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# 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 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 US, except surface  $O_3$  over the western and central US under RCP8.5 conditions, where rising background ozone counteracts domestic emissions reductions. Under the RCP4.5, surface  $O_3$  is substantially reduced (about 5 ppb), with daily maximum 8 h averages below the primary US EPA NAAQS of 75 ppb (and even 65 ppb) in all the NPs.  $PM_{2.5}$  is significantly reduced in both scenarios ( $4 \mu\text{g m}^{-3}$ ;  $\sim 50\%$ ), with levels below the annual US EPA NAAQS of  $12 \mu\text{g m}^{-3}$  across all the NPs; visibility is also improved (10–15 deciviews;  $> 75$  km in visibility range), although some parks over the western US (40–74 % of total sites in the US) may not reach the 2050 target to restore visibility to natural conditions by 2064. We estimate that climate-driven increases in fire activity may dominate summertime  $PM_{2.5}$  over the western US, 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 anthropogenic emission patterns. However, climate and land use changes alone may lead to a substantial increase in surface  $O_3$  (2–3 ppb) with important consequences for  $O_3$  air quality and ecosystem degradation at the US 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  $< 2.5 \mu\text{m}$ ;  $PM_{2.5}$ ), has evolved in both urban and rural regions around the world over

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to the US National Park Service for air quality management. Surface O<sub>3</sub> is toxic to humans and thus poses a threat to visitor and park staff health. In addition, accumulated exposure to elevated levels of O<sub>3</sub> can damage vegetation (e.g. 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 (e.g. 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 US and needleleaf evergreen forest in the western US (e.g. Arbaugh et al., 1998; Schaub et al., 2005). In addition, rising O<sub>3</sub> 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<sub>2.5</sub> 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 (e.g. Pope and Dockery, 2006). Fine particles and gases cause haze, which degrades visibility. The importance of visibility at NPs and wilderness areas is recognized and protected by the US Environmental Protection Agency's (EPA) Regional Haze Rule (RHR), which establishes the goal of returning visibility to natural conditions. 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 US (e.g. 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 pro-



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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). 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 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 US and Europe in CAM-Chem documented by Lamarque et al. (2012); we discuss this further in Sect. 2.2.

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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 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  $8.5 \text{ Wm}^{-2}$ ) associated with greenhouse gases: RCP2.6, RCP4.5, RCP6.0 and RCP8.5, respectively. The RCP2.6 assumes a peak forcing ( $3.0 \text{ Wm}^{-2}$ ) 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  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  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 US because of the different emission patterns. Emissions of  $\text{NO}_x$ ,  $\text{NH}_3$ ,  $\text{CO}$ , non-methane volatile organic compounds (VOCs),  $\text{SO}_2$  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 and vegetation density (Heald et al., 2008). Both dust and seasalt are also emitted interactively in CESM (Mahowald et al., 2006a, b; Albani et al.,









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<sub>3</sub> 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 US is predicted to generally increase, with the largest increase over in central US (> 100 m) (Fig. 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<sub>2.5</sub> and O<sub>3</sub> concentrations) and precipitation (and cloud cover) decreases (i.e., favoring high PM<sub>2.5</sub> and O<sub>3</sub> concentrations) are generally co-located. These two processes have opposite effects on air quality and this highlights the difficulty of predicting possible air quality impact resulting from climate change.

Higher temperature and solar radiation will also affect biogenic emissions, which in turn will influence PM<sub>2.5</sub> and surface O<sub>3</sub>. Biogenic emissions will also depend on land use changes. In 2050, isoprene emissions are predicted to increase from 28 to 43 Tg C in the US (Table 2), with 10% of this increase driven by land use changes. This effect is more significant in the southeastern US (about 25%) due to afforestation (Fig. 5d). The RCP8.5 scenario also predicts an increase in biogenic emissions, but with a lower influence from land use and climate changes (2%; not shown). We note that our isoprene emissions are slightly overestimated because they do not include the effect of CO<sub>2</sub>, which suppresses isoprene production at elevated levels (e.g. Heald et al., 2008). The impact of changing biogenic emissions on surface O<sub>3</sub> depends critically on the fate of isoprene nitrates, i.e., whether isoprene nitrate is a terminal or temporal sink of NO<sub>x</sub> (e.g. Horowitz et al., 2007; Wu et al., 2012). In our model, isoprene nitrate recycles 40% of NO<sub>x</sub> (Horowitz et al., 2007). Therefore, increases in biogenic emissions tend to enhance surface O<sub>3</sub>. In addition, increased biogenic volatile organic compounds (e.g. isoprene) will lead to increases in PM<sub>2.5</sub> through SOA formation (Heald et al., 2008).

