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February 11, 2014

Prof. Robert Harley
Editor
Atmospheric Chemistry and Physics

Dear Prof. Harley:

Thank you for sending the reviews to our research article acp-2014-784 "How emissions, climate, and land use change will impact mid-century air quality over the United States: A focus on effects at National Parks".

In response to the reviewers' comments, several changes were made to improve the manuscript. Please, find below descriptions of our response to each reviewer comment, and the marked-up manuscript version. In summary, we made the main following changes:

- An evaluation of the simulated O_3 W126 index was included and the W126 index results were modified accordingly.
- The sources of model uncertainties were added and discussed throughout the manuscript.
- Many minor changes were made, including addition of information, and correction of minor lapses in the English.

All line numbers listed below are given for the current version of the manuscript, with the exception of the line numbers noted by the reviewers, which refer to our previously submitted version.

We thank all the reviewers for the careful review and constructive comments on our paper. All coauthors concur with submission of this work in its revised form.

Sincerely,

Maria Val Martin

Response to Reviewer #1

We thank the reviewer for her/his thorough evaluation and constructive comments for improving this manuscript. Our responses to these comments (in blue) are given below.

The paper presents a modeling study investigating the influences of future changes of emissions, climate and land use on air quality in the US National Parks. The authors analyze the future changes in surface ozone and PM2.5 along the RCP4.5 and RCP 8.5 projections in 2050 compared to the present day. They find that while PM2.5 is significantly reduced in the future for both RCP scenarios, surface ozone improves for RCP4.5 but deteriorates in RCP8.5 for some regions. While several studies analyzed future ozone and PM2.5, the novelty of this manuscript is that they separate the individual effects. The paper is well written, analysis is sound and the results are important. However, a number of revisions should be made before the paper is acceptable for publication in Atmospheric Chemistry and Physics.

My main comment is that not enough validation/error analysis was presented for the simulated W126 ozone index, which the authors use to conclude on potential violations of the secondary standard in the future. Models are known to have large errors when simulating this metric (e.g., Tong et al. , 2009; Hollaway et al. , 2012) and the discussion on uncertainty for the modeled results is essential. This is especially true, as the authors conclude that the modeled MDA8 for the summer are biased and these errors are likely to amplify for the modeled W126 metric. Indeed, the simulated present-day W126 values presented in Table 4 are extremely high – how do these compare to observations? E.g., for Shenandoah NP the authors have 66.5 ppm-hrs. The CASTNET annual reports (http://epa.gov/castnet/javaweb/docs/annual_report_2010.pdf) list

W126 at Shenandoah NP as low as 10ppm-hrs in 2010 and 4ppm–hrs in 2009, much lower than simulated in this paper. The authors should either discuss these errors and how they may affect their conclusions on W126 projections or remove Figure 12 and associated discussion.

We agree with the reviewer that there was not enough validation of the simulated O_3 W126 index. To address this comment, we have evaluated the W126 index simulations with observations and included the following discussion in the text.

Section 2.2 (Page 7 Lines 203-215)

> We also evaluate the secondary metric W126 established to protect ecosystems and crops. The W126 is a biologically based index that estimates a cumulative ozone exposure over a 3-month growing season and applies sigmoidal weighting to hourly ozone concentrations [eg, Lefohn et al., 1988, Lapina et al., 2014]. The spatial distribution of W126 (not shown) is similar to the daily MDA-8 O_3 (Figure 2a), but exhibits larger values over regions of low and high ozone due to the sigmoidal weighting of the W126 function as discussed in Lapina et al., [2014]. We find that the model captures the spatial

distribution of W126 across the US (r^2=0.7). However, the model tends to overestimate the magnitude by a factor of 3, in particular over the eastern United States. In previous studies, the lower performance of model simulations of W126 compared to those of daily MDA-8 O_3 has been attributed to the unbalanced sensitivity to model errors at the high end of the ozone concentration range [eg, Tong et al 2009, Holloway et al., 2012, Lapina et al., 2014]. For example, Lapina et al., 2014 report an overestimation of a factor varying between 2 and 4 over the United States in three chemical transport models.

In addition, we acknowledge that our simulated W126 index shows a positive bias, in particular over the eastern US, as many other chemical transport models. We modified the discussion in Section 5.2 and Conclusions accordingly, and modified Figure 12 to show the relative changes in W126 instead of absolute values.

Section 5.4 (Page 16 Lines 547-548)

[…] and show the 2050–2000 difference in W126 to minimize the influence of the positive bias in the simulated W126 index, as discussed in section 2.2.

Section 5.2 (Pages 16-17 Lines 551-558)

[….] *The RCP4.5 scenario projects a general decline in W126: from strong decreases (-39 ppm-hr) in the North East to more moderate decreases (-8 ppm-hr) in the Great Plains. Under RCP8.5 conditions, the changes in W126 are more modest, with decreases of -37 ppm-hr in the North East and increases of +7 ppm-hr in the Great Plains.* Despite the general decrease in daily surface O_3 predicted by both scenarios from strong emission reductions, *our results suggest that the decreases in the W126 index may not be sufficient to keep W126 above the suggested range for a secondary standard (7-15 ppm-hours) to protect vegetation (not shown); however this is difficult to quantitatively assess here given the current model bias.*

Section 5.2 Page 17 Lines 563-568

[…]. *We note that our results indicate an upper limit on the impacts of surface O_3 concentrations on vegetation given the model positive bias in the W126 index. Nonetheless, this study suggests* that O_3 pollution may remain a threat to ecosystems in the U.S. NPs and wilderness areas despite the substantial general decrease in surface O_3 concentrations.

Conclusions Page 18 Lines 601

Furthermore, despite the substantial general decrease in surface O_3, our study *suggests* that the secondary standard W126 may remain above the recommended limits (7--15 ppm-hrs) to protect vegetation […].

Figure 12 caption

Simulated 2050-2000 summertime changes in O_3 W126 for RCP4.5 (blue) and RCP8.5 (red) averaged over the six U.S. climatic regions identified in Figure 4. Numerals indicate the simulated changes in O_3 W126 (ppm-hr).

Other comments:

p 26495, line 19: The sentence "Our study. . ." doesn't fit well with the conclusions before or after. Also, if true, it is rather consistent with many previous works showing that AQ is primarily driven by anthropogenic emissions changes, thus reducing the novelty of this work or the conclusion that climate change and land use are factors that "need" to be considered.

We smoothed the statement as suggested to highlight the relevance and novelty of our findings. The text reads now:

> Our study indicates that anthropogenic emission patterns will be important for air quality in 2050. However, climate and land use changes alone may lead [...]

p 26499, lines 5-7: Did the authors find that the effects of emissions, climate change, and land use interact in a strongly nonlinear fashion? Or could one have estimated the results of treating all of these components together from previous works that have treated them individually? The former would support the novelty of this work.

We find that the effects of emissions, climate and land use changes interact in a nonlinear fashion, that is, the linear sum of the individual forcings (emissions, climate and land use) does not equal the combined effect, in particular for surface O_3. We added a discussion of the non-linearity in the manuscript.

Section 5.1 Page 15 Lines 515-519

> Finally, it is important to note that the effects of emissions, climate and land use need to be considered together when studying changes in surface O_3 since these individual forcings interact in a strongly non-linear fashion. For example, surface O_3 changes in the RCP8.5 scenario are 15% larger in the linear sum of the individual forcings than in the combined effects.

p 26502, lines 1-4: It is difficult to believe that the changes in land use or climate have negligible effects on soil NOx or fertilizer NH3 emissions, which are likely to be important for the air quality in National Parks. Can the authors provide more justification for their assumptions? Is there a previous work citable here?

We agree that soil NO_x and other natural emissions may have an important effect on air quality, in particular for the National Parks. Unfortunately, CAM-Chem does not have a reliable soil NO_x parameterization and it uses a 10-year climatology of monthly-averaged soil moisture from NCEP/NCAR-reanalysis meteorological fields. In addition, we think that emissions of NO from soil, for example, are highly uncertain and the quality of predicted estimates is still not well known [e.g., Fowler et al, 2008]. To make our assumptions clearer, we added the following clarification to the text.

Section 2.1 Page 5 Lines 156-159

> Other natural emissions of O_3 and aerosols precursors (e.g., volcanoes, ocean and soil) may have some impact on surface O_3 and $PM_{2.5}$ on a regional scale over the United States. However, given the large uncertainties on how these emissions might vary in the future, we keep them constant at year 2000 levels.

p 26502, lines 15-19: Please add description for the "2050 Total Change" simulation.

Added as indicated

p 26503, lines 3-4: What was the data coverage for the monitoring sites during the 1998-2010 period?

For CASTNET we used observations from January to December (1995-2005) from a total of 90 sites; for IMPROVE we considered observations from January to December (1998-2010) from a total of 194 sites. We added this additional information in the paper.

Section 2.2 Page 6 Lines 185-187

> Both networks monitor air quality in rural areas at the surface *all year round*. *We calculate long-term means from observations in 90 sites for CASTNet (1995-2005), and 194 sites for IMPROVE (1998-2010).*

P 26504, lines 1-2: While mostly true, this statement exposes a larger problem. The forward model performance can/has been evaluated via comparison to observations. The model sensitivity (i.e., response of the model to changes in emissions or climate) however has not been evaluated, and that is what the authors base their main conclusions on. Thus they really need to consider the validation of their model sensitivities. Below are a couple of major issues with regards to ozone.

First, recent work (Parrish et al., JGR, 2014) has shown that CAM-Chem's response of O3 to changes in emissions underestimates observed responses. How does this impact the projections presented here?

Second, treatment of ozone chemistry (particularly organic nitrates) can drastically alter the sign of the sensitivity of air quality models to changes in VOC emissions (see Mao et al., JGR, 2013, Fig 8). Does the CAM-Chem chemical mechanism lead to positive or negative sensitivities w.r.t. changes in isoprene emissions? How does the response of O3 to NOx in this model compare to others? I suggest at the very least a considerable discussion of these sources of uncertainties, as they are critical for interpretation of the results from this single model.

We thank the reviewer for bringing out the model sensitivity evaluation. We agree that this evaluation is important to support the conclusions in modeling predictions. Such a thorough evaluation is outside of the scope of the work here. Instead, we addressed throughout the paper the two main issues with respect to our ozone simulations highlighted by the reviewer. We also thank the reviewer for pointing out the work of Parrish et al., [2014] and Mao et al., [2013].

First, as highlighted by the reviewer based on findings from Parrish et al [2014], our model tends to underestimate the response of O_3 to changes in emissions. We acknowledged this now in the discussion.

Section 5.1 Page 15 Lines 512-517

> Furthermore, Parrish et al., [2014] show that models (including CAM-Chem) typically underestimate the O_3 response to emissions changes; thus, our sensitivities likely represent a lower limit, and even larger emission-driven changes in O_3 surface concentrations may be anticipated in coming decades.

Second, we agree with the reviewer that the treatment of organic nitrates, i.e., whether isoprene nitrate is a terminal or a temporary sink of NO_x, determines the ozone sensitivity of the model to changes in biogenic emissions. As we explained in the paper (Page 9 Line 293), the chemical mechanism in CAM-Chem recycles 40% of NO_x from isoprene nitrate [Horowitz et al., 2007]. Thus, CAM-Chem has a positive sensitivity with respect to changes in biogenic emissions, i.e., increases in biogenic emissions tend to enhance surface O_3 regardless of the NO_x concentrations. This O_3 response to NO_x with biogenic emission changes in CAM-Chem is slightly different than other models (e.g. GEOS-Chem), where isoprene nitrates represent a terminal sink of NO_x. In this chemical mechanism, increases in isoprene emissions lead to decreases of surface O_3 concentrations in regions with low NO_x concentrations (e.g. southeastern US) and increases in regions with sufficient NO_x (e.g. northern US). We expanded our discussion on the treatment of the organic nitrates in the paper as suggested.

Section 3 Page 9 Lines 287-296

> *Increased biogenic volatile organic compounds (e.g. isoprene) will lead to increases in $PM_{2.5}$ through SOA formation [Heald et al, 2008]. For ozone,* the impact of changing biogenic emissions depends critically on the fate of isoprene nitrates, i.e., whether isoprene nitrate is a terminal or temporal sink of NO_x [e.g., Horowitz et al., 2007,Wu et al., 2012]. In our model, isoprene nitrate recycles 40% of NO_x [Horowitz et al., 2007]. Therefore, increases in biogenic emissions tend to enhance surface O_3 *regardless of the NO_x concentrations. This O_3 response to NO_x with respect to changes in biogenic emissions is slightly different than other models where isoprene nitrates represent a terminal sink of NO_x. In those cases, increases in isoprene emissions lead to increases or decreases in surface O_3 concentrations depending on the availability of NO_x [e.g. Wu et al., 2012, Mao et al 2013].*

p 26506: Please clarify how climate-driven biogenic emissions were calculated in the "Land Use" change simulation, which used the 2000 climate. Were they pre-calculated using the 2050 climate first?

The reviewer is correct and we clarified this point in the text.

Section 2.1 Page 6 Lines 173-175

> In the “2050 Land Use” simulation, climate-driven biogenic emissions in our land use simulations are pre-calculated using the 2050 RCP4.5 and RCP8.5 climate projections.

Figures 2, 6: The model’s horizontal resolution is 1.9x2.5 degrees. The plotted maps however show features on a much finer scale, perhaps due to interpolated contours

The authors should state this in the Figures’ captions, as it can be misleading otherwise.

Clarified as indicated. We added the following in caption of Figures 2, 5, 6, 9 and 10.

> Maps show interpolated contours from the 1.9x2.5 degree horizontal resolution output.

p 26513 and Figure 10: Figure 10 presents the annual mean MDA8 ozone, but it would be more informative and relevant to present estimates on the changes in summertime MDA8.

We thank the reviewer for pointing out the summertime surface MDA-8 O_3 estimates. In this paper, we reported the annual average because we feel it was more useful and relevant for regulatory purposes as the annual average includes the overall effect. In addition, some results in this paper focus on effects over the US National Parks (NP) and wilderness areas, and the NP Service expressed an interest on the annual average results. However, we agree with the reviewer that summertime O_3 changes may be more informative and interesting in terms of changes in surface ozone since concentrations are the highest during this season. In addition to our discussion in section 5.1 (Page 16 Lines 536-538), we have added the following information in the text.

