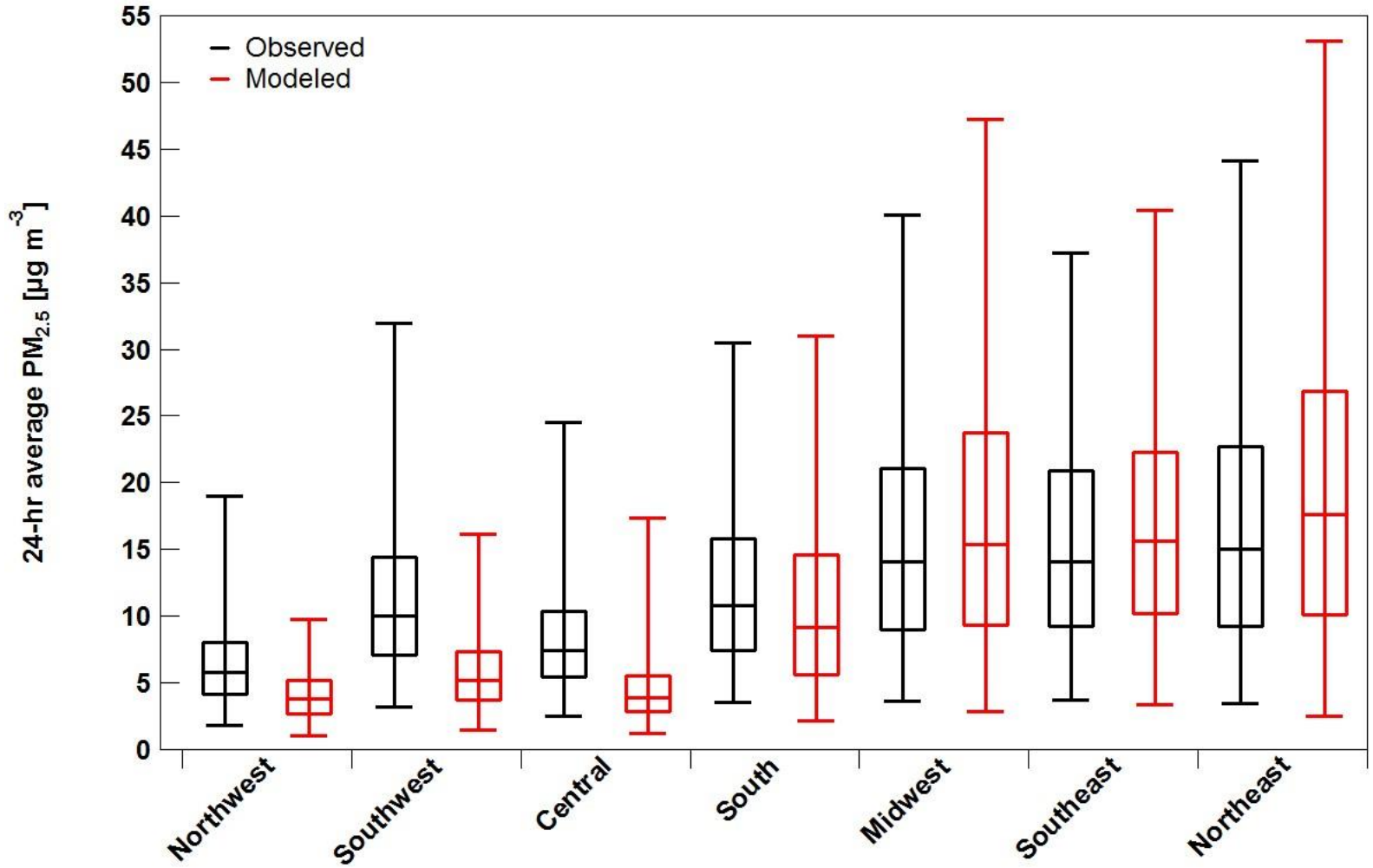

Figure 7. 2nd, 25th, 50th, 75th, 98th percentiles of observed vs modeled values of 24-hr average $PM_{2.5}$ for each region.