Land use changes can also influence deposition processes. For example, large O<sub>3</sub> dry deposition velocities are associated with denser, broadleaf forests (i.e., with high

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LAI) and crops (e.g. 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 small decrease across the US ( $0.2\text{--}1.0\text{ cm min}^{-1}$ ; about 1–3 %) (Fig. 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, 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.

## 4 Future $PM_{2.5}$ air quality

In this section, we first examine how  $PM_{2.5}$  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 the “land use” simula-



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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  $\text{SO}_4$ ,  $\text{NH}_4\text{NO}_3$  and BC. However,  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  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 Sect. 3.

Our results are consistent with previous studies, which have shown the small impact of climate change on  $\text{PM}_{2.5}$  levels and the significant contribution from projected emissions reductions (e.g. Tagaris et al., 2007; Pye et al., 2009; Lam et al., 2011; Kelly et al., 2012). Comparing  $\text{PM}_{2.5}$  projections from different studies is not straightforward due to variations in the study region, reported  $\text{PM}_{2.5}$  metrics and use of different climate and emissions (Fiore et al., 2012). A decrease of about  $2 \mu\text{g m}^{-3}$  (25 %) over the US 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 ( $4\text{--}5 \mu\text{g m}^{-3}$ ), with 90 % of the reduction due to emission reductions. Most recently, Kelly et al. (2012) reported summertime regional decreases of more than  $3 \mu\text{g m}^{-3}$  over the US, with the SRES A2 climate and RCP6.0 emission scenarios.

We summarize the simulated  $\text{PM}_{2.5}$  changes over the US 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  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$ , but concentrations are predicted to remain below  $12 \mu\text{g m}^{-3}$ , the primary annual US National Ambient Air Quality Standards (NAAQS) for  $\text{PM}_{2.5}$ . In the Joshua Tree NP, both RCP scenarios predict a significant improvement of  $\text{PM}_{2.5}$  air quality, but with an annual average above  $12 \mu\text{g m}^{-3}$  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<sub>2.5</sub> levels associated with climate change, as discussed in the following section.

## 4.2 Effects of increased fire activity on summertime PM<sub>2.5</sub>

Climate-driven changes in fire emissions can be an important factor controlling PM<sub>2.5</sub> 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 US 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 US, 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<sub>2.5</sub>, we perform an additional simulation (not shown in Table 3), where we increase the RCP fire emissions over the US 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 US and Canada, with the exception of the eastern US, 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<sub>2.5</sub>. We focus on the summer here which is the peak fire season in the United States. We compare the PM<sub>2.5</sub> 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<sub>2.5</sub> is affected by fire, i.e., West, Great Plains, Southwest and Northeast. PM<sub>2.5</sub> concentrations in these regions increase significantly as a result of increased fire activity. These

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increases are most prominent over the West and Great Plain regions, in which fire-driven  $\text{PM}_{2.5}$  may potentially offset anticipated reductions in anthropogenic emissions. For example, over the West region we estimate that fire activity may increase future summertime  $\text{PM}_{2.5}$  from  $3.2$  to  $5.2 \mu\text{g m}^{-3}$  (63%) in the RCP8.5 scenario and from  $4.5$  to  $5.6 \mu\text{g m}^{-3}$  (22%) in the RCP4.5 scenario. The concentration of organic aerosol nearly doubles in both scenarios, and this dominates the total change in  $\text{PM}_{2.5}$ . 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  $\text{PM}_{2.5}$  further downwind from the fires. We estimate that summertime  $\text{PM}_{2.5}$  may increase up to 4–10% in the Northeast region due to smoke transported from fires in the western US and the boreal region.