Section 5.1 Page 14 Lines 471-475

> During summertime (not shown), these changes are similar, but more pronounced because O_3 concentrations are the highest during this season: summertime MDA-8 decreases from 62 to 51 ppb in the RCP4.5 scenario and increases (about 6 ppb) over the Great Plain region and decreases (up to 25 ppb) over the eastern U.S. and California in the RCP8.5 scenario.

p 26513, line 8: “daily surface ozone” is too ambiguous, as there can be a number of daily surface ozone metrics.

To clarify the surface ozone concentrations, we specified the daily surface ozone metrics used in the analysis.

Section 5 Page 14 Line 455

> In this section, we first examine future projections on daily surface *MDA-8* O_3 concentrations […]

p 26513, line 16: In my understanding "reducing emissions" is inaccurate here, as in addition to the reduced anthropogenic emissions these simulations include emissions from biomass burning plus varying concentrations of methane.

We agree with the reviewer that the statement "reducing emissions" is not accurate as the 'emissions' simulation includes changes in anthropogenic and biomass burning emissions as well as methane concentrations. We addressed this issue here and throughout the text:

Section 2.1 Page 6 Line 171

> […] anthropogenic emissions including biomass burning emissions *and methane levels* ("2050 Emissions"), […]

Section 4.1 Page 10 Lines 322

> The "emissions" simulation takes into account changes in anthropogenic and biomass burning emissions *and methane levels;* the "land use" simulation […]

Section 5.1 Page 14 Lines 462

> […] show the individual perturbations resulting from changing climate, land use, and *emissions including methane concentrations*.

p 26514, lines 7-10: From Figure 10 the perturbation due to climate appears to be smaller (1%) compared to land use, which is opposite to what is said in the text.

We believe the reviewer was confused by the meaning of the bar plots in Figure 10b. Here the bars are not cumulative, that is, each bar represents the change (in %) for each individual forcing. Therefore, in our example, the contribution from climate is +3% (rather than 1%, as interpreted by the reviewer) and from land use (+2%). To clarify, we added the following to the caption of Figure 10b (and also Figure 6b).

> Bars represent the changes (in %) for each individual forcing, i.e., emissions and methane levels (grey), climate (yellow) and land use (dark red).

Table 1: The units of concentrations are given as "(ppm)". However, the values for N2O and CH4 are given in ppb.

We corrected the concentration units in Table 1 as indicated.

Table 3: I presume that the 2050 methane concentrations were used for both the "2050 Total" and "Emissions" simulations, but this information is currently missing.

I suggest adding another row to the table with “Methane” and/or state this in the text.

As suggested, we added a row in Table 3 with the information on the methane levels

Figure 12: From caption alone it’s not clear whether the presented values are the means for the whole regions or sampled only at the locations of National Parks.

We clarified this point in the caption as follows:

> […] averaged over the six U.S. climatic regions *identified in Figure 4.*

Minor comments:

p 26505, line 10: Please replace “than” with “to”.

Changed as indicated.

p 26507, line 6: Please change to “Interestingly”.

Changed as indicated.

p 26507, line 4: Please remove “s” in “The RCP8.5 scenarios”.

Changed as indicated.

26514 , line 9: Please change to “counterbalanced”.

Changed as indicated.

Figures 6, 8, caption: Please remove coma after “individual perturbations (b)”

Removed as indicated.

Figure 12, caption: Missing “s” in “grey area represent”.

Added as indicated.

Response to Reviewer #2

We thank the reviewer for her/his comments. Our responses to these comments (in blue) are given below.

This paper examines the impact of future emission scenarios, climate, and land use change on air quality over the U.S. with particular emphasis on the National Parks. The paper examines ozone levels, visibility, pm2.5 and the impact of ozone on ecosystems and crops.

The paper is very thorough, well written and provides a good concise analysis with appropriate figures and graphics. I recommend publication although the authors should address the minor comments below.

1) There should be somewhat more discussion concerning the response of VOC emissions to CO2. It is my understanding that the consensus opinion is that there should be a CO2 response (at odds with the assumptions made in the simulations analyzed here). This seems a rather important point in interpreting the paper's results. Page 26501, line 28 states what VOC emissions respond to, but does not mention there is not a response to CO2. It should be explicitly stated the emissions do not respond to CO2 increases. I think there should be some more discussion about this up front.

On page 26506, line 20 the paper states "our isoprene emissions are slightly overestimated", but on page 26511 the paper states "the offsetting effects of climate and CO2 inhibition substantially reduce the role of isoprene emission changes". These statements seem inconsistent. Is the role of CO2 inhibition really slight? At any rate the impact of not including a CO2 response needs to be addressed.

We agree with the reviewer that the response of biogenic emissions to CO_2 inhibition is an important effect that needs to be considered in modeling analyses. Unfortunately just before submitting this manuscript we found a bug in CESM that affected the CO_2 inhibition scheme and learned that our estimated biogenic emissions did not take this effect into account. The bug is now fixed in CESM and we will take this effect in our future studies. For this work, we reviewed the text and clarified better how our results were affected by not including the CO_2 response.

Section 2.1 Page 5 Lines 145-150

> [biogenic emissions] are allowed to respond interactively to temperature, light, soil moisture, leaf age, *carbon dioxide (CO_2) concentrations* and vegetation density [Heald et al., 2008]. *In this work, we do not include the effect of CO_2, which suppresses isoprene production at elevated levels [eg, Heald et al., 2008]; we acknowledge that this is a limitation, which will lead to a slight overestimate in isoprene emissions in 2050 because the CO_2 inhibition would suppress about 10% the isoprene emission efficiency.*

Section 3 Page 9 Lines 286-287

> We note that our isoprene emissions are slightly overestimated *as explained in*

Section 2.1.

Section 5.1 Page 15 Lines 505-508

> It is clear that our land use impacts may be slightly overestimated because we do not include the effect of CO_2 inhibition in our isoprene emissions, as discussed in Sect. 2.1. However, this does not change the positive effect that changes in land use cover have on our surface ozone concentrations.

2) Page 26502, line 2. Are lightning emissions really held constant? This would seem difficult to do as lightning is usually computed interactively.

We thank the reviewer for observing that lightning emissions are usually computed interactively in the models. In CAM-Chem, emissions of NO from lightning use the Price parameterization (Price and Rind,1992; Price et al., 1997) as explained in Lamarque et al. (2012). We clarified this in the text

Section 2.1 Page 5 Lines 151-155

> Lightning NO emissions are also calculated interactively in the model as described in Lamarque et al., [2012]. These emissions respond to climate and cannot be modified in the time-slice experiments. However, they are expected to have a very small impact on the overall surface ozone concentrations [Kaynak et al., 2008]. The global annual lightning emissions change from 4.2 Tg N yr^{-1} in 2000 to 4.4 and 4.8 Tg N yr^{-1}in 2050 for the RCP4.5 and RCP8.5 scenarios, respectively.

3) Page 26503, line 25. "rather overestimated". You don't really need the euphemism, "somewhat". The overestimate is almost a factor of 2.

Modified as suggested.

4) Figure 5. It was not clear to me if the changes in dry-deposition velocity were solely due to landuse change or to landuse change and climate. Could you clarify?

Figure 5 shows O_3 dry deposition velocity changes solely due to land use changes. We clarified this in the caption.

> All maps show changes predicted by the RCP4.5 as a result of the combination of climate, land use and emissions changes, except *for* O_3 dry deposition velocity that shows only *the changes* from land use.

5) Page 26506, line 18. Please give percentage increase of biogenic emissions.

Added as indicated

6) Page 26506, line 28. Paragraph beginning with Land use changes. This is a somewhat strange paragraph as it involves a rather extensive discussion of the impact of dry deposition and land use change, discusses the contrast between this study and other studies, but ends with the fact that the results are not significant. If the results are not significant they should not be discussed at length.

While we agree with the reviewer that the structure of this paragraph may seem strange, we think that it is important to compare our results of land use changes on O_3 dry deposition velocity with previous studies since there are only two examples in the literature [Wu et al., 2012 and Ganzeveld et al., 2010] that focus on future dry deposition changes from land cover on surface ozone and we find different results. Yet, we also find that our results for land cover changes are not statistically significant, and feel that it is critical to state this. However, the two previous studies do not evaluate the statistical significance of their results and that makes the comparison even more difficult. We reworded the paragraph accordingly.

Section 3 Pages 9-10 Lines 306-312

> However, these studies focus on either summertime changes when the broadleaf forests have a larger dry deposition velocity than crops [Wu et al., 2012] or use a different dry deposition parameterization [Ganzeveld et al., 2010]. *We note that the resulting changes in the deposition velocities in our model are not significant at the 95% confidence level and these two previous studies do not evaluate the statistical significance of their results. Nonetheless, this comparison* underlines the important effect that land-use change assumptions have on the projections of future air quality.

7) Page 26507, line 16. What is a pm2.5 chemical species?

We mean with $PM_{2.5}$ chemical species as the major chemical compounds that form $PM_{2.5}$ (e.g. SO_4, NO_3, fine dust, etc). We rephrased the sentence to clarify this point.

Section 4 Page 10 Lines 314

> In this section, we first examine how total and speciated $PM_{2.5}$ are predicted to change in the future [...]

8) Page 26511, line 19. I presume the authors are comparing best days to best days and worse days to worse days in the two scenarios. Is this correct?

We assume the reviewer refers here to our definition of the Haze Index in polluted/cleanest episodes (page 26511 line 21). We clarified this in the text.

Section 4.3 Page 13 Line 417

> Figure 9a shows changes in HI for the most polluted and the cleanest episodes *(ie, worst and best days, respectively)* predicted by the RCP4.5 and RCP8.5 scenarios.

9) Why did you not examine the impact of climate dependent forest fires on ozone? Is it not important?

The effect of climate-driven fire emissions on surface ozone is indeed a very interesting and important question. We are currently working on a project funded by the Joint Fire Science Program that focuses on the effect of climate change in fire activity and the consequences for air quality over the United States. This new project uses CESM at high resolution (50x50 km) with a fire module prediction scheme. Using CESM at such high resolution will resolve the ozone chemistry from fire emissions more accurately and as a consequence, we decided to address this issue in this later study.

10) Table 1. Is the globally area-averaged SST given here?

The reviewer is correct. We specified that in Table 1 now.

11) While the paper is very well written there are the occasional minor lapses. This could easily be remedied by the coauthors who speak English as a native language.

We reviewed the manuscript and corrected the minor lapses.

Manuscript prepared for Atmos. Chem. Phys.
with version 2014/07/29 7.12 Copernicus papers of the LaTeX class copernicus.cls.
Date: 12 February 2015

How emissions, climate, and land use change will impact mid-century air quality over the United States: A focus on effects at National Parks

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Abstract. We use a global coupled chemistry-climate-land model (CESM) to assess the integrated effect of climate, emissions and land use changes on annual surface O_3 and $PM_{2.5}$ ~~on~~ in the United States with a focus on National Parks (NPs) and wilderness areas, using the RCP4.5 and RCP8.5 projections. We show that, when stringent domestic emission controls are applied, air quality is predicted to improve across the U.S., except surface O_3 over the western and central U.S. under RCP8.5 conditions, where rising background ozone counteracts domestic emissions reductions. Under the RCP4.5 scenario, surface O_3 is substantially reduced (about 5 ppb), with daily maximum 8-hour averages below the primary U.S. EPA NAAQS of 75 ppb (and even 65 ppb) in all the NPs. $PM_{2.5}$ is significantly reduced in both scenarios (4 μg/m^3; $\sim$ 50%), with levels below the annual U.S. EPA NAAQS of 12 μg/m^3 across all the NPs; visibility is also improved (10–15 deciviews; >75 km in visibility range), although some ~~parks over the~~ western U.S. parks with Class I status (40–74% of total sites in the U.S.) ~~may not reach~~ are still above the 2050 ~~target to restore visibility to natural~~ planned target level to reach the goal of natural visibility conditions by 2064. We estimate that climate-driven increases in fire activity may dominate summertime $PM_{2.5}$ over the western U.S., potentially offsetting the large $PM_{2.5}$ reductions from domestic emission controls, and keeping visibility at present-day levels in many parks. Our study ~~suggests that air quality in 2050 will be primarily controlled by~~ indicates that anthropogenic emission patterns ~~.~~ will be important for air quality in 2050. However, climate and land use changes alone may lead to a substantial ~~increases~~ increase in surface O_3 (2–3 ppb) with important consequences for O_3 air quality and ecosystem degradation at the U.S. NPs. Our study illustrates the need to consider the effects of changes in

climate, vegetation, and fires in future air quality management and planning and emission policy making.

1 Introduction

Air pollution, such as surface ozone (O_3) and fine particulate matter (with diameter$<$2.5 μm; $PM_{2.5}$), has evolved in both urban and rural regions around the world over the last centuries. Air pollution changes have resulted in part from direct changes in natural and anthropogenic emissions and in part from indirect changes in associated with climate and land use (Jacob and Winner, 2009; Arneth et al., 2010; Fiore et al., 2012). A changing climate is projected to significantly modify both natural and anthropogenic emissions and the atmospheric processes that govern air pollution transport, transformation, and deposition. For example, a warming climate is expected to increase wildfires and associated emissions of trace gases and particulate matter (Spracklen et al., 2009; Yue et al., 2013), cause a general increase in biogenic emissions (Heald et al., 2008), and increase emissions of O_3 and aerosol precursors, such as nitrogen oxides (NO_x) and ammonia (NH_3), from soil and agricultural activities. Anthropogenic emissions are likely to change in response to economic, climatic, and political pressures and policies (IPCC, 2013). In addition, a changing climate is likely to alter precipitation and cloud patterns and synoptic-scale transport processes (Jacob and Winner, 2009). At the same time, changes in land cover and land use will influence the deposition of pollution, as well as the emission of O_3 and aerosol precursors (Ganzeveld et al., 2010; Wu et al., 2012). For example, deforestation decreases turbulent exchange and foliar uptake, prompting a rise in air pollutants. These effects may drive significant local increases or decreases in air pollution.

National Parks (NPs) and wilderness areas in the United States (U.S.) are visited by millions of people every year to enjoy pristine nature. Maintaining adequate air quality conditions in these areas is key to preserving natural ecosystems, preventing negative impacts on visitor and staff health, and maximizing the beauty of landscapes. Air quality management in these regions, including efforts to develop meaningful emissions control strategies, relies on assessment of the current as well as future contributions of natural and anthropogenic sources to local air quality.