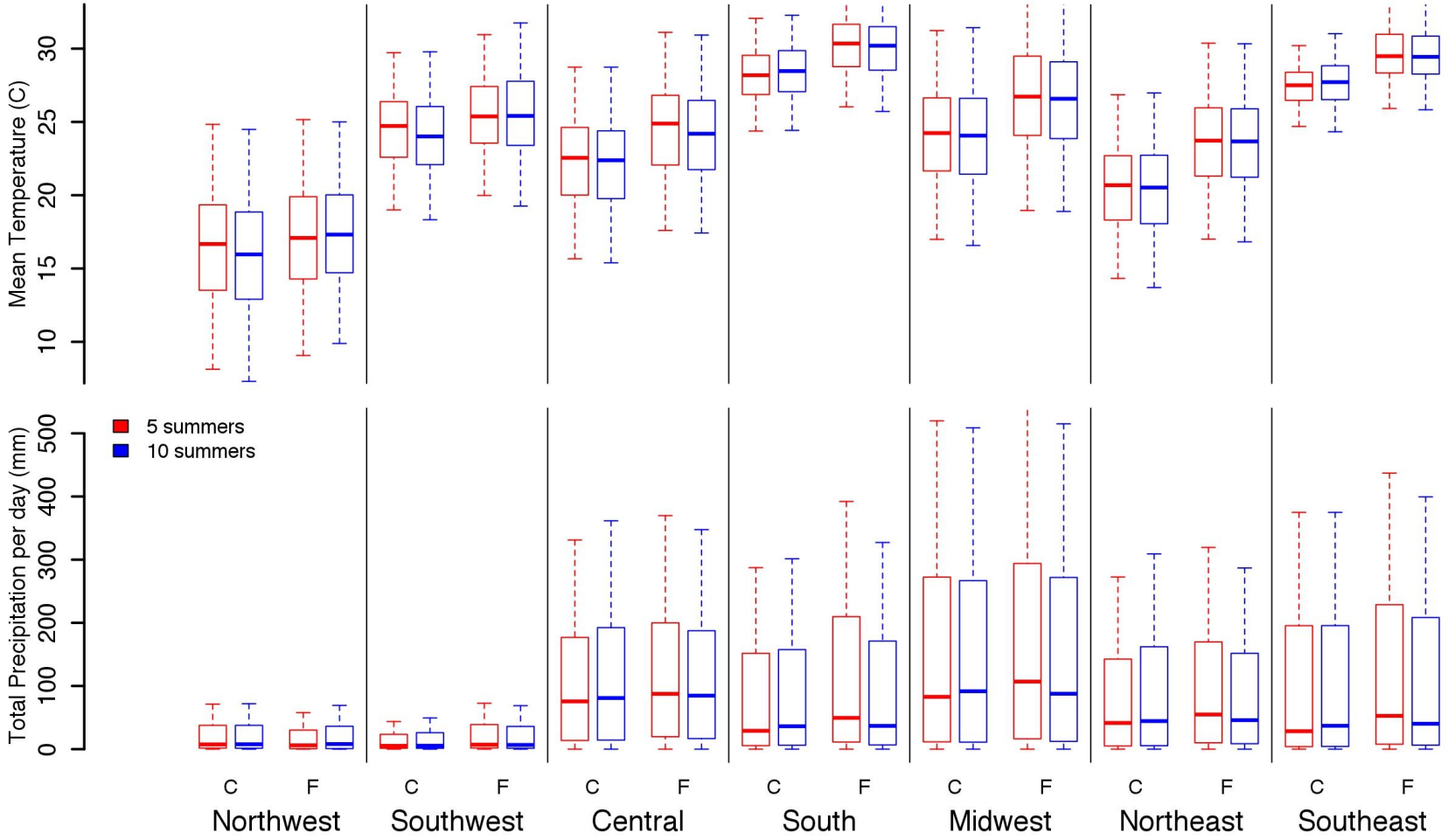

Figure ~~7~~8. (Top Panel) Mean regional temperature for the five chosen summers (red) and ten summers (blue) of the current (C) and future (F) decades. (Bottom Panel) Total regional precipitation per day for the five chosen summers (red) and ten summers (blue) of the current (C) and future (F) decades. Each box-and-whisker indicates median, 5%, 25%, 75% and 95% quartiles within each region.