Therefore, changes in summertime  $\text{PM}_{2.5}$  concentrations may be dominated by changes in fire activity in most of the western US in a future climate. This same fire pollution may contribute significantly to impairing 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 US NP and wilderness areas across the continental US 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  $\text{PM}_{2.5}$  chemical species. Figure 9a shows changes in HI for the most polluted and the cleanest episodes 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 Fig. 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 US 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 US 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; Fig. 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

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regions (e.g., Acadia NP; Fig. 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 vs. 400 km) and the spatial distribution of the fire emissions.

## 5 Future changes in surface O<sub>3</sub>

In this section, we first examine future projections on daily surface O<sub>3</sub> concentrations and evaluate the contributing factors to this future change. We then discuss how future changes in surface O<sub>3</sub> may impact ecosystems.

### 5.1 Predictions of daily O<sub>3</sub> concentrations

Figure 10 shows the 2050–2000 changes in annual mean surface O<sub>3</sub> concentrations predicted by the RCP4.5 and RCP8.5 scenarios in the continental United States. As in the PM<sub>2.5</sub> analysis, we present total changes in the simulated daily MDA-8 O<sub>3</sub> concentrations and show the individual perturbations resulting from changing climate, land use, and reducing emission (Fig. 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<sub>3</sub> across the continental US in the RCP4.5 scenario, with the strongest absolute reductions (up to 10 ppb) over the eastern US and California, regions with the highest O<sub>3</sub> concentrations (Fig. 2a) and strongest anthropogenic precursors emissions reductions. The average MDA-8 over the US decreases from 52 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 ppb) over the eastern US and California. The RCP4.5 and RCP8.5 scenarios project

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strong and similar decreases in domestic O<sub>3</sub> precursor emissions (Table 2), however global CH<sub>4</sub> concentrations are 50 % larger in RCP8.5 compared to RCP4.5 (2740 vs. 1838 ppb; Table 1). Rising surface O<sub>3</sub> levels over central US are therefore the result of elevated background O<sub>3</sub> due to rising CH<sub>4</sub> levels in combination with climate and land use changes. These individual effects can be clearly seen in Fig. 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<sub>3</sub> (~ 3 ppb), in which the contribution from emission reductions (−2 %) is counterbalance by climate (+3 %) and land (+2 %) changes.

The impact of the rising background O<sub>3</sub> in the RCP8.5 scenario can also be seen on the surface O<sub>3</sub> concentrations over the ocean. Similar to previous studies (e.g. Wu et al., 2008; Fiore et al., 2012), the RCP4.5 scenario projects a decrease in O<sub>3</sub> levels (up to 5 ppb) over the Pacific and Atlantic oceans in a changing climate due to the decrease of O<sub>3</sub> lifetime associated with higher water vapor. The shorter lifetime of PAN in a future climate may also contribute to the decrease of O<sub>3</sub> levels over remote areas (e.g. Wu et al., 2008; Doherty et al., 2013). By contrast, the RCP8.5 scenario projects an increase in surface O<sub>3</sub> (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<sub>3</sub> 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<sub>3</sub> increases by 1 and 2 ppb across the US in the RCP4.5 and RCP8.5 scenarios respectively, with the largest absolute changes over the eastern US (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<sub>2</sub> 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 compara-







region, the US 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 US, 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 US NPs and wilderness areas in the RCP4.5 scenario, with MDA-8 levels below the primary US 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 US 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 that the secondary standard W126 may remain above the recommended limits (7–15 ppm h) to protect vegetation in many regions across the United States. Thus, future  $O_3$  pollution may be a threat to the US NP ecosystems. In the US, 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 US NPs and wilderness areas.