Two recent literature reviews (Jacob and Winner, 2009; Fiore et al., 2012) indicate that climate change alone will increase summertime surface ozone in polluted areas by 1–10 ppb. Pfister et al. (2014) predict an increase of about 5 ppb over the Rocky Mountain region during the summer in a future climate, with important potential results to implications for the U.S. National Park Service for air quality management. Surface O_3 is toxic to humans and thus poses a threat to visitor and park staff health. In addition, accumulated exposure to elevated levels of O_3 can damage vegetation (eg Reich and Amundson, 1985; Schaub et al., 2005). Ozone levels have been shown to cause significant yield reduction in a number of major crops on a global scale (eg Avnery et al., 2011; Ghude et al., 2014), and in combination with warming may reduce global crop production by up to 15% in 2050 (Tai et al., 2014), leading to substantial economic losses and potentially worsening global malnutrition. Studies have also reported many other negative impacts on ecosystems, such as reductions in tree and seedling growth, decreases in photosynthetic rates, and visible foliar injuries on multiple plant species, including broadleaf deciduous forest in the northeastern U.S. and needleleaf evergreen forest in the western U.S. (eg Arbaugh et al., 1998; Schaub et al., 2005). In addition, rising O_3 levels may substantially suppress the global land-carbon sink via its negative effect on ~~the~~ photosynthesis, leading to a greater accumulation of carbon dioxide in the atmosphere (Sitch et al., 2007).

Atmospheric fine particles are also harmful to human and ecosystem health. Short-term exposure to $PM_{2.5}$ can lead to respiratory illness such as asthma; longer-term exposure may result in more severe cardiovascular and respiratory diseases as well as lung cancer, increasing the risk of premature mortality (eg Pope and Dockery, 2006). Fine particles and gases cause haze, which degrades visibility. ~~The importance of visibility at~~ Visibility is a protected attribute of some remote locations known as Class I areas, which includes many NPs and wilderness areas~~is recognized and protected by~~. The 1977 Clean Air Act set forth the goal to prevent future and remedy existing visibility impairment in Class I areas. In response, the U.S. Environmental Protection Agency ~~'s~~ (EPA) promulgated the Regional Haze Rule (RHR), which ~~establishes~~ established the goal of returning visibility to natural conditions ~~. The~~ by the year 2064. Specifically, the RHR mandates that each state set "reasonable progress" goals to return visibility to natural conditions on the 20% haziest days by 2064, while preventing further degradation of visibility on the 20% clearest days (US EPA, 2003). Wild and prescribed fires are one of the primary contributors to air pollution, including haze-causing pollutants, in the western and southeastern U.S. (eg Val Martin et al., 2013). Previous studies project that increased fire activity over the western United States will nearly double carbonaceous aerosol by 2050, and produce a significant increase in annual mean $PM_{2.5}$ and haze (Spracklen et al., 2009; Yue et al., 2013)~~, exacerbating efforts to achieve "natural visibility"~~.

In this study, we examine the integrated effect of climate change, anthropogenic emission changes, and land use change on air quality over the United States, with a particular focus on the U.S. National Parks. To our knowledge, this is the first time that the relative effect of these three factors has been considered for U.S. air quality projection. We use a global earth system model to estimate how surface O_3 and $PM_{2.5}$ are expected to change using two Representative Concentration Pathway (RCP) scenarios, represented in the IPCC (2013). We assess the changes in surface O_3 and $PM_{2.5}$ in 2050 relative to present-day levels and discuss the meteorological and chemical drivers behind these changes.

2 Modeling Analysis

2.1 Model description and future changes

To simulate the impact of future changes on the U.S. air quality, we use the Community Earth System Model (CESM) [http://www2.cesm.ucar.edu/]. CESM is a global model, which includes atmospheric, land, ocean and sea ice models that can be run in stand-alone or coupled configurations. We run CESM version 1.1.1 with online computed meteorology and prescribed sea-surface and sea-ice distributions, corresponding to previous fully-coupled simulations. Simulations are performed at the horizontal resolution of 1.9° x 2.5°, and vertical resolution of 26 layers from the surface to about 4 hPa, with a time step of 30 minutes.

To simulate land processes, we use the Community Land Model (CLM) version 4 (Oleson et al., 2010). CLM describes the physical, chemical, and biological processes of terrestrial ecosystems, including the hydrology and carbon cycling of the terrestrial biosphere.

For the atmospheric model, we use the Community Atmospheric Model (CAM) version 4 (Neale et al., 2013) fully coupled with an interactive gas-aerosol scheme (CAM-Chem) ~~(Lamarque et al., 2012)~~ (Lamarque et al., 2012; Tilm The chemical mechanism includes full tropospheric O_3–NO_x–CO–VOC and aerosol phase chemistry, based on the MOZART-4 chemical transport model (Emmons et al., 2010). Simulated aerosol mass classes include sulfate (SO_4), ammonium nitrate (NH_4NO_3), primary carbonaceous aerosols (black carbon, organic carbon), secondary organic aerosols (SOA), sea salt and dust. SO_4 is formed from the oxidation of SO_2 in the gas phase (by reaction with the hydroxyl radical) and in the aqueous phase (by reaction with ozone and hydrogen peroxide). NH_4NO_3 is determined from NH_3 emissions and the parameterization of gas/aerosol partitioning by Metzger et al. (2002), which is based on the level of sulfate present. Black carbon (BC) and organic carbon (OC) aerosols are directly emitted in a combination of hydrophobic and hydrophilic forms (80% and 50% hydrophobic, respectively), and hydrophobic aerosol is converted to hydrophilic with a fixed 1.6 days e-folding time (Tie et al., 2005). Dust and sea salt are implemented following Mahowald et al. (2006a, b), with improvements from Albani et al. (2014); the sources of these natural aerosols are derived based on the model calculated wind speed and surface conditions. SOA ~~are~~ formation is linked to the gas-phase chemistry through the oxidation of isoprene, monoterpenes, alkenes and toluene as in Lack et al. (2004). Finally, dry deposition is represented by the multiple resistance approach of Wesely (1989), with some updates (Emmons et al., 2010; Lamarque et al., 2012; Val Martin et al., 2014). The calculation of dry deposition velocities is performed in CLM and linked to land cover types. Therefore, dry deposition responds to changes in land cover and climate. In this work, we use the optimized dry deposition scheme described in Val Martin et al. (2014), in which the vegetation resistances are linked to the leaf area index (LAI). This optimized dry deposition scheme improves the simulation of O_3 dry deposition velocity, particularly over broadleaf forested regions, and significantly reduces the well-known, long lasting summertime surface O_3 bias over eastern U.S. and Europe in CAM-Chem documented by Lamarque et al. (2012); we discuss this further in section 2.2.

We perform time-slice experiments for 2000 (present-day and baseline) and 2050 (future), under the RCP scenarios designed in support of the IPCC AR5. The RCP RCPs include four scenarios, each of which corresponds to a specific pathway towards reaching a 2100 target radiative forcing (RF) (i.e., 2.6, 4.5, 6.0 and 8.5 Wm^{-2}) associated with greenhouse gases: RCP2.6, RCP4.5, RCP6.0 and RCP8.5, respectively. The RCP2.6 assumes a peak forcing (3.0 Wm^{-2}) in the early 21st century and a decline out to 2100, the RCP4.5 and RCP6.0 scenarios assume RF stabilization after 2100, and the RCP8.5 scenario assumes continuing growth in RF after 2100 (Moss et al., 2011). In this work, we select the RCP4.5 and RCP8.5 scenarios to bracket our results, i.e., we use a stabilization scenario (RCP4.5) and the largest forcing scenario (RCP8.5). Table 1 summarizes the main climate input data for 2000 and 2050. We apply monthly mean time varying sea-surface temperatures and sea-ice distributions generated by the Community Climate System Model, version 4 for the Coupled Model Intercomparison Project Phase 5 (Meehl et al., 2012). Our simulations also consider time varying, zonally averaged greenhouse gas distributions for CO_2, CH_4, N_2O and halogens, and future changes in stratospheric ozone levels.

Table 2 summarizes the main anthropogenic emissions for short-lived air pollutants and biogenic emissions projected over the United States. We divide the emissions for eastern and western U.S. because of the different emission patterns. Emissions of NO_x, NH_3, CO, non-methane volatile organic compounds (VOCs), SO_2 and carbonaceous aerosols for anthropogenic activities and biomass burning are provided in 2000 by Lamarque et al. (2010) and in 2050 by the RCP database (van Vuuren et al., 2011, and references therein). Biomass burning emissions vary among the RCPs and in time, following changes in land cover and land use; however, they do not respond to changes in climate. Biogenic VOCs (e.g., isoprene and monoterpenes) are computed within CLM using the Model of Emissions of Gases and Aerosols from Nature (MEGAN2.1) algorithms (Guenther et al., 2012), and are allowed to respond interactively to temperature, light, soil moisture, leaf age, vegetation density, and carbon dioxide concentrations (Heald et al., 2008) . CO_2 concentrations and vegetation density (Heald et al., 2008) . In this work, we do not include the effect of CO_2, which suppresses isoprene production at elevated levels (eg Heald et al., 2008) ; we acknowledge that this is a limitation which will lead to a slight overestimate in isoprene emissions in 2050 because the CO_2 inhibition would suppress about 10% the isoprene emission efficiency. Both dust and seasalt are also emitted interactively in CESM (Mahowald et al., 2006a, b; Albani et al., 2014). Lightning NO_x emissions are also calculated interactively in the model, as described in Lamarque et al. (2012) . These emissions respond to climate and cannot be modified in the time-slice experiments. However, they are expected to have a very small impact on the overall surface ozone concentrations (Kaynak et al., 2008) . The global annual lightning emissions change from 4.2 Tg N yr^{-1} in 2000 to 4.4 and 4.8 Tg N yr^{-1} in 2050 for the RCP4.5 and RCP8.5 scenarios, respectively. Other natural emissions of O_3 and aerosols precursors (e.g., volcanoes, ocean ~~, soil and lightning) are kept constant at year 2000 levels; potential climate feedbacks on these sources is expected to have a small~~ and soil) may have some impact on surface O_3 and $PM_{2.5}$ on a regional scale over the United States. However, given the large uncertainties on how these emissions might vary in the future, we keep them constant at year 2000 levels.

In addition to climate forcing and emission changes, we include changes in land use induced by human activities in our simulations (Hurtt et al., 2011). We show projected 2050-2000 changes in crops, grasslands and trees over the U.S. for the RCP4.5 and RCP8.5 scenarios in Figure 1 as an example. The RCP4.5 scenario predicts an expansion of forested area, in particular over the eastern U.S. (10%) as a result of mitigation strategies for carbon emission reductions and a decline in agricultural land (8%) due to this afforestation. Conversely, the RCP8.5 scenario predicts an important increase in agricultural land ~~resulting from increasing population~~ (up to 5% in eastern U.S.) ~~and~~ resulting from increasing population as well as grasslands (∼10%) and a decline in forest cover (2%).

For this study, we perform nine simulations: one simulation for present-day and four for each future scenario (Table 3). For the four simulations in the future, we modify one forcing at a time, and name these simulations after their future conditions, i.e., climate alone ("2050 Climate"), anthropogenic emissions including biomass burning emissions and methane levels ("2050 Emissions")~~and~~ , land cover and land use changes including climate-driven biogenic emissions ("2050 Land Use") ~~.~~ and the combined effects of all the individual forcings ("2050 Total Change"). In the "2050 Land Use" simulation, climate-driven biogenic emissions are pre-calculated using the 2050 RCP4.5 and RCP8.5 climate projections. Each model simulation is initialized with a 1-year spin-up run. Following initialization, present-day and future "snapshot" forcing simulations are run for 9 years. We then average the results, and use all years to evaluate interannual variability and ultimately define statistical significance. We replicate these simulations for the RCP4.5 and RCP8.5 scenarios.

2.2 Model evaluation

The CESM simulations driven by online and offline meteorology have been extensively evaluated by comparison with satellite, sonde, aircraft and ground observations of key pollutants on a global scale (Lamarque et al., 2012). Here we focus our evaluation on annual $PM_{2.5}$ and O_3 over the United States and use long-term means from the Interagency Monitoring of Protected Visual Environments (IMPROVE) and the Clean Air Status and Trends Network (CASTNet) datasets. Both networks monitor air quality in rural areas at the surface ~~.~~ all year round. We calculate long-term means from observations in 90 sites for CASTNet (1995–2005), and 194 sites for IMPROVE (1998–2010). Figure 2 compares observed and simulated surface O_3 and $PM_{2.5}$. For O_3, we use the metric for the U.S. EPA air quality standard of daily maximum 8-hour average (MDA-8); for $PM_{2.5}$, we focus on the annual average and determine $PM_{2.5}$ fine mass as the sum of SO_4, NH_4NO_3, organic aerosol

(OA), BC, fine dust and seasalt. We compute OA assuming an average molecular weight of 2.0 per carbon weight for organic carbon (Malm and Hand, 2007). Organic carbon includes SOA. We summarize the comparison between the model and observations using the squared-correlation coefficient (r^2) and the normalized mean bias (NMB) (Figure 2c-d). In Figure 2c, we divide the O_3 comparison into eastern and western U.S. because of the different chemical regimes (eg Murazaki and Hess, 2006; Lamarque et al., 2012). For O_3, we find that simulated surface concentrations show good agreement with the mean observations over the western U.S. (r^2=0.77; NMB=4%), whereas slightly overestimates but slightly overestimate O_3 (r^2=0.47; NMB=16%) over the eastern United States. This annual overestimation is due to a positive bias in summertime O_3 (about 10 ppb), which is a well-known issue and has been previously documented in CESM (Lamarque et al., 2012) as well as other global and regional models (eg Murazaki and Hess, 2006; Fiore et al., 2009; Lapina et al., 2014). Using the optimized dry deposition scheme (section 2.1), we significantly improve the simulation of summertime surface O_3, which has a 30 ppb bias (NMB=60%) over eastern U.S. in the standard dry deposition scheme (Val Martin et al., 2014).

We also evaluate the secondary metric W126 established to protect ecosystems and crops. The W126 is a biologically based index that estimates a cumulative ozone exposure over a 3-month growing season and applies sigmoidal weighting to hourly ozone concentrations (eg Lefohn et al., 1988; Lapina et al., 2014) . The spatial distribution of W126 (not shown) is similar to the daily MDA-8 O_3 (Figure 2a), but exhibits larger values over regions of low and high ozone more emphasized due to the sigmoidal weighting of the W126 function as discussed in Lapina et al. (2014) . We find that the model captures the spatial distribution of W126 across the US (r^2=0.70). However, the model tends to overestimate the magnitude by a factor of 3, in particular over the eastern United States. In previous studies, the lower performance of model simulations of W126 compared to those of daily MDA-8 O_3 has been attributed to the unbalanced sensitivity to model errors at the high end of the ozone concentration range (eg Tong et al., 2009; Hollaway et al., 2012; Lapina et al., 2014) . For example, Lapina et al. (2014) report an overestimation of a factor varying between 2 and 4 over the United States in three chemical transport models.