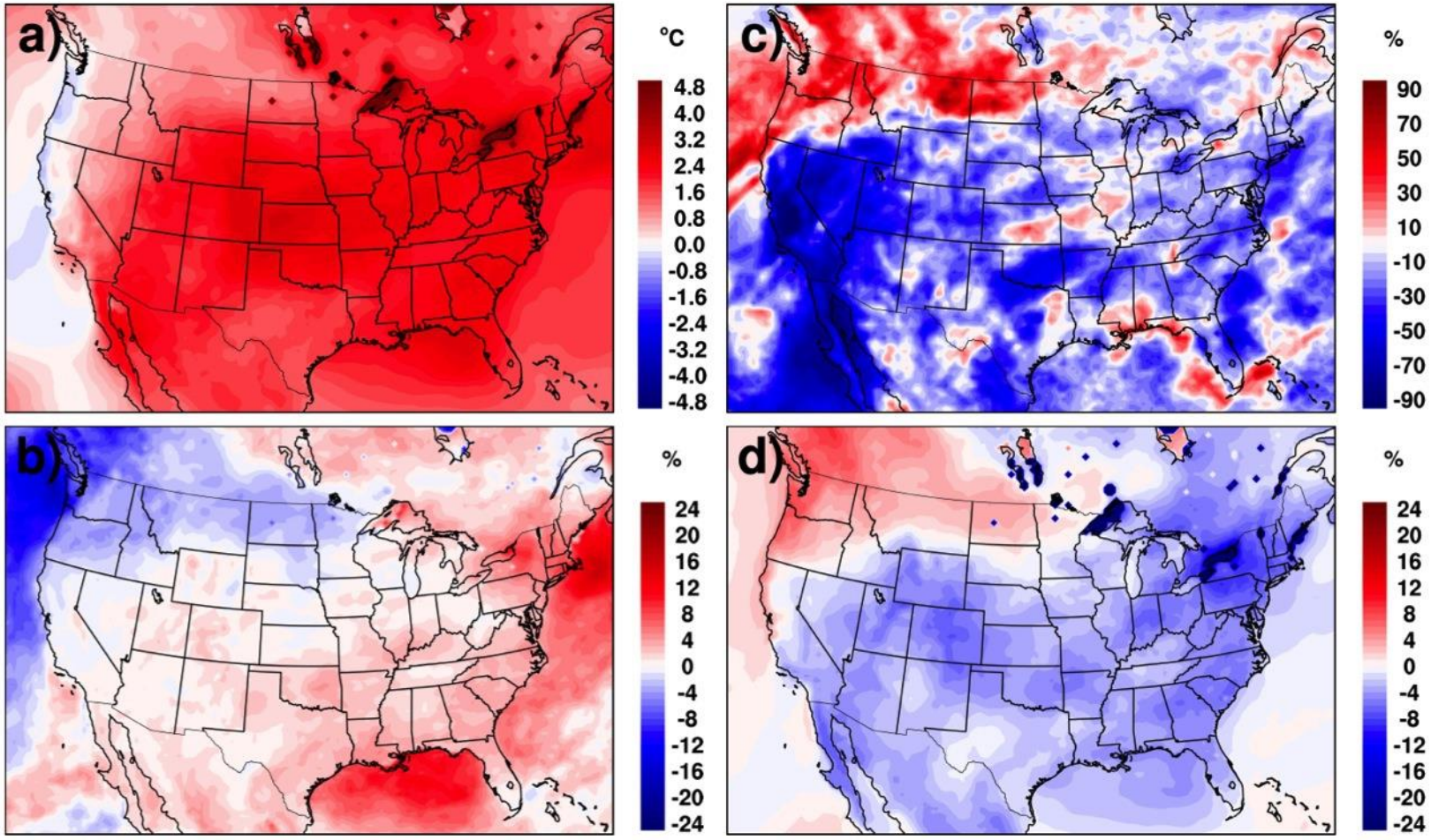

Figure ~~8~~ 9 Projected changes in summertime meteorological fields (future decade - current decade): a) changes in 2-m temperature (°C); b) percent change in solar radiation reaching the ground; c) percent change in precipitation; d) change in relative humidity.