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Our results suggest that 2050 air quality in the US 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 and they are not always significant. However, climate alone can lead to a substantial increase in surface MDA-8 O<sub>3</sub> by 2050 over most of the US with important implications for O<sub>3</sub> air quality and ecosystem health degradation at the US National Parks. Projected changes in temperature, cloud cover, and biogenic emissions suggest that these drivers may exacerbate future O<sub>3</sub> pollution across the United States. Furthermore, land use change may have an important regional effect on surface O<sub>3</sub>, 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.

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**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.

Year	Scenario	Anthropogenic Emissions <sup>a</sup>						Biogenic Emissions <sup>b</sup>		
		BC	OC	CO	NO <sub>x</sub>	NH <sub>3</sub>	NMVOCs	SO <sub>2</sub>	Isoprene	Monoterpenes
eastern US										
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 US										
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

<sup>a</sup> Reported Tg C year<sup>-1</sup> for BC, OC and NMVOCs; Tg N year<sup>-1</sup> for NO<sub>x</sub> and NH<sub>3</sub>; Tg S year<sup>-1</sup> for SO<sub>2</sub>; and Tg CO year<sup>-1</sup> for CO.

<sup>b</sup> Reported Tg C year<sup>-1</sup>.

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**Table 3.** List of simulations<sup>a</sup>.

Forcings	2000 Baseline	2050 Total	2050 Climate	2050 Emissions	2050 Land Use
Climate	2000	2050	2050	2000	2000
Emissions <sup>b</sup> :					
Anthropogenic	2000	2050	2000	2050	2000
BB	2000	2050	2000	2050	2000
Biogenic	2000	2050	2000	2000	2050
Land Use <sup>c</sup>	2000	2050	2000	2000	2050

<sup>a</sup> Years represent the year forcing parameter selected for each simulation.

<sup>b</sup> 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).

<sup>c</sup> Land is the human induced land cover and land use projected by the RCP scenarios.

**Table 4.** Simulated annual air quality over the US National Parks and wilderness areas<sup>a</sup>.

National Park	PM <sub>2.5</sub> (µg m <sup>-3</sup> )			MDA-8 O <sub>3</sub> (ppb)			W126 O <sub>3</sub> (ppm h)		
	2000 Base	2050 RCP4.5	2050 RCP8.5	2000 Base	2050 RCP4.5	2050 RCP8.5	2000 Base	2050 RCP4.5	2050 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 (25° N, 83° W)	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

<sup>a</sup> 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.

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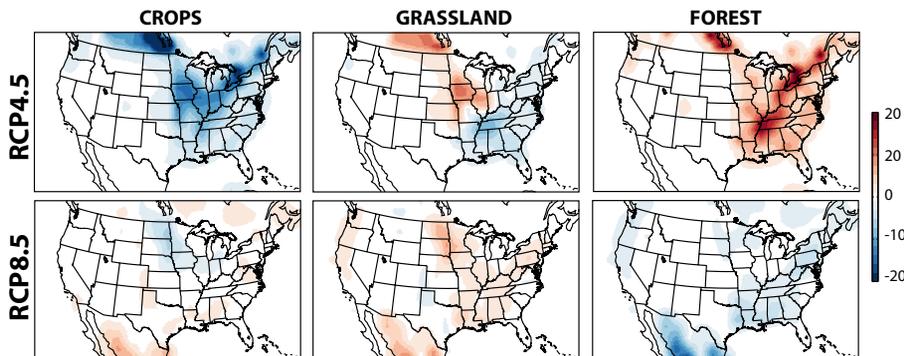
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**Figure 1.** Projected 2050–2000 changes (%) in forest, grasslands and croplands by the RCP4.5 and RCP8.5 scenarios.