For $PM_{2.5}$, we find that annual levels are well represented by CESM (r^2=0.70 and NMB=12%; Figure 2d). We further compare the simulated speciated $PM_{2.5}$ chemical species with observations in Figure 3. In our simulations, SO_4 and NH_4NO_3 are somewhat overestimated, whereas OA is underestimated. BC, dust and seasalt concentrations show good agreement with the mean observations, although with some scatter in the relationship (r^2 <0.40; not shown). These results are consistent with previous comparisons over the U.S. (Lamarque et al., 2012; Albani et al., 2014).

It is important to note that in our analysis we mainly concentrate in differences between present-day and future simulations, minimizing the impact of model biases.

2.3 Studied locations

We focus our analysis ~~in~~ on the National Parks and wilderness areas located in the continental United States as shown in Figure 4. We consider the 352 units designated by the U.S. National Park System in the lower 48 states, ~~in~~ of which 46 are classified as protected parks and the ~~rest~~ remaining as monuments, reserves, historical parks and sites and recreational areas. Additionally, we include 109 Class I ~~Mandatory Federal sites~~ areas in the lower 48 states that are not classified as National Park units, but in which air quality is also given special protection. In this work, we present results clustering the NPs and wilderness areas in six climatic regions (ie., Northeast, Southeast, Midsouth, Southwest, West and Great Plains) (Hand et al., 2012). We define these regions and highlight the protected parks in Figure 4.

3 Future changes in meteorological and chemical drivers

Climate and land cover and land use changes affect air pollution through changes in chemistry, transport, removal and natural emissions (eg Heald et al., 2008; Tai et al., 2012; Fiore et al., 2012). We examine here how some meteorological and chemical drivers are predicted to change in the future. Figure 5 shows present-day conditions and 2050-2000 changes in surface temperature, precipitation, boundary layer (BL) depth, isoprene emissions and O_3 dry deposition velocity. We only show changes predicted by the RCP4.5 scenario since the RCP4.5 and RCP8.5 scenarios have similar climates, but the RCP4.5 scenario has a more pronounced increase in isoprene emissions due to land use and climate change. Ozone deposition velocities also differ between the RCP4.5 and RCP8.5 simulations due to differences in projected land use change (Figure 1). To evaluate the statistical significance of our results, we use the Student-t test for a 95% confidence level and highlight the regions which are significant. Previous studies have investigated in detail the sensitivity of surface O_3 (eg Murazaki and Hess, 2006; Leung and Gustafson, 2005) and $PM_{2.5}$ (eg Tai et al., 2012; Leibensperger et al., 2012) to numerous climatic variables. In this work, we do not intend to assess the impact that each climatic variable has on the total change in $PM_{2.5}$ and surface O_3. Instead, we provide here an overview on how these drivers may impact our simulated O_3 and $PM_{2.5}$.

Surface temperature is predicted to increase ~~with~~ by an average of 1.7°C across the U.S. due to the rising greenhouse gases in the RCP 4.5 scenario (Figure 5a). The extent of this increase varies across the U.S., with ~~the~~ a maximum increase of 4°C observed over the central United States. The RCP8.5 scenario predicts a similar increase ~~than~~ to the RCP4.5 scenario: 2.0°C. We find that the 9-year simulations generate robust increases in surface temperature changes across most of the continental United States. Previous studies have reported similar results with distributions and magnitudes differing slightly depending on the model, resolution and the climate scenario considered (eg Murazaki and Hess, 2006; Kelly et al., 2012; Pfister et al., 2014). It is known that high ozone levels correlate well with temperature in many polluted regions due to the connection between temperature to stagnation conditions, enhanced photochemistry and biogenic and wildfire emissions (Fiore et al., 2012, and references therein). $PM_{2.5}$ is also affected by many of the same meteorological processes as surface O_3, although the relationship is more complex and the sign of the effect can be positive or negative because of the different sensitivities of the $PM_{2.5}$ chemical species (eg Tai et al., 2012). Thus, our simulated increase in temperature will intensify surface O_3 and most probably $PM_{2.5}$ pollution over the United States.

Air quality is also sensitive to precipitation and cloud cover. For example, $PM_{2.5}$ is expected to decrease in regions with increased precipitation (eg Pye et al., 2009; Racherla and Adams, 2008). In our simulations, precipitation decreases over most of the continental U.S. (30%), with some small increases over some regions in the northwestern U.S. (8%) (Figure 5b). However, not all of the changes in precipitation are significant and the absolute changes are generally small (<1 mm/day) despite the large percentage change. We find similar pattern in the cloud cover (not shown). A decrease in cloudiness is associated with an increase in solar radiation, which favors surface O_3 production in our simulations.

An important meteorological process for diluting and transporting air pollutant is mixing within the boundary layer. In our simulations, the boundary layer depth across the U.S. is predicted to generally increase, with the largest increase over in central U.S. (>100 m; about 20%) (Figure 5c). Increases in BL depth favors ventilation and reduces pollutant accumulation. In our simulations, we notice that BL depth increases (i.e., favoring low $PM_{2.5}$ and O_3 concentrations) and precipitation (and cloud cover) decreases (i.e., favoring high $PM_{2.5}$ and O_3 concentrations) are generally co-located. These two processes have opposite effects on air quality and this highlights the ~~difficulty of~~ challenges in predicting possible air quality ~~impact~~ impacts resulting from climate change.

Higher temperature and solar radiation will also affect biogenic emissions, which in turn will influence $PM_{2.5}$ and surface O_3. Biogenic emissions will also depend on land use changes. In 2050, isoprene emissions are predicted to increase from 28 to 43 Tg C (about 53%) in the U.S. (Table 2), with 10% of this increase driven by land use changes. This effect is more significant in the south-eastern U.S. (about 25%) due to afforestation (Figure 5d). The RCP8.5 scenario also predicts an increase in biogenic emissions, but with a lower influence from land use and climate changes (~~2~~33%; not shown). ~~The~~ We note that our isoprene emissions are slightly overestimated as we neglect the effect of CO_2 inhibition, explained in Section 2.1. Increased emissions of biogenic volatile organic compounds (e.g. isoprene) will increase $PM_{2.5}$ through SOA formation (Heald et al., 2008) . For ozone, the impact of changing biogenic emissions ~~on surface O_3~~ depends critically on the fate of isoprene nitrates, i.e., whether isoprene nitrate is a terminal or temporal sink of NO_x (eg Horowitz et al., 2007; Wu et al., 2012). In our model, isoprene nitrate recycles 40% of NO_x (Horowitz et al., 2007). Therefore, increases in biogenic emissions tend to enhance surface O_3~~. In addition, increased biogenic volatile organic compounds (e. g. isoprene) will~~ , regardless of the NO_x concentrations. This O_3 response to NO_x with respect to changes in biogenic emissions is slightly different than other models, where isoprene nitrates represent a terminal sink of NO_x. In those cases, increases in isoprene emissions lead to increases ~~in $PM_{2.5}$ through SOA formation (Heald et al., 2008)~~ or decreases in surface O_3 concentrations depending on the availability of NO_x (eg Wu et al., 2012; Mao et al., 2013) .

Land use changes can also influence deposition processes. For example, large O_3 dry deposition velocities are associated with denser, broadleaf forests (i.e., with high LAI) and crops (eg Wesely, 1989; Val Martin et al., 2014), whereas grasslands and needleleaf forests (i.e., with low LAI) are characterized by low deposition velocities. In our simulations, the O_3 dry deposition velocity ~~shows a general~~ generally shows a small decrease across the U.S. (0.2–1.0 cm/min; about 1–3%) (Figure 5e). The RCP8.5 scenarios projects more variable, but even smaller changes in the O_3 dry deposition velocity (<0.6%), associated with a less pronounced change in vegetation. ~~Interesting~~Interestingly, in this study we find a reduction in the annual O_3 dry deposition velocity due to the shift from croplands to grasslands and forests. This result contrasts with previous studies that report decreased dry deposition velocities in regions with increased agricultural land (Ganzeveld et al., 2010; Wu et al., 2012). However, these studies focus on either summertime changes when ~~the~~ broadleaf forests have a larger dry deposition velocity than crops (Wu et al., 2012) or use a different dry deposition parameterization (Ganzeveld et al., 2010)~~, and this underlines the important effect that land-use change assumptions have on the projections of future air quality~~. We note that the resulting changes in the deposition velocities in our model are not significant at the 95% confidence level ~~.~~ and these two previous studies do not evaluate the statistical significance of their results. Nonetheless, this comparison underlines the important effect that land-use change assumptions may have on the projections of future air quality.

4 Future $PM_{2.5}$ air quality

In this section, we first examine how total and speciated PM~~$_{2.5}$ and PM~~$_{2.5}$ ~~chemical species concentrations~~ are predicted to change in the future due to climate, emissions and land use changes. We then discuss the impacts of future climate-driven wildfire activity in $PM_{2.5}$ and haze.

4.1 Regional annual changes in $PM_{2.5}$

Figure 6 shows changes in annual surface $PM_{2.5}$ concentrations following the RCP4.5 and RCP8.5 scenarios over the continental United States. The projected changes in 2050 from the combined effects and the individual effects of emissions, climate and land use change are also shown. The "emissions" simulation takes into account changes in anthropogenic and biomass burning emissions and methane levels; the "land use" simulation is associated with changes in climate-driven biogenic emissions and land cover. We also indicate the regions with confidence levels higher than 95% from the Student-t test; we find that the 9-year simulations generate robust results across most of the continental U.S. for the simulations with the combined effects and emissions alone.

The combined effects of changing climate, land use, and emissions lead to a strong decrease in $PM_{2.5}$ concentrations across the continental U.S. (Figure 6a), with an average projected decrease of about 4 μg/m^3 ($\sim$50%) for both the RCP4.5 and RCP8.5 scenarios. The absolute decrease is stronger in the eastern than in the western U.S., about 4 μg/m^3 versus 2 μg/m^3, because the eastern U.S. is characterized by larger $PM_{2.5}$ concentrations (Figure 2b). Projected changes in U.S. $PM_{2.5}$ for 2050 largely reflect changes in anthropogenic emissions, which drive the majority (>95%) of this decrease all over the United States. The contribution of climate and land use changes, although minor and rather insignificant in most of the U.S., may counteract the benefits of emissions reductions in some regions (Figure 6b). For example, the RCP4.5 scenario projects ~~about~~ a 47% total average decrease in $PM_{2.5}$ in the Southwest region, with about 52% drop due to emission reductions, but a counter veiling increase of 5% and 0.1% from climate and land use, respectively. In many regions the impact of climate ~~and~~ or land use change is not significant compared to climate variability when averaging over 9 years.

To examine in more detail future changes in $PM_{2.5}$ we show changes in ~~the individual~~ speciated $PM_{2.5}$ ~~chemical species~~ in Figure 7. We find that the decrease in $PM_{2.5}$ concentrations is mainly driven by decreases in SO_4 and, to a lesser extent, in NH_4NO_3 and BC. Under the RCP4.5 and RCP8.5 scenarios, anthropogenic SO_2 emissions are projected to decrease substantially in the western and the eastern U.S. compared to present-day (84% and 89% in RCP4.5 and 69% and 90% in RCP8.5, respectively; Table 2). Large decreases in NO_x emissions are also projected (75% and 78% in RCP4.5 and 50% and 72% in RCP8.5), whereas NH_3 emissions increase (33% and 25% in RCP4.5 and 59% and 44% in RCP8.5). The largest significant change in $PM_{2.5}$ is projected with the RCP8.5 scenario over the Northeast region, with a decrease of 90% in BC, 79% in SO_4 and 46% in NH_4NO_3. Organic aerosol increases slightly, in particular over the Northeast, Southeast and West regions. This increase does not offset the decreases in the other species, yet it can be important in some regions. Over the Southeast, the RCP4.5 and RCP8.5 scenarios project similar decreases in SO_4, NH_4NO_3 and BC. However, $PM_{2.5}$ concentrations are predicted to be lower in the RCP8.5 than in the RCP4.5 scenario because of the relative importance of OA in the total $PM_{2.5}$ loading. Higher OA concentrations in the RCP4.5 scenarios result from higher VOC emissions (Table 2) associated with reforestation and climate change, as discussed in section 3.

Our results are consistent with previous studies, which have shown the small impact of climate change on $PM_{2.5}$ levels and the significant contribution from projected emissions reductions (eg Tagaris et al., 2007; Pye et al., 2009; Lam et al., 2011; Kelly et al., 2012). Comparing $PM_{2.5}$ projections from different studies is not straightforward due to variations in the study region, reported $PM_{2.5}$ metrics and use of different climate and emissions (Fiore et al., 2012). A decrease of about 2 μg/m^3 (25%) over the U.S. was projected for the SRES A1B scenario by Tagaris et al. (2007) for the combined effect of climate and emissions, with the bulk of this decrease resulting from sulfate, nitrate and ammonium reductions. Using the same scenario, Lam et al. (2011) found a similar decrease (4–5 μg/m^3), with 90% of the reduction due to emission reductions. Most recently, Kelly et al. (2012) reported summertime regional decreases of more than 3 μg/m^3 over the U.S., with the SRES A2 climate and RCP6.0 emission scenarios.

We summarize the simulated $PM_{2.5}$ changes over the U.S. NP and wilderness areas in Table 4. We show results for the 46 protected National Parks located in the continental United States. We find that the RCP4.5 and RCP8.5 scenarios predict a significant reduction of $PM_{2.5}$ levels across the protected NPs, with the exception of the Crater Lake and Lassen Volcanic NPs. In these two NPs, the RCP8.5 scenario projects a slight increase in annual $PM_{2.5}$, but concentrations are predicted to remain below 12 μg/m^3, the primary annual U.S. National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$. In the Joshua Tree NP, both RCP scenarios predict a significant improvement of $PM_{2.5}$ air quality, but with an annual average above 12 μg/m^3 due to the dominance of natural dust in this region.

It is important to note that changes in the frequency and magnitude of the fire resulting from climate change are not included in this analysis, and this effect may have an important impact on the $PM_{2.5}$ levels associated with climate change, as discussed in the following section.