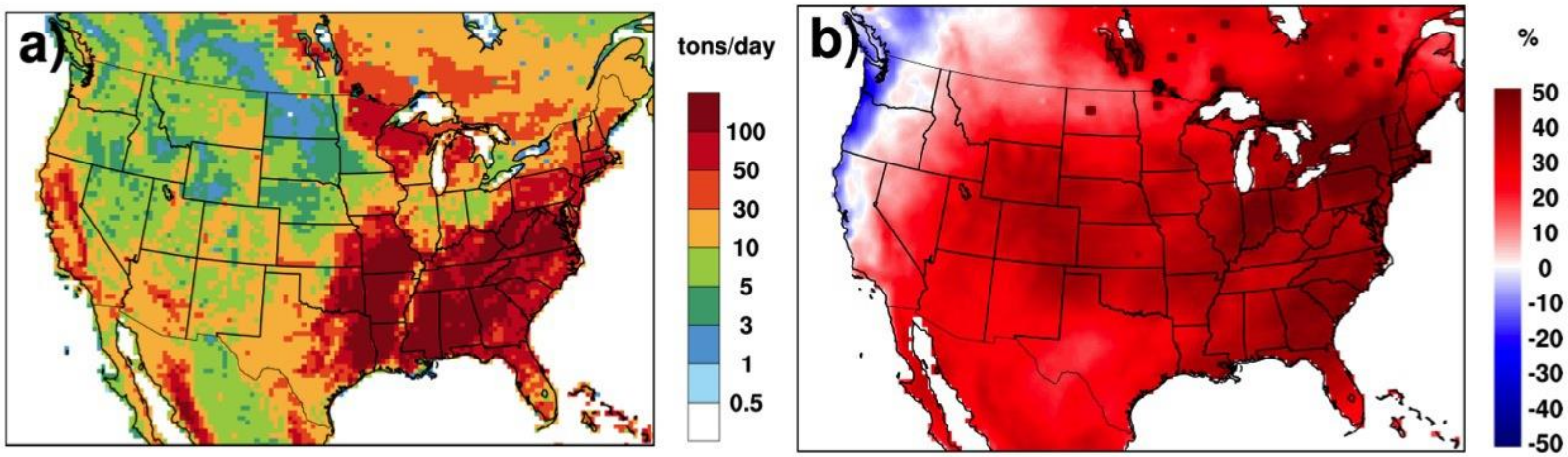

Figure ~~9~~10. a) Current decade summertime isoprene emissions, and b) percent change induced by climate on future summertime isoprene emissions with current decade land use.