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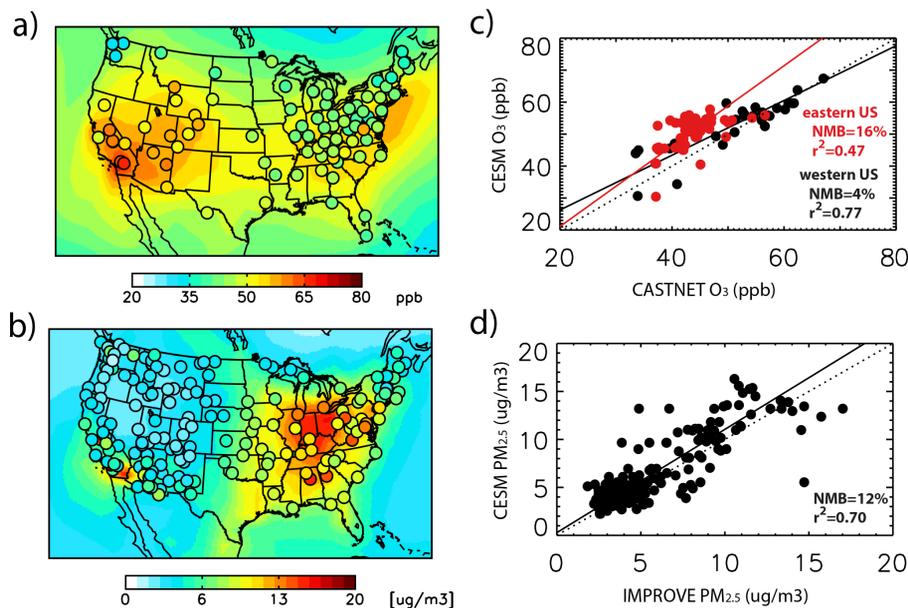
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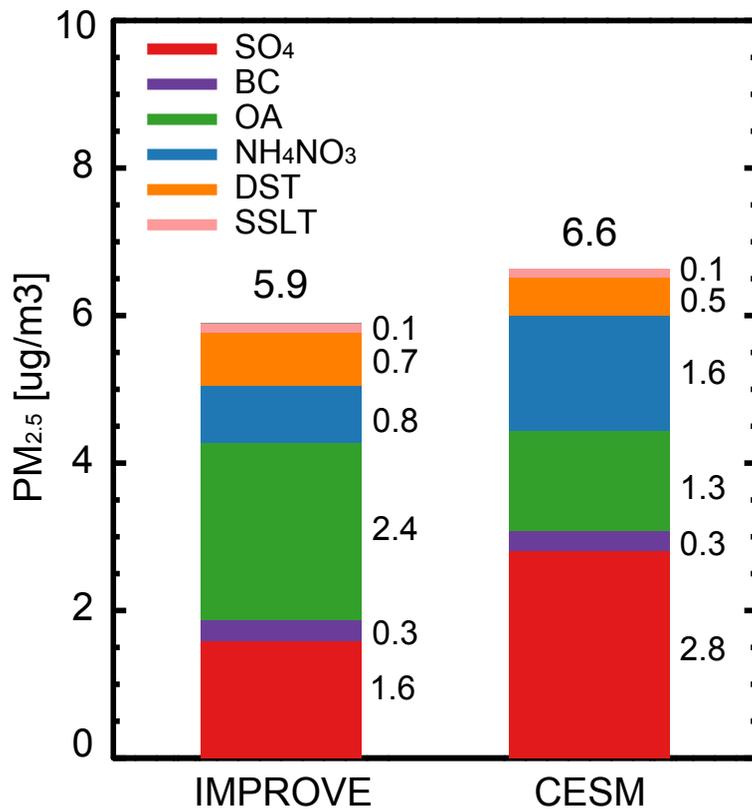


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**Figure 2.** Simulated and observed present-day surface MDA-8 O<sub>3</sub> and PM<sub>2.5</sub> (**a**, **b**) and the scatter plots with modeled and observed values at the individual sites (**c**, **d**). Observations are long-term means (1998–2010) from the CASTNET and IMPROVE 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.