4.2 Effects of increased fire activity on summertime $PM_{2.5}$

Climate-driven changes in fire emissions can be an important factor controlling $PM_{2.5}$ concentrations (Spracklen et al., 2009; Yue et al., 2013). Yue et al. (2013), use results from 15 climate models following the SRES A1B scenario and a fire prediction model of area burned to predict increases of 63–169% in area burned over the western U.S. in 2050, which leads to about 150–170% increases in OC and BC fire emissions. The RCP4.5 scenario predicts an increase of about 60% in OC fire emissions over the western U.S., whereas the RCP8.5 projects a marginal decrease of 0.3%. These two RCP scenarios clearly underestimate the average increase in carbonaceous aerosol fire emissions associated with climate feedbacks as projected by Yue et al. (2013).

To assess the importance of climate-driven fire emissions on future $PM_{2.5}$, we perform an additional simulation (not shown in Table 3), where we increase the RCP fire emissions over the U.S. in order to match Yue et al. (2013)'s projection. In doing so, we keep the spatial distribution of fire as described by the RCP scenarios and apply a homogeneous increase on a monthly basis. We scale the RCP fire emissions over the U.S. and Canada, with the exception of the eastern U.S., where fire activity is not predicted to significantly increase in the future due to climate (Scholze et al., 2006; Moritz et al., 2012).

Figure 8 shows the effect of climate-driven fire emissions on summertime $PM_{2.5}$. We focus on the summer here which is the peak fire season in the United States. We compare the $PM_{2.5}$ levels predicted by the RCP scenarios in 2050 to those when climate-driven fire activity is included, and only show those climatic regions where $PM_{2.5}$ is affected by fire, i.e., West, Great Plains, Southwest and Northeast. $PM_{2.5}$ concentrations in these regions increase significantly as a result of increased fire activity. These increases are most prominent over the West and Great Plain regions, in which fire-driven $PM_{2.5}$ may potentially offset anticipated reductions in anthropogenic emissions. For example, over the West region we estimate that fire activity may increase future summertime $PM_{2.5}$ from 3.2 $\mu g/m^3$ to 5.2 $\mu g/m^3$ (63%) in the RCP8.5 scenario and from 4.5 to 5.6 $\mu g/m^3$ (22%) in the RCP4.5 scenario. The concentration of organic aerosol nearly doubles in both scenarios, and this dominates the total change in $PM_{2.5}$. It is important to note that our fire OA may be underestimated as we do not include secondary production of OA from fire emissions. Increased fire activity may also affect $PM_{2.5}$ further downwind from the fires. We estimate that summertime $PM_{2.5}$ may increase up to 4–10% in the Northeast region due to smoke transported from fires in the western U.S. and the boreal region.

Therefore, changes in summertime $PM_{2.5}$ concentrations may be dominated by changes in fire activity in most of the western U.S. in a future climate. This same fire pollution may ~~contribute significantly to impairing~~ significantly impair visibility over this region, as well as hundreds of kilometers downwind from the fire sources.

4.3 Effects on future visibility

We evaluate the effects of future changes in visibility in the U.S. NP and wilderness areas across the continental U.S. by examining changes in the haze index (HI) and visibility range. We calculate the HI based on the definition of the US EPA (2003) and the visibility range as in Pitchford and Malm (1994) using the results of the daily averages of $PM_{2.5}$ chemical species. Figure 9a shows changes in HI for the most polluted and the cleanest episodes (ie, worst and best days, respectively) predicted by the RCP4.5 and RCP8.5 scenarios. We define most polluted and cleanest episodes as those days characterized by aerosol levels with the 20% worst and best visibility, that is, with the HI above the 90th percentile or below the 10th percentile, respectively (US EPA, 2003). As an example, we show in Figure 9b the cumulative distribution function of daily HI over two protected national parks: Crater Lake and Acadia NP, located over the West and Northeast region, respectively. We also include the impact of fire pollution in this analysis and indicate the 2050 HI target required to reach natural background conditions by 2064 as mandated by the Regional Haze Rule.

Consistent with the $PM_{2.5}$ projections, we predict a significant visibility improvement in both polluted and background conditions over the continental United States. This improvement results mainly from the large reduction in anthropogenic emissions, with the strongest absolute reductions in areas with high $PM_{2.5}$ and high anthropogenic aerosol precursor emissions such as the Northeast region. In this region, our results show a reduction of up to 15 deciviews during cleanest days and up to 10 deciviews during most polluted events in both RCP scenarios, which corresponds to an increase of more than 75 km in visibility range.

The improvement in $PM_{2.5}$ air quality is reflected in the projected visibility over the U.S. National Parks and wilderness areas. For example, in Acadia NP, we find that both RCP scenarios predict HI level decreases of about 10 deciviews during the most polluted events, leading to an improvement in visibility range of more than 70 km. This NP is estimated to reach the 2050 target to restore natural visibility conditions by 2064, even during most polluted conditions. However, this is not the case for all the protected NPs and wilderness areas. Our results show that visibility in Crater Lake NP is estimated to improve by 2050, with moderate HI decreases ($\sim$ 4 deciviews) predicted by both RCP scenarios, and a general improvement of visibility range of 30–40 km. However, HI levels are predicted to remain higher than the 2050 target. This is also the case for other important NPs located in the western U.S. such as Yellowstone, Grand Canyon, and Mount Rainier NPs; about 40% and 74% of the total parks may not reach the 2050 target as predicted by the RCP4.5 and RCP8.5 scenarios, respectively.

Future regional visibility may also be impaired by fire pollution resulting from climate change. We find that fire pollution may maintain visibility levels at present-day conditions during the most polluted events in some NPs and wilderness areas (e.g. Crater Lake NP; Figure 9b) or may impede the attainment of the 2050 visibility target (e.g. Yellowstone NP; not shown). Our analysis shows little or no effect of fire in visibility impairment in NPs and wilderness areas located in the Northeast and Southeast climatic regions (e.g., Acadia NP; Figure 9b). Yue et al. (2013) estimate that future fire activity would lead to an average visibility decrease of 30 km in the 32 Federal Class I areas located in Rocky Mountains Forest. Our predictions for the Rocky Mountain NP show more moderate decreases in visibility (4–6 km; not shown). However, our work differs from Yue et al. (2013) in both the model resolution (200 km versus 400 km) and the spatial distribution of the fire emissions.

5 Future changes in surface O_3

In this section, we first examine future projections on daily surface MDA-8 O_3 concentrations and evaluate the contributing factors to this future change. We then discuss how future changes in surface O_3 may impact ecosystems.

5.1 Predictions of daily O_3 concentrations

Figure ~~11~~ 10 shows the 2050–2000 changes in annual mean surface O_3 concentrations predicted by the RCP4.5 and RCP8.5 scenarios in the continental United States. As in the $PM_{2.5}$ analysis, we present total changes in the simulated daily MDA-8 O_3 concentrations and show the individual perturbations resulting from changing climate, land use, and ~~reducing emission~~ emissions including methane concentrations (Figure ~~11~~10a). We also highlight the regions with confidence levels higher than 95% from the Student-t test.

The combined effects of changing emissions, climate and land use produce a strong decrease in surface O_3 across the continental U.S. in the RCP4.5 scenario, with the strongest absolute reductions (up to 10 ppb) over the eastern U.S. and California, regions with the highest O_3 concentrations (Figure 2a) and strongest anthropogenic precursors emissions reductions. The average MDA-8 over the U.S. decreases from 52 ppb to 47 ppb from present to future days. However, the RCP8.5 scenario predicts important increases over the Great Plain region (about 5 ppb) and marginal decreases (about ~~1~~ 2 ppb) over the eastern U.S. and California. During summertime (not shown), these changes are similar but more pronounced because O_3 concentrations are the highest during this season: summertime MDA-8 decreases from 62 to 51 ppb in the RCP4.5 scenario and increases (about 6 ppb) over the Great Plain region and decreases (up to 15 ppb) over the eastern U.S. and California in the RCP8.5 scenario.

The RCP4.5 and RCP8.5 scenarios project strong and similar decreases in domestic O_3 precursor emissions (Table 2), however global CH_4 concentrations are 50% larger in RCP8.5 compared to RCP4.5 (2740 versus 1838 ppb; Table 1). Rising surface O_3 levels over central U.S. in the RCP8.5 scenario are therefore the result of elevated background O_3 due to rising CH_4 levels in combination with climate and land use changes. These individual effects can be clearly seen in Figure ~~11~~10b, which shows that climate and land use changes completely offset the emission reductions over the West and Midsouth regions in the RCP8.5 scenario. For example, in the West region, the RCP8.5 scenario predicts an overall increase of 3% in surface O_3 ($\sim$3 ppb), in which the contribution from emission reductions (-2%) is ~~counterbalance~~ counterbalanced by climate (+3%) and land (+2%) changes.

The impact of the rising background O_3 in the RCP8.5 scenario can also be seen on the surface O_3 concentrations over the ocean. Similar to previous studies (eg Wu et al., 2008; Fiore et al., 2012), the RCP4.5 scenario projects a decrease in O_3 levels (up to 5 ppb) over the Pacific and Atlantic oceans in a changing climate due to the decrease of O_3 lifetime associated with higher water vapor. The shorter lifetime of PAN in a future climate may also contribute to the decrease of O_3 levels over remote areas (eg Wu et al., 2008; Doherty et al., 2013). By contrast, the RCP8.5 scenario projects an increase in surface O_3 (up to 8 ppb) due to the rising background over these remote regions.

Climate and land use changes alone are also expected to significantly impact future O_3 air quality. When only climate change is considered and the emissions of ozone precursors are held at present-day levels ("Climate" simulation), simulated surface O_3 increases by 1 and 2 ppb across the U.S. in the RCP4.5 and RCP8.5 scenarios respectively, with the largest absolute changes over the eastern U.S. (up to 3 and 5 ppb, respectively). Note that this "climate penalty" does not include the effect of changing biogenic emissions, which is incorporated in the land use change simulations. However, Tai et al. (2013) show that the offsetting effects of climate and CO_2 inhibition substantially reduce the role of isoprene emission changes in the climate penalty. Thus, the climate effect shown here may be a good proxy for the climate penalty and is comparable to values shown by Tai et al. (2013). In the land use change simulation, surface O_3 increases by 2 ppb in both scenarios, with the largest increases over the central U.S. (up to 8 and 4 ppb, respectively). Increases in surface O_3 result ~~from a decrease in dry deposition velocity due to the shift from croplands to grasslands projected in~~

both scenarios over this region (Figure 1), as discussed in section 3. Increases in surface O_3 result mainly from climate-driven increases in biogenic VOCs and, to a lesser extent, from a decrease in dry deposition velocity due to the shift from croplands to grasslands projected in both scenarios over this region. It is clear that our land use impacts may be slightly overestimated because we do not include the effect of CO_2 inhibition in our isoprene emissions, as discussed in section 3. Sect. 2.1. However, this does not change the positive effect that changes in land use cover have on surface ozone concentrations.

Our projected change in surface O_3 is more moderate than that reported in previous studies (eg Tagaris et al., 2007; Nolte et al., 2008; Kelly et al., 2012; Pfister et al., 2014). However, these studies do not account for changes in land cover, which our work indicates can be regionally quite substantial. Furthermore, Parrish et al. (2014) show that models (including CAM-Chem) typically underestimate the O_3 response to emissions changes; thus, our sensitivities likely represent a lower limit, and even larger emission-driven changes in O_3 surface concentrations may be anticipated in coming decades. Finally, it is important to note that the effects of emissions, climate and land use need to be considered together when studying changes in surface O_3 since these individual forcings interact in a strongly non-linear fashion. For example, surface O_3 changes in the RCP8.5 scenario are 15% larger in the linear sum of the individual forcings than in the combined effects.

Figure 11 shows the impact of these changes on surface O_3 over the U.S. National Parks and wilderness areas. Under RCP8.5 conditions, we find an improvement of surface O_3 air quality for most polluted days (ie. high tail of the distribution is lower than present-day), except in the Great Plains region, and a deterioration in the background O_3 (i.e., the low tail of the distribution is higher than present-day) all across the United States. These results are due to increases in CH_4 emissions in combination with the effects of climate and land use changes as discussed above. However, under RCP4.5 conditions, there is a clear general improvement of surface O_3 air quality across the U.S., with the exception of increasing background O_3 in the Northeast, Southeast and Midsouth regions. Furthermore, as discussed in Pfister et al. (2014), background O_3 at high elevations may be affected by long-range transport of pollution and stratospheric intrusions (eg Eyring et al., 2010; Lin et al., 2012). Both processes are taken into account in our simulations (but not disaggregated), and are expected to change in the future due to decreasing NO_x emissions in Asia (van Vuuren et al., 2011) and the recovery of the stratosphere O_3 layer (Eyring et al., 2010; Kawase et al., 2011).

In all of the U.S. protected NPs and wilderness areas (Table 4), surface O_3 levels are predicted to improve under the RCP4.5 scenario. We estimate that annual concentrations are projected to be below the current primary EPA NAAQS of 75 ppb to protect public health, and even below a more restrictive potential future standard of 65 ppb. In contrast, under RCP8.5 conditions, numerous parks and wilderness areas are predicted to have poorer O_3 air quality. For example, 34 out of the 46 protected NPs in the lower 48 states may encounter surface O_3 increases with respect to present-day levels (e.g., Glacier and Yellowstone NPs), although projected concentrations are below 65 ppb.

However, during the summer, when O_3 concentrations are higher, 16 out of 46 NPs are predicted to have summertime surface O_3 levels above 65 ppb (e.g., Rocky Mountain and Yosemite NPs) (not shown).

5.2 Effects on future ecosystem O_3 damage

To investigate the effect of projected changes in surface O_3 levels in the U.S. NPs and wilderness areas, we use the secondary metric W126 established to protect ecosystems and crops. ~~The W126 is a biologically based index that estimates a cumulative ozone exposure over a 3 month growing season and applies sigmoidal weighting to hourly ozone concentrations (eg Lefohn et al., 1988; Lapina et al., 2014) .~~ Figure 12 presents average W126 over the U.S. NPs and wilderness areas divided in the 6 climatic regions for present-day and future. We focus on summertime W126 as the summer season is the growing season for many ecosystems ~~. The spatial distribution of~~ and show the 2050–2000 difference in W126 ~~(not shown) is similar to~~ to minimize the influence of the ~~MDA-8 O_3 (Figure 2a), but with the regions of low and high ozone more emphasized due to the sigmoidal weighting of the~~ positive bias in the simulated W126 ~~function~~ index, as discussed in ~~Lapina et al. (2014)~~ section 2.2.