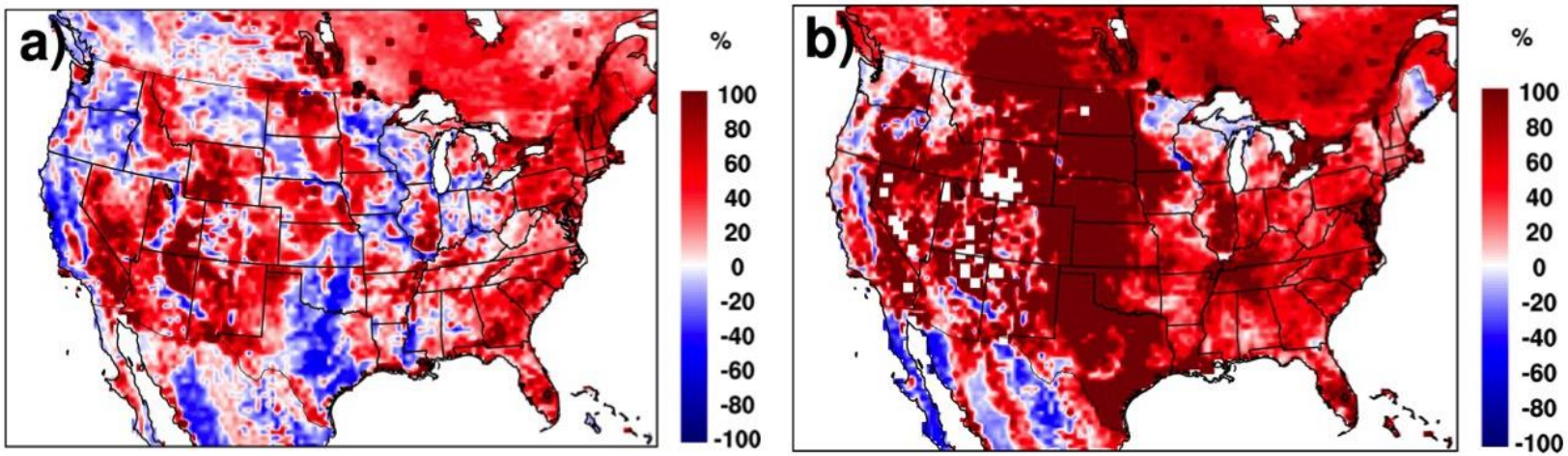

Figure 1~~10~~1. Percent change between future and current decade summertime emissions for future climate and land use for a) isoprene and b) monoterpene.