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

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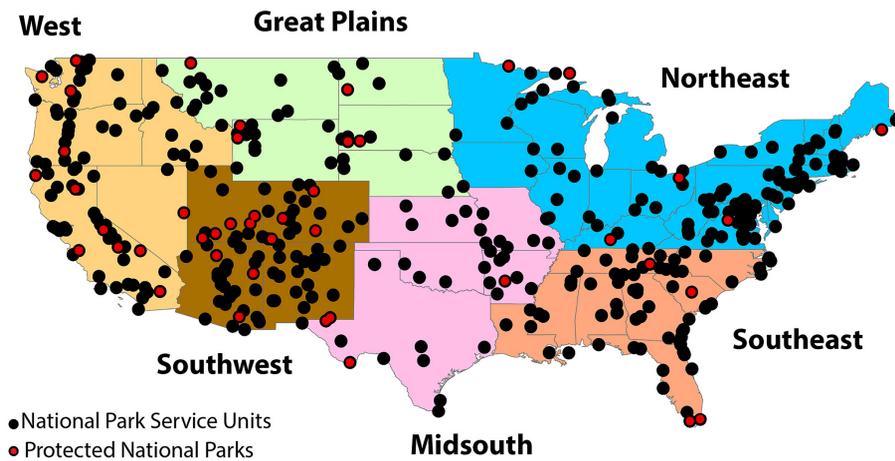
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**Figure 4.** Location of the US National Park units and wilderness areas used in this study. The US protected National Parks are highlighted in red; the six US climatic regions are also identified.

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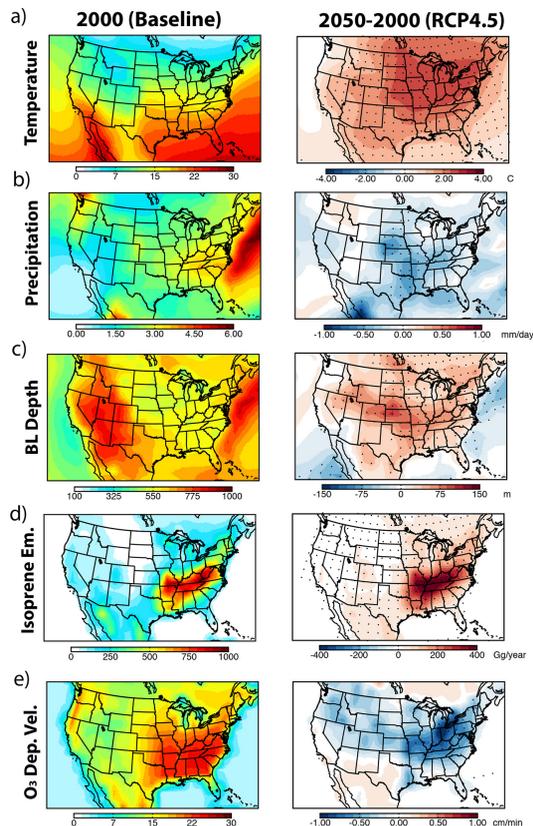
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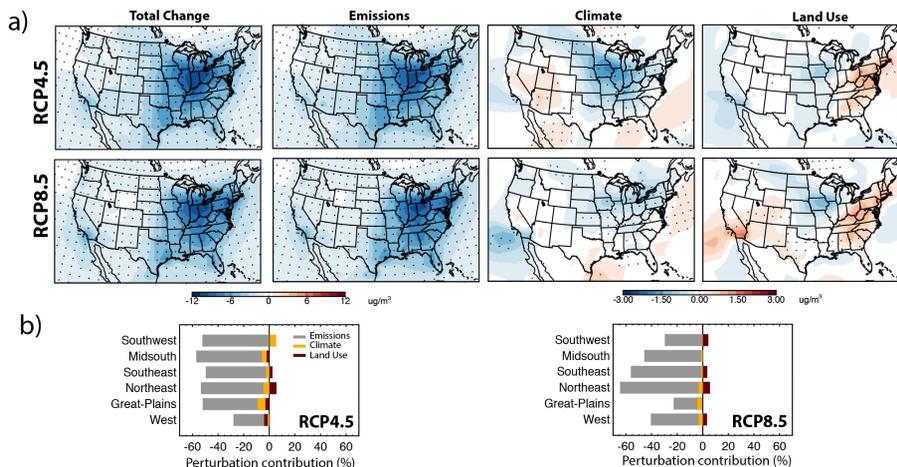
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**Figure 5.** Simulated annual average present-day (left) and projected 2050–2000 changes (right) for surface temperature **(a)**, precipitation **(b)**, boundary layer depth **(c)**, isoprene emissions **(d)** and O<sub>3</sub> dry deposition velocity **(e)**. All maps show changes predicted by the RCP4.5 as a result of the combination of climate, land use and emissions changes, except O<sub>3</sub> dry deposition velocity that shows only land use changes. Regions with changes that are significant at the 95 % confidence level are indicated in the maps with dots.