Consistent with the daily O_3 pattern, the RCP4.5 and RCP8.5 scenarios project a decrease in the W126 index across the continental U.S., with the exception of the Great Plain region by the RCP8.5 scenario. The RCP4.5 scenario projects a general decline in W126: from strong decreases (-39 ppm-hr) in the North East to more moderate decreases (-8 ppm-hr) in the Great Plains. Under RCP8.5 conditions, the changes in W126 are more modest, with decreases of -37 ppm-hr in the North East and increases of $+7$ ppm-hr in the Great Plains. Despite the general decrease in daily surface O_3 predicted by both scenarios from strong emission reductions, our results ~~show that the~~ suggest that the decreases in the W126 index may ~~remain~~ not be sufficient to keep W126 above the suggested range for a secondary standard (7–15 ~~ppm-hours) throughout most of the United States. Under RCP8.5 conditions, our simulations predict that the W126 index will remain above the maximum recommended limit (15 ppm-hours)~~ ppm-hr) to protect vegetation ~~, with the exception of the Southeast region, where W126 will still remain above the minimum recommended limit (7 ppm-hours). The RCP4.5 scenario predicts lower W126 levels, yet still above the minimum recommended standard in almost all the regions~~(not shown); however, this is difficult to quantitatively assess here given the current model bias.

The simulated W126 over the U.S. protected NPs is summarized in Table 4. Our study shows that a number of protected NPs will experience W126 levels exceeding the secondary standard to protect vegetation. The RCP8.5 ~~scenarios~~ scenario projects that the majority of the protected parks will have an W126 index above the recommended limits, with 34 parks above 7 ppm-hours and 26 parks above 15 ppm-hours; projections from the RCP4.5 result in 26 and 6 parks, respectively. ~~Therefore,~~ We note that our results indicate an upper limit on the impacts of surface O_3 concentrations on vegetation given the model positive bias in the W126 index. Nonetheless, this study suggests that

O_3 pollution may remain a threat to ecosystems in the U.S. NPs and wilderness areas despite the substantial general decrease in surface O_3 concentrations.

6 Conclusions

We have quantified for the first time changes in air quality between present and a 2050 future period associated with changes in emissions, climate, and land use change over the United States. In particular, we focus on the implications of these projections for air quality in National Parks and wilderness areas.

We find that, if stringent domestic emission controls are applied in the future such as those projected by the RCP4.5 and RCP8.5 scenarios, air quality is predicted to improve significantly across the U.S., except surface O_3 in the central U.S. under RCP8.5 conditions. We estimate that $PM_{2.5}$ concentrations in the majority of the U.S. NPs and wilderness areas will be substantially reduced, below the annual U.S. EPA NAAQS of 12 μg/m^3. In addition, visibility will be in general significantly improved. Over the eastern U.S., we estimate that most of the parks will reach the 2050 target to restore visibility to natural conditions by 2064, whereas some parks may not reach this target during most polluted episodes over the western U.S. (e.g., Yellowstone and Grand Canyon NP). This result suggests that, to obtain acceptable future visibility conditions over this region, the U.S. National Park Service may have to develop specific air quality management plans to include further mitigation strategies beyond those projected by the RCP scenarios.

Our analysis shows that climate-driven fires may dominate summertime $PM_{2.5}$ concentrations in the future over the western U.S., potentially offsetting the large $PM_{2.5}$ reductions from anthropogenic emission controls. Future regional visibility is also estimated to be impaired by fire pollution, which may keep visibility at present-day levels during the most polluted episodes in many parks (e.g., Crater Lake NP). However, our analysis has important limitations. For example, it considers an average fire emission projection based on SRES A1B climate and applies this projection homogeneously to all the fire species on a monthly basis and with the spatial distribution formulated by the RCP fire emission database. More work is needed to directly couple climate-driven fire emissions, vegetation dynamics, and air quality.

We find that daily surface O_3 is projected to drop in all U.S. NPs and wilderness areas in the RCP4.5 scenario, with MDA-8 levels below the primary U.S. EPA NAAQS of 75 ppb to protect human health, and even below 65 ppb, a level considered for future regulation. In contrast, our projections with the RCP8.5 scenario indicate that numerous parks in the western and central U.S. are predicted to have a poorer O_3 air quality, with MDA-8 above 65 ppb in some cases during the summer (e.g., Rocky Mountain and Yellowstone NP). In this case, the rising O_3 resulting from a growing O_3 background associated with increases in CH_4 levels ($\sim$1000 ppb) as well as climate and land use changes exceeds the important surface O_3 reductions projected from anthropogenic emission controls. Furthermore, despite the substantial general decrease in surface O_3, our study ~~indicates~~ suggests that the secondary standard W126 may remain above the recommended limits (7–15 ppm-hrs) to protect vegetation in many regions across the United States. Thus, future O_3 pollution may be a threat to the U.S. NP ecosystems. In the U.S., W126 levels are most sensitive to domestic anthropogenic NO_x emissions (Lapina et al., 2014) and our results suggest that more restricted policies for NO_x control may be needed to preserve natural ecosystems in the U.S. NPs and wilderness areas.

Our results suggest that 2050 air quality in the U.S. will likely be dominated by anthropogenic emission trajectories. Changes in air quality driven by climate and land use are small over the 50-year time horizon studied here and they are not always significant. However, climate alone can lead to a substantial increase in surface MDA-8 O_3 by 2050 over most of the U.S. with important implications for O_3 air quality and ecosystem health degradation at the U.S. National Parks. Projected changes in temperature, cloud cover, and biogenic emissions suggest that these drivers may exacerbate future O_3 pollution across the United States. Furthermore, land use change may have an important regional effect on surface O_3, due to changes in biogenic emissions and dry deposition. Our study suggests that the effects of climate, vegetation, and fires are important in future air quality projections and these processes should be considered in air quality management and planning in the coming decades.

Acknowledgements. This work was supported by the U.S. National Park Service (grant H2370 094000/J2350103006), the U.S. National Science Foundation (AGS-1238109), and the JFSP (project ID 13-1-01-4). We thank Xu Yue (Yale University) and Jenny Hand (CIRAS-CSU) for helpful discussions and Samuel Albani (Cornell University) for the updated dust scheme. The CESM project is supported by the National Science Foundation and the Office of Science (BER) of the U.S. Department of Energy. Computing resources were provided by the Climate Simulation Laboratory at NCAR's Computational and Information Systems Laboratory (CISL), sponsored by the National Science Foundation and other agencies. The National Center for Atmospheric Research is operated by the University Corporation for Atmospheric Research under sponsorship of the National Science Foundation. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the National Science Foundation.

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Table 1. Summary of main RCP4.5 and RCP8.5 anthropogenic greenhouse gas concentrations and global area-averaged sea-surface temperature (SST) for 2000 and 2050.

Year	Scenario	~~Year Scenario~~ CO_2 (ppm)	CH_4 (ppb)	N_2~~0~~O (ppb)	SST (°C)
2000	Baseline	367	1760	316	12.2
2050	RCP4.5	487	1833	350	12.6
	RCP8.5	541	2740	367	13.0

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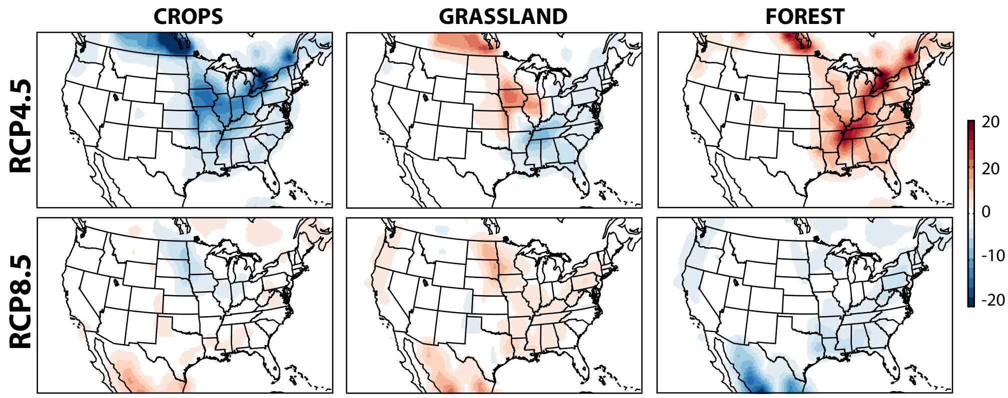

Figure 1. Projected 2050-2000 changes (%) in forest, grasslands and croplands by the RCP4.5 and RCP8.5 scenarios.

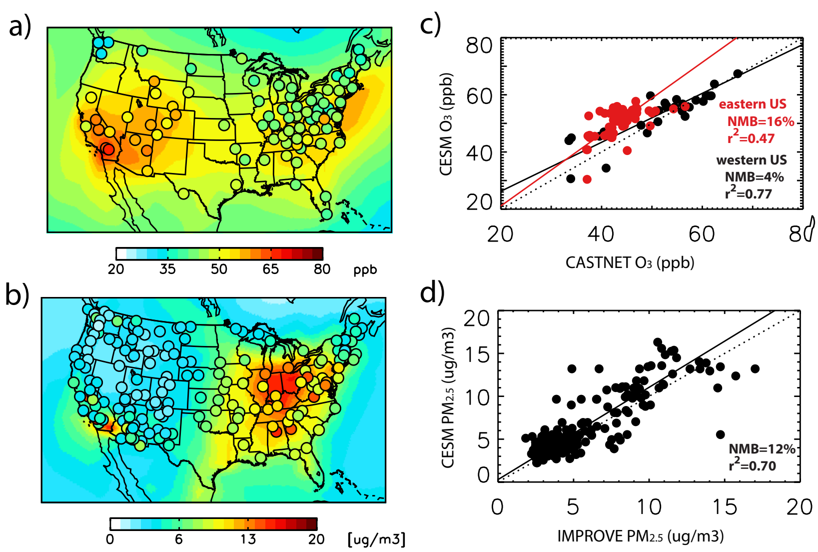

Figure 2. Simulated and observed present-day surface MDA-8 O_3 and $PM_{2.5}$ (a,b) and the scatter plots with modeled and observed values at the individual sites (c,d). Observations are long-term means (1998–2010) from the CASTNET (1995–2005) and IMPROVE (1998–2010) networks. The squared-correlation coefficients (r^2) and normalized mean biases (NMB) are shown in the inset. Reduced-major axis regression lines (solid) and the 1:1 lines (dash) are also shown. Maps show interpolated contours from the 1.9x2.5 degree horizontal resolution output.

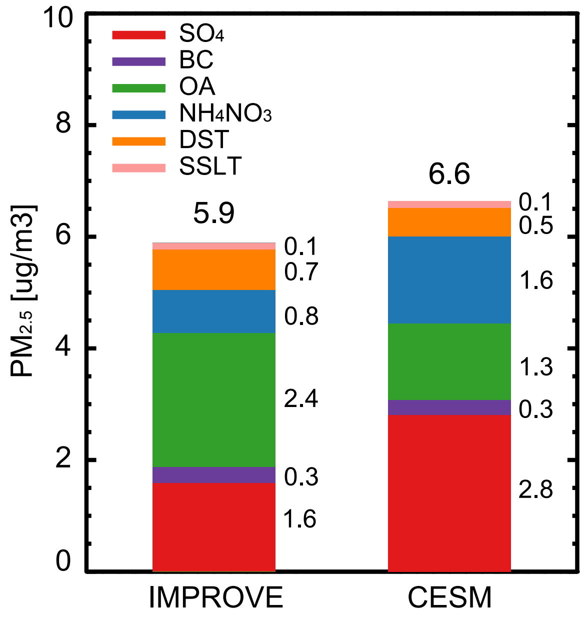

Figure 3. Simulated and observed $PM_{2.5}$ chemical ~~speciation~~ species over the United States. Big numerals indicate the annual $PM_{2.5}$ concentrations, whereas small numerals indicate $PM_{2.5}$ chemical species concentrations.

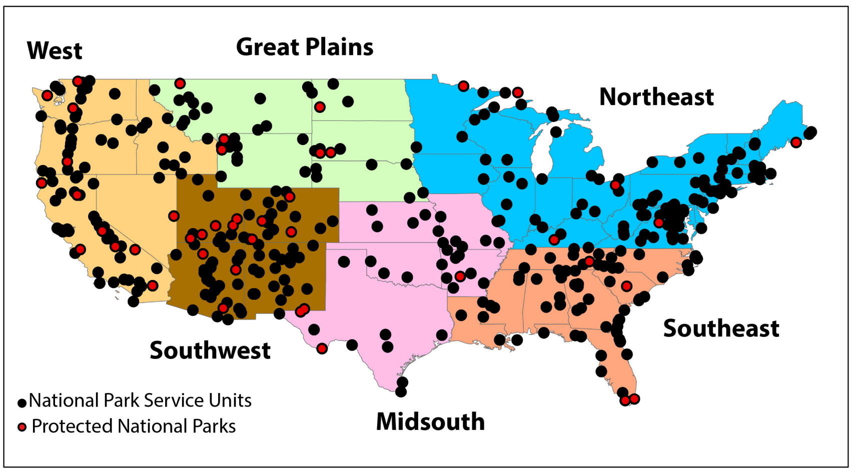

Figure 4. Location of the U.S. National Park units and wilderness areas used in this study. The U.S. protected National Parks are highlighted in red; the six U.S. climatic regions are also identified.