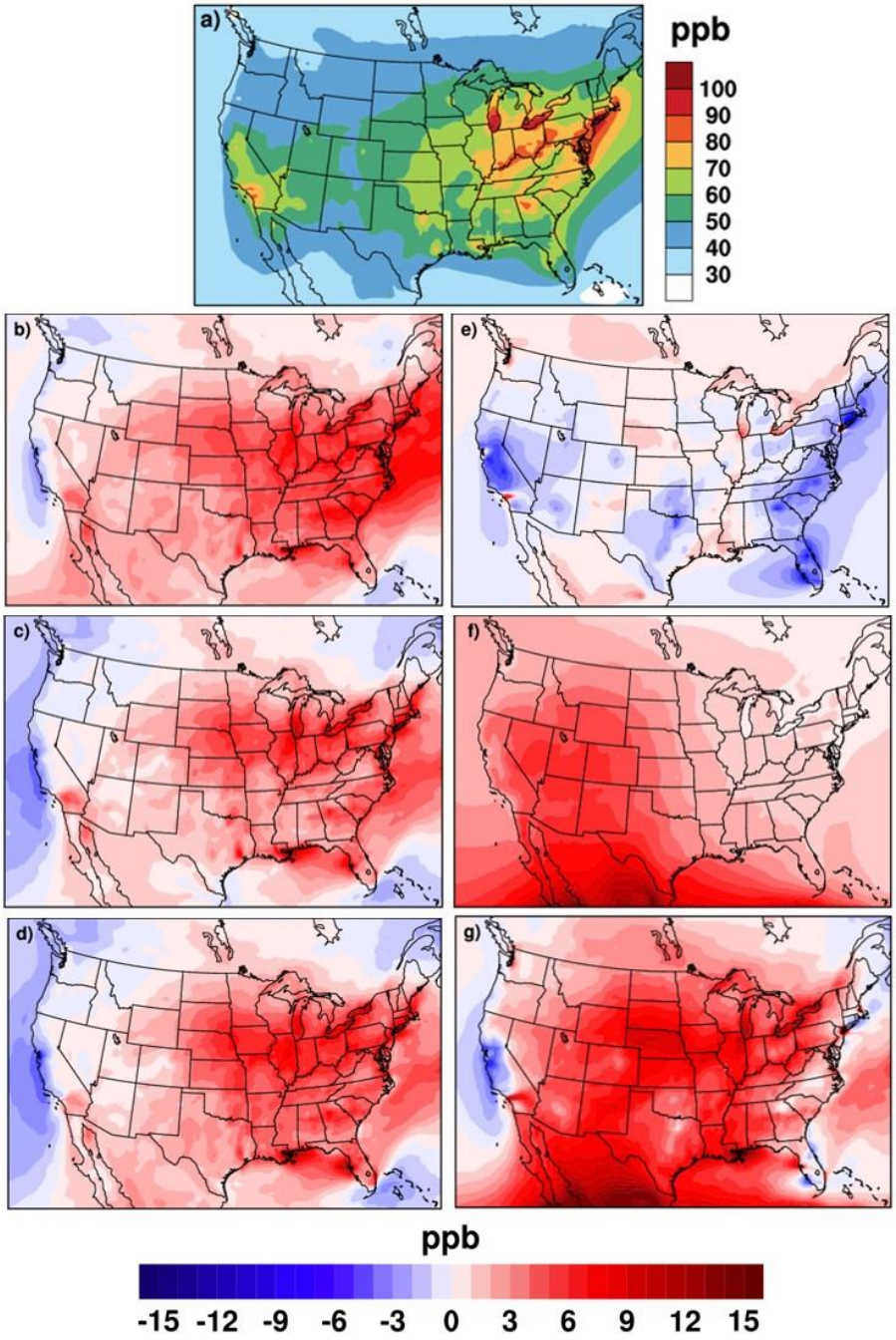

Figure -1211. a) Current decade base case daily maximum 8-hour ozone average concentrations for five summers in the 2000s; spatial distribution; and regional effect on maximum 8-hour ozone due to: b) changes in meteorology (Simulation 1 – Simulation 0); c) changes in meteorology and biogenic emissions (Simulation 2 – Simulation 0); d) changes in meteorology, biogenic emissions, and land use (Simulation 3 – Simulation 0); e) changes in US anthropogenic emissions (Simulation 4 – Simulation 0); f) changes in global anthropogenic emissions (Simulation 5 – Simulation 0); and g) all the changes above combined (Simulation 6 – Simulation 0).