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**Figure 6.** Projected simulated 2050–2000 changes in annual PM<sub>2.5</sub> 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.

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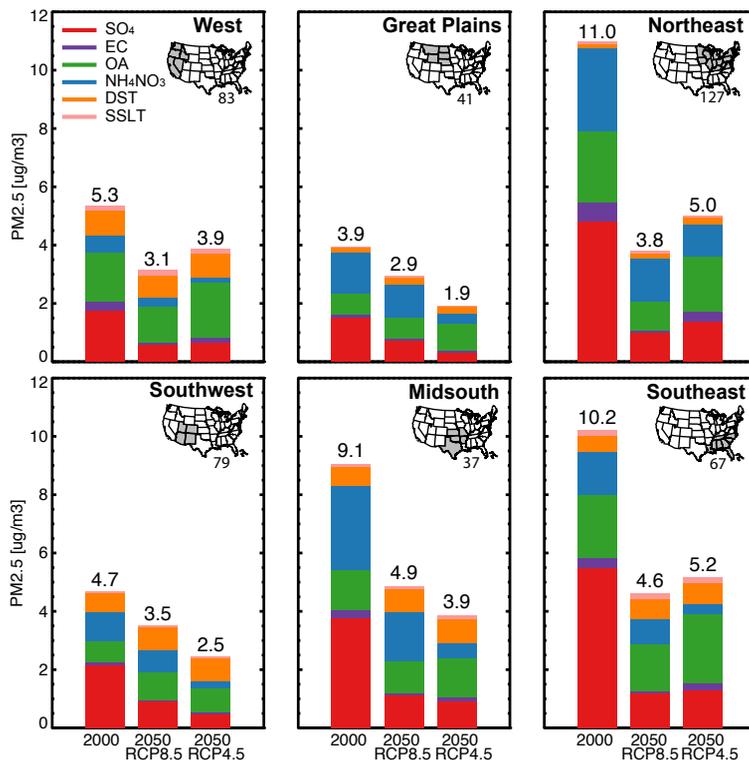
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**Figure 7.** Annual PM<sub>2.5</sub> chemical speciation for present-day and 2050 as predicted by the RCP4.5 and RCP8.5 scenarios in the US climatic regions. The inset maps show the states in the region in gray, and the numerals indicate the numbers of US National Parks and wilderness areas in each climatic region. Big numerals indicate the annual PM<sub>2.5</sub> concentrations.

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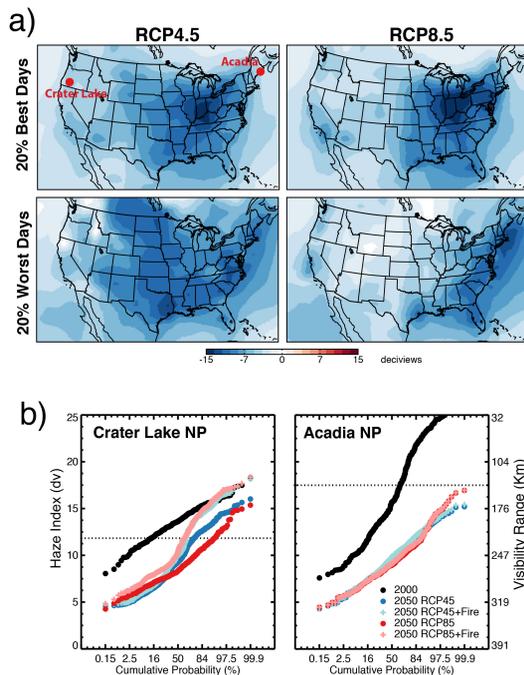
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