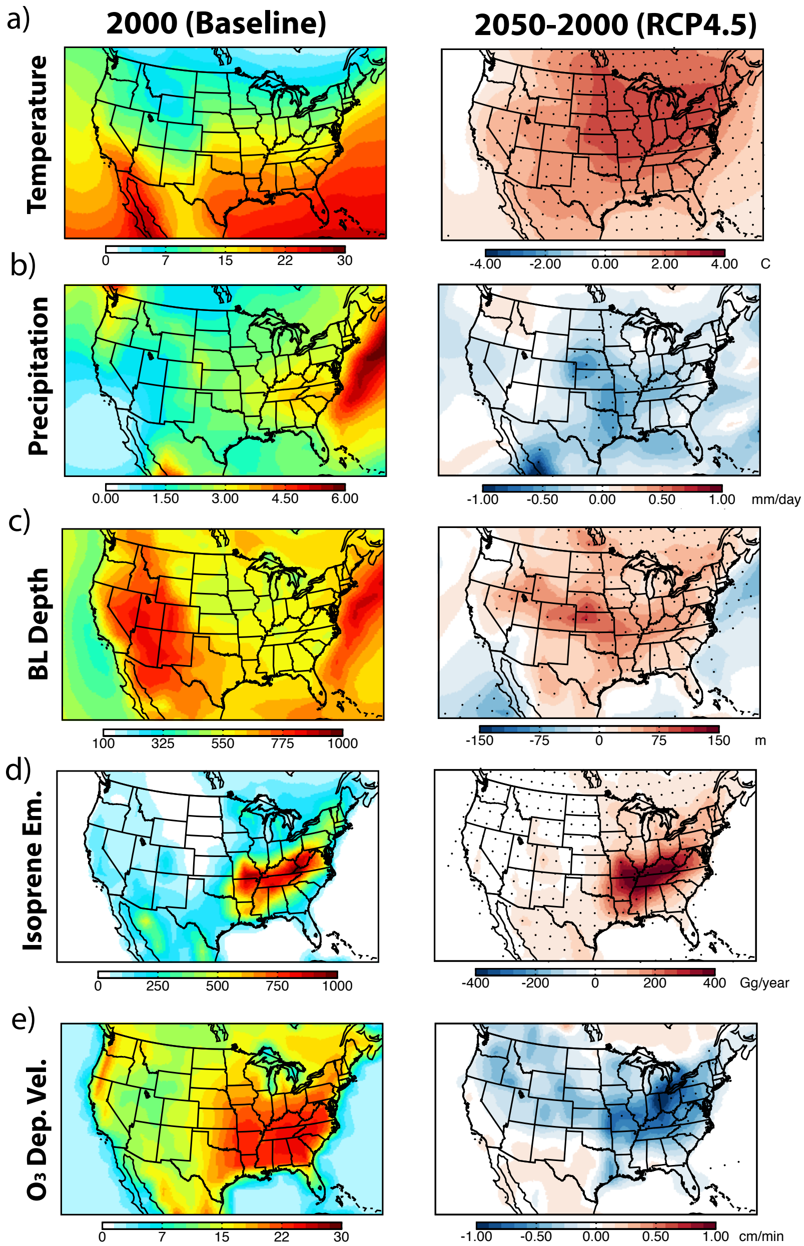

Figure 5. Simulated annual average present-day (left) and projected 2050–2000 changes (right) for surface temperature (a), precipitation (b), boundary layer depth (c), isoprene emissions (d) and O_3 dry deposition velocity (e). All maps show changes predicted by the RCP4.5 as a result of the combination of climate, land use and emissions changes, except for the O_3 dry deposition velocity that shows only the changes from land use~~changes~~. Regions with changes that are significant at the 95% confidence level are indicated in the maps with dots, and maps show interpolated contours from the 1.9x2.5 degree horizontal resolution output.

Table 2. Anthropogenic short-lived air pollutants and biogenic emissions in 2000 and 2050, projected by the RCP4.5 and RCP8.5 scenarios over the United States.

				Anthropogenic Emissions[a]					Biogenic Emissions[b]	
Year	Scenario	BC	OC	CO	NO_x	NH_3	NMVOCs	SO_2	Isoprene	Monoterpenes
				eastern U.S.						
2000	Baseline	0.29	0.41	70.38	4.82	1.53	1.96	6.30	20.5	6.3
2050	RCP4.5	0.15	0.19	8.70	1.04	1.92	0.90	0.80	33.7	9.4
2050	RCP8.5	0.03	0.06	7.99	1.35	2.21	0.36	0.63	27.8	8.0
				western U.S.						
2000	Baseline	0.10	0.15	25.82	1.58	1.36	0.64	1.77	7.4	2.5
2050	RCP4.5	0.05	0.08	3.55	0.39	1.81	0.30	0.29	9.4	3.0
2050	RCP8.5	0.02	0.03	7.20	0.79	2.16	0.15	0.54	9.6	3.3

[a] Reported Tg C/year for BC, OC and NMVOCs; Tg N/year for NO_x and NH_3; Tg S/year for SO_2; and Tg CO/year for CO.
[b] Reported Tg C/year.

Table 3. List of simulations[a]

Forcings	2000 Baseline	2050 Total	2050 Climate	2050 Emissions	2050 Land Use
Climate	2000	2050	2050	2000	2000
Emissions[b]:					
Anthropogenic	2000	2050	2000	2050	2000
BB	2000	2050	2000	2050	2000
Biogenic	2000	2050	2000	2000	2050
Land Use[c]	2000	2050	2000	2000	2050
Methane	2000	2050	2000	2050	2000

[a] Years represent the year forcing parameter selected for each simulation.
[b] Anthropogenic is the RCP surface and ship emissions, BB is the RCP biomass burning emissions and are considered anthropogenic impact; Biogenic is biogenic emissions calculated by MEGAN v2.1 (see text for further explanation).
[c] Land is the human induced land cover and land use projected by the RCP scenarios.

Table 4. Simulated annual air quality over the U.S. National Parks and wilderness areas[a]

	$PM_{2.5}$ (μg/m^3)			MDA-8 O_3 (ppb)			W126 O_3 (ppm-hr)		
	2000	2050	2050	2000	2050	2050	2000	2050	2050
National Park	Base	RCP4.5	RCP8.5	Base	RCP4.5	RCP8.5	Base	RCP4.5	RCP8.5
Acadia, ME (44° N, 68° W)	4.3	2.2	2.2	48.0	43.9	48.8	13.1	2.5	4.7
Arches, UT (39° N, 110° W)	3.2	1.6	2.6	57.8	51.2	60.8	39.9	10.4	39.1
Badlands, SD (44° N, 102° W)	4.1	1.7	3.1	49.4	47.8	55.1	15.9	11.5	29.2
Big Bend, TX (29° N, 103° W)	5.2	2.9	3.8	47.0	42.9	49.4	5.9	2.9	8.5
Biscayne, FL (26° N, 80° W)	5.9	3.9	3.3	45.7	40.4	45.6	1.1	0.5	1.0
Black Canyon, CO (39° N, 108° W)	3.3	1.8	2.8	57.0	50.9	60.4	34.5	8.7	34.6
Bryce Canyon, UT (38° N, 112° W)	4.1	2.1	3.2	58.7	51.7	59.7	45.8	12.1	33.3
Canyonlands, UT (38° N, 110° W)	3.2	1.6	2.6	57.8	51.2	60.8	39.9	10.4	39.1
Capitol Reef, UT (38° N, 111° W)	3.2	1.6	2.6	57.8	51.2	60.8	39.9	10.4	39.1
Carlsbad Caverns, NM (32° N, 104° W)	5.0	2.6	3.2	50.7	45.5	52.3	13.4	4.9	13.1
Channel Islands, CA (34° N, 119° W)	8.6	6.2	5.6	52.4	50.2	55.5	12.2	4.6	9.6
Congaree, SC (34° N, 81° W)	10.2	4.6	4.7	53.6	46.3	52.3	23.4	4.2	11.8
Crater Lake, OR (43° N, 122° W)	4.2	4.2	3.0	50.1	46.6	52.2	11.9	3.1	5.4
Cuyahoga Valley, OH (41° N, 82° W)	15.9	5.8	5.2	53.2	49.0	52.3	61.3	21.7	28.4
Death Valley, CA (36° N, 117° W)	3.4	2.1	2.1	58.9	52.7	58.9	45.5	15.1	28.3
Dry Tortugas, FL (25N,83W)	6.0	4.2	4.0	40.8	38.4	44.6	0.6	0.5	1.2
Everglades, FL (25° N, 81° W)	5.9	3.9	3.3	45.7	40.4	45.6	1.1	0.5	1.0
Glacier, MT (49° N, 114° W)	3.1	2.6	2.5	48.5	46.8	52.7	9.5	4.2	9.7
Grand Canyon, AZ (36° N, 113° W)	4.1	2.1	3.2	58.7	51.7	59.7	45.8	12.1	33.3
Grand Teton, WY (44° N, 111° W)	2.2	1.4	1.8	53.8	50.8	58.2	18.1	8.3	22.8
Great Basin, NV (39° N, 114° W)	2.5	1.5	1.7	57.0	51.6	59.0	34.7	11.5	28.2
Great Sand Dunes, CO (38° N, 105° W)	4.2	2.0	3.3	55.9	49.7	59.1	29.7	8.5	32.1
Great Smoky Mountains, NC, TN (36° N, 83° W)	10.9	5.6	4.2	55.7	46.4	51.8	43.8	5.7	14.9
Guadalupe Mountains, TX (32° N, 105° W)	5.0	2.6	3.2	50.7	45.5	52.3	13.4	4.9	13.1
Hot Springs, AR (34° N, 93° W)	10.8	4.7	5.5	53.0	43.9	51.0	32.0	3.9	13.3
Isle Royale, MI (48° N, 88° W)	3.7	2.6	3.0	43.3	42.8	47.7	4.8	2.8	6.5
Joshua Tree, CA (34° N, 116° W)	16.9	13.4	13.9	62.3	53.7	58.4	57.9	20.2	28.2
Kings Canyon, CA (37° N, 118° W)	3.4	2.1	2.1	58.9	52.7	58.9	45.5	15.1	28.3
Lassen Volcanic, CA (40° N, 121° W)	4.7	5.2	3.6	51.2	48.0	54.4	14.0	4.0	9.3
Mammoth Cave, KY (37° N, 86° W)	15.1	6.4	6.2	54.5	46.4	52.4	49.1	7.6	22.6
Mesa Verde, CO (37° N, 108° W)	4.6	2.1	3.9	57.8	50.5	60.6	40.8	8.8	38.6
Mount Rainier, WA (47° N, 122° W)	5.1	3.4	2.6	45.9	43.4	47.8	5.2	1.0	1.6
North Cascades, WA (49° N, 121° W)	4.9	3.2	2.4	45.2	43.3	47.7	6.3	1.2	1.9
Olympic, WA (48° N, 123° W)	4.9	3.2	2.4	45.2	43.3	47.7	6.3	1.2	1.9
Petrified Forest, AZ (35° N, 110° W)	5.5	2.6	4.4	58.2	50.3	59.3	44.0	10.2	34.6
Redwood, CA (41° N, 124° W)	3.4	3.4	2.6	44.9	44.4	49.5	1.2	0.8	1.4
Rocky Mountain, CO (40° N, 106° W)	4.6	2.0	3.1	56.8	51.8	60.0	37.9	13.6	36.4
Saguaro, AZ (32° N, 110° W)	6.1	3.3	4.3	57.8	49.6	56.3	45.0	10.0	23.0
Sequoia, CA (36° N, 119° W)	3.4	2.1	2.1	58.9	52.7	58.9	45.5	15.1	28.3
Shenandoah, VA (38° N, 78° W)	13.2	6.2	4.0	57.0	49.0	51.7	66.5	11.7	13.3
Theodore Roosevelt, ND (47° N, 103° W)	4.8	1.8	3.6	47.8	46.8	53.6	16.2	11.0	29.3
Voyageurs, MN (48° N, 93° W)	4.1	2.3	3.2	43.5	42.9	48.0	5.7	3.0	7.4
Wind Cave, SD (44° N, 103° W)	4.1	1.7	3.1	49.4	47.8	55.1	15.9	11.5	29.2
Yellowstone, WY, MT, ID (45° N, 110° W)	2.2	1.4	1.8	53.8	50.8	58.2	18.1	8.3	22.8
Yosemite, CA (38° N, 119° W)	5.5	3.3	2.7	60.1	52.8	58.5	60.4	18.9	29.1
Zion, UT (37° N, 113° W)	4.1	2.1	3.2	58.7	51.7	59.7	45.8	12.1	33.3

[a] Shown only results for the 46 protected National Parks located in the continental United States; Results from other NPs and wilderness areas can be provided by request.

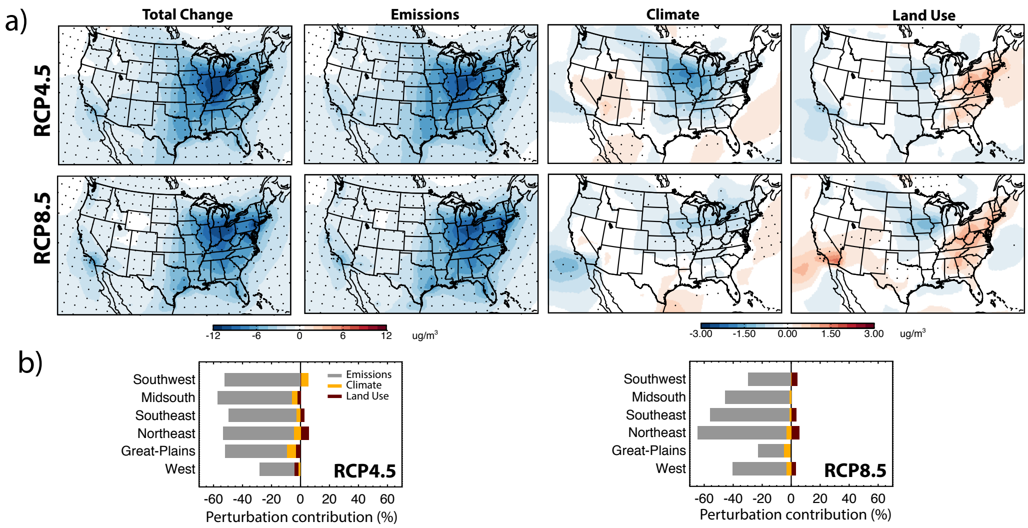

Figure 6. Projected simulated 2050–2000 changes in annual $PM_{2.5}$ as a result of the combination of climate, land use and emissions changes, and the individual changes (a), and the percentage contribution of the individual perturbation (b) , for the RCP4.5 and RCP8.5 scenarios. Regions with changes that are significant at the 95% confidence level are indicated in the maps with dots, and maps show interpolated contours from the 1.9x2.5 degree horizontal resolution output. Bars represent the changes (in %) for each individual forcing, i.e., emissions and methane levels (grey), climate (yellow) and land use (dark red).

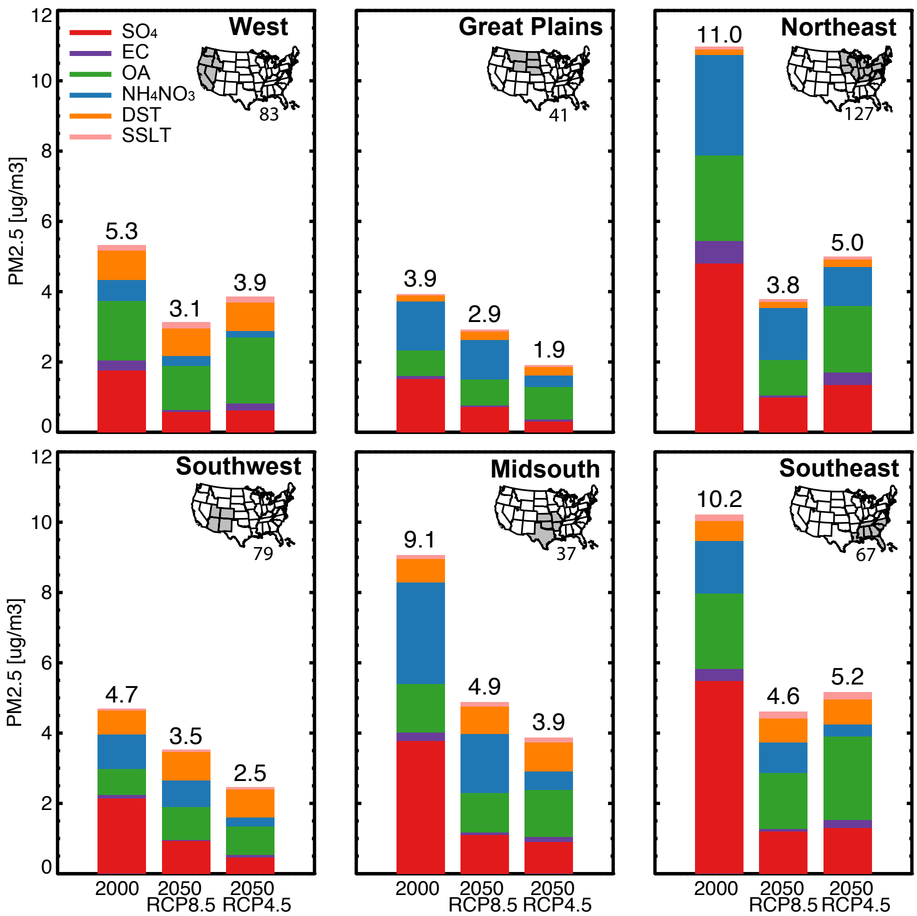

Figure 7. Annual $PM_{2.5}$ chemical ~~speciation~~ species for present-day and 2050 as predicted by the RCP4.5 and RCP8.5 scenarios in the U.S. climatic regions. The inset maps show the states in the region in gray, and the numerals indicate the numbers of U.S. National Parks and wilderness areas in each climatic region. Big numerals indicate the annual $PM_{2.5}$ concentrations.

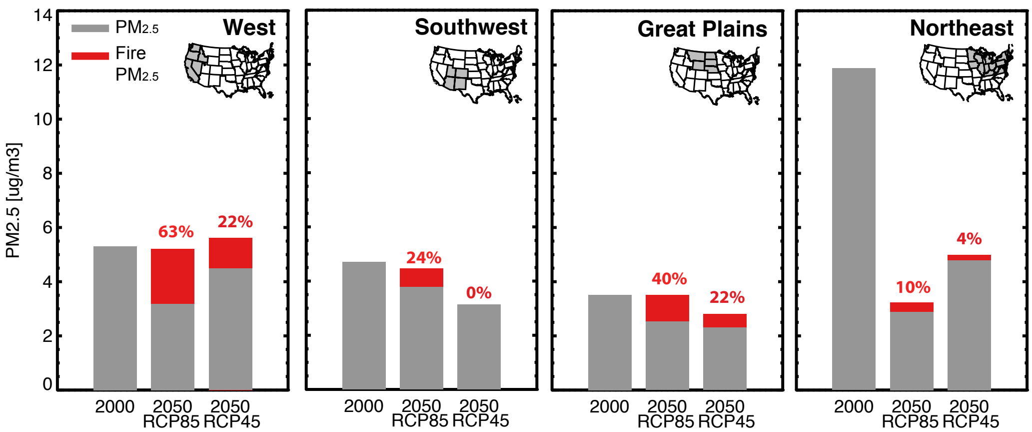

Figure 8. Changes in $PM_{2.5}$ resulting from climate-driven fire activity in the U.S. regions affected by fire. Simulated $PM_{2.5}$ by the RCP scenarios is shown in gray and future $PM_{2.5}$ from climate-driven fire emissions in red. The inset maps show the states in the region in gray and the red numerals indicate the percentage change in $PM_{2.5}$ when climate-driven fire activity is included in the simulation.

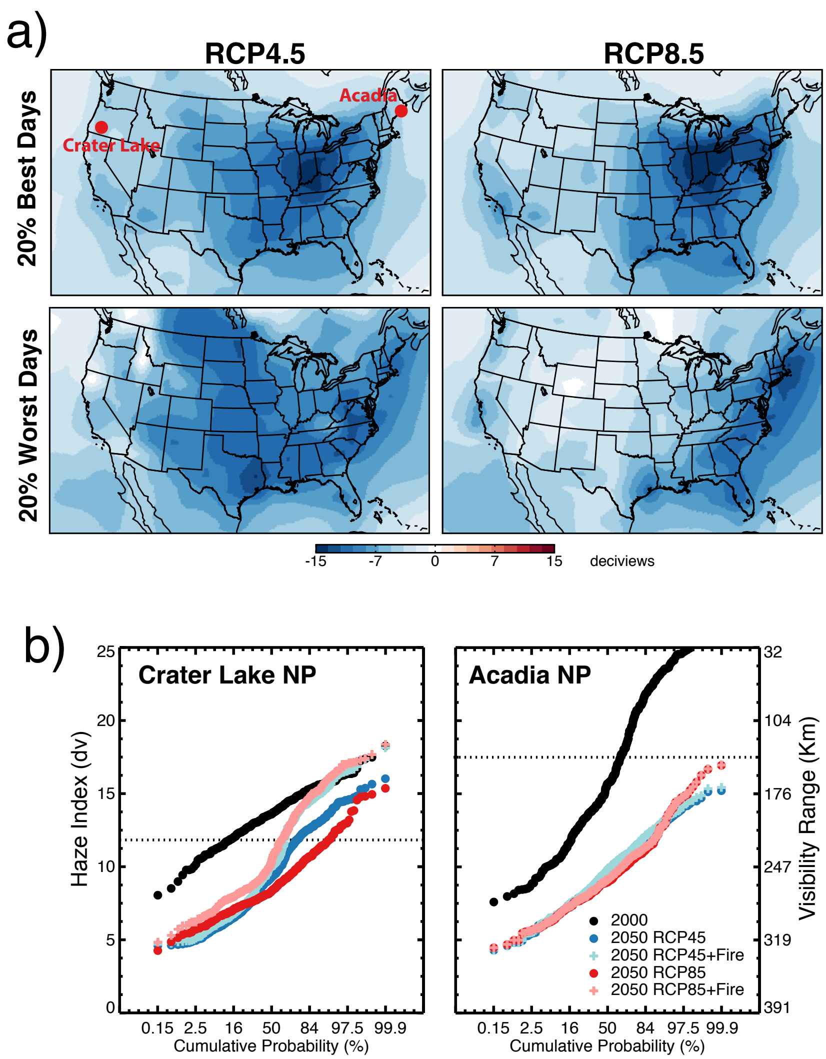

Figure 9. Projected simulated 2050–2000 changes in haze index (HI) as a result of the combination of climate, land use and emissions changes (a) and the cumulative probability distributions of daily mean haze index in the Crater Lake and Acadia NPs (b). ~~The maps~~ Maps show "20% Best Days" as the averaged HI during the cleanest days and "20% Worst Days" as averaged HI during the haziest days (see text for further explanation); data are shown as interpolated contours from the 1.9°x2.5° horizontal resolution output. The location of the Crater Lake and Acadia NPs are indicated in the top left map. The cumulative distribution plots show simulated daily HI for present-day (black circles), 2050 projected by RCP4.5 (blue circles) and by RCP8.5 (red circles), and 2050 with the effects of climate-driven fires by RCP4.5 (light blue cross) and by RCP8.5 (light red cross). The 2050 HI target to reach natural visibility conditions by 2064 are indicated with a horizontal dotted line.

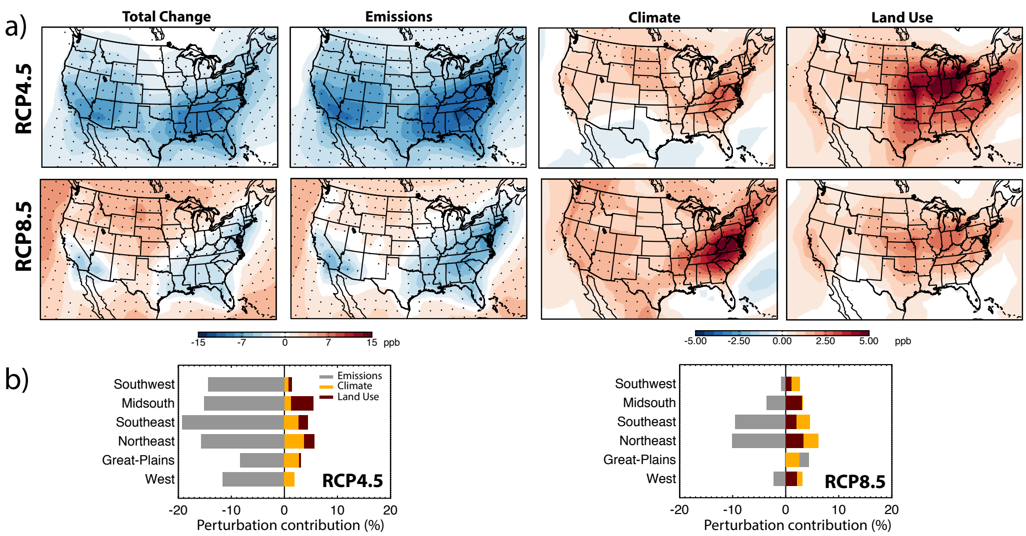

Figure 10. Projected simulated 2050–2000 changes in surface O_3 as a result of the combination of climate, land use and emissions changes, and the individual changes (a) and the percentage contribution of the individual perturbations (b) , for the RCP4.5 and RCP8.5 scenarios. Maps show interpolated contours from the 1.9x2.5 degree horizontal resolution output. Regions with changes that are significant at the 95% confidence level are indicated in the maps with dots, and O_3 concentrations are annual maximum daily 8-hour (MDA-8) averages. Bars represent the changes (in %) for each individual forcing, i.e., emissions and methane levels (grey), climate (yellow) and land use (dark red).

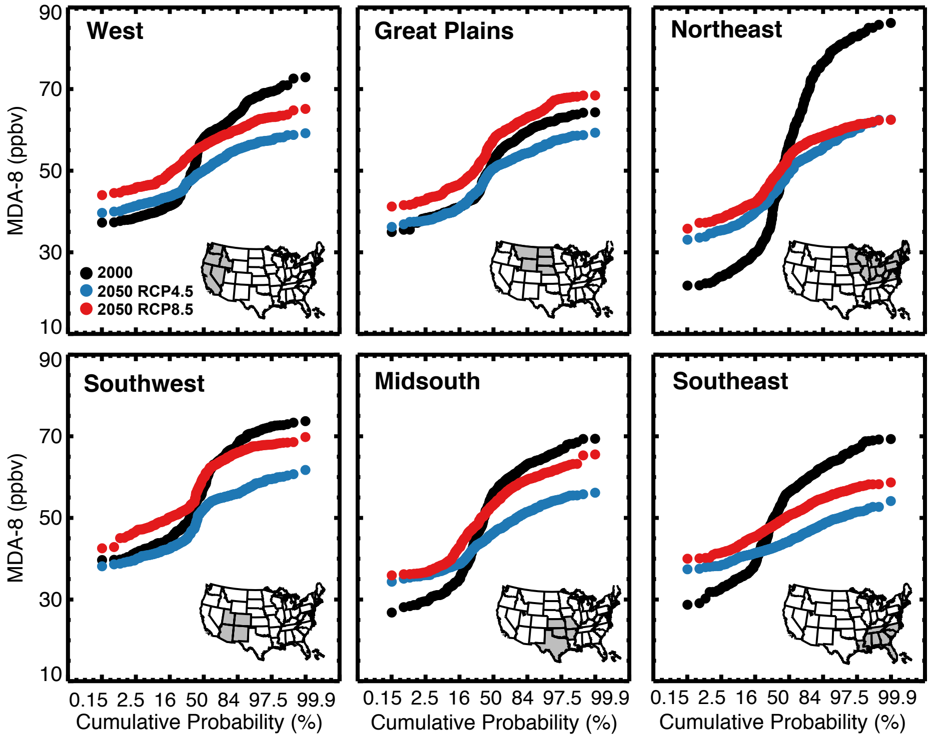

Figure 11. Cumulative probability distributions of simulated surface O_3 MDA-8 averaged over the U.S. National Parks and wilderness areas in the U.S. climatic regions, for present-day (black circles) and 2050 predicted by the RCP4.5 (blue circles) and RCP8.5 (red circles) scenarios. The inset maps show the states in the region in gray.

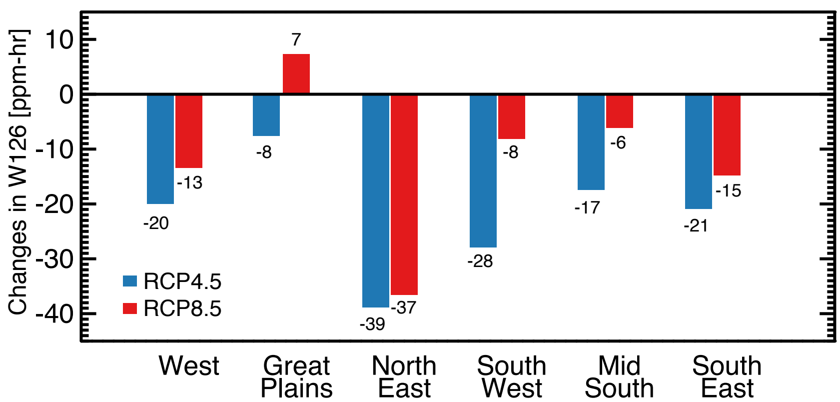

Figure 12. Simulated 2050–2000 summertime ~~W126~~ changes in O_3 ~~in 2000 (black) and 2050~~ W126 for RCP4.5 (blue) and RCP8.5 (red) averaged over the six U.S. climatic regions identified in Figure 4. Numerals indicate the simulated changes in O_3 W126 ~~value, and grey shaded area represent the minimum and maximum recommended standard~~ (~~7–15 ppm-hours~~ppm-hr).