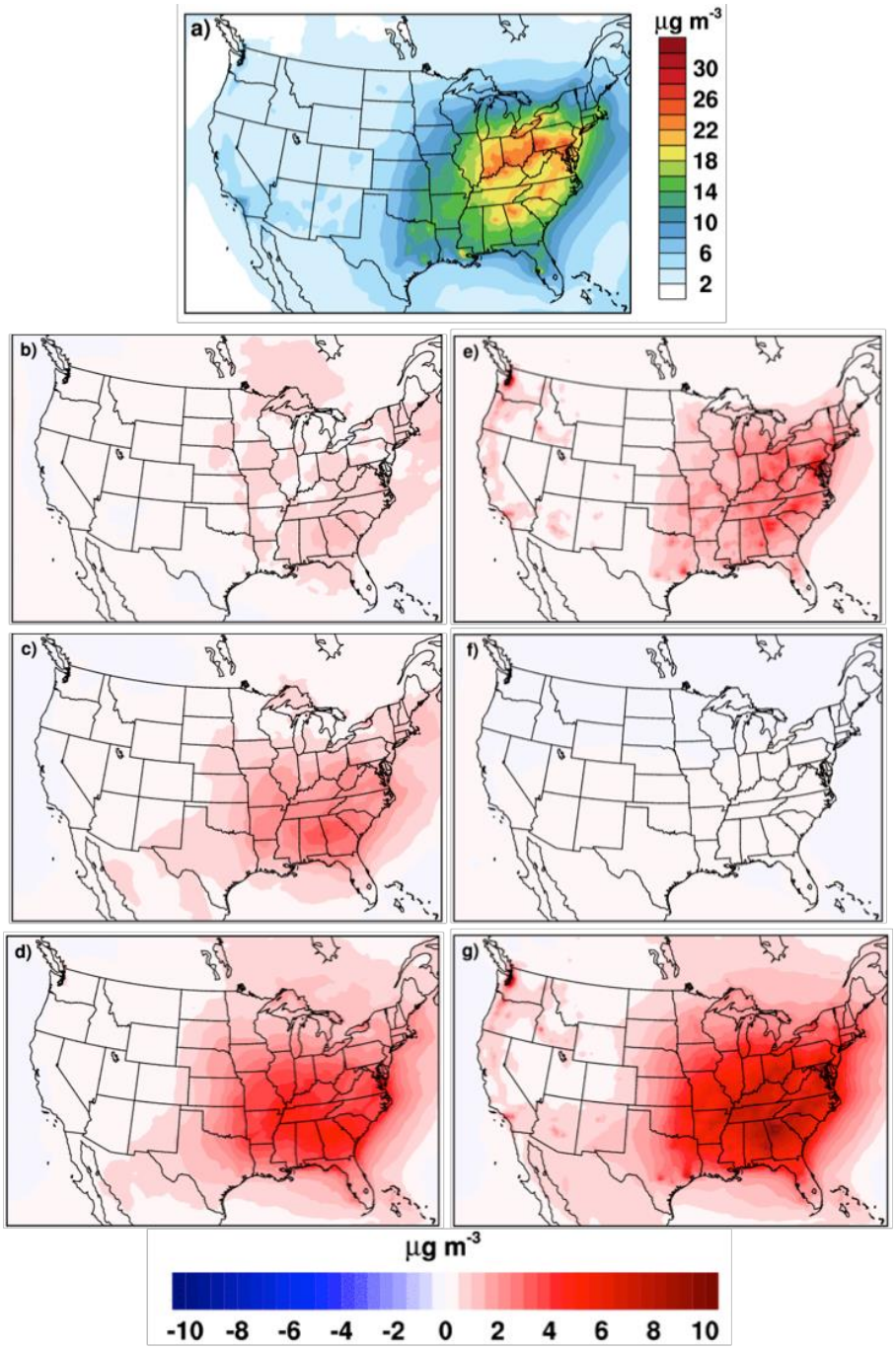

Figure 1~~1~~23. a) Current decade base case $PM_{2.5}$ average concentrations for five summers in the 2000s; and spatial distribution and regional effect on $PM_{2.5}$ due to: b) changes in meteorology (Simulation 1 – Simulation 0); c) changes in meteorology and biogenic emissions (Simulation 2 – Simulation 0); d) changes in meteorology, biogenic emissions, and land use (Simulation 3 – Simulation 0); e) changes in US anthropogenic emissions (Simulation 4 – Simulation 0); f) changes in global anthropogenic emissions (Simulation 5 – Simulation 0); and g) all the changes above combined (Simulation 6 – Simulation 0).

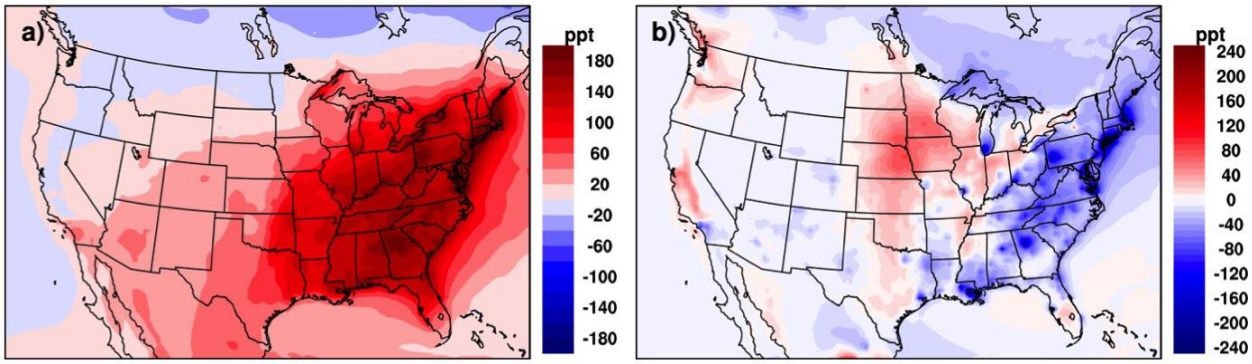

Figure 14.– Differences in (a) RNO_3 and (b) NO_x concentrations between Simulation 2 and Simulation 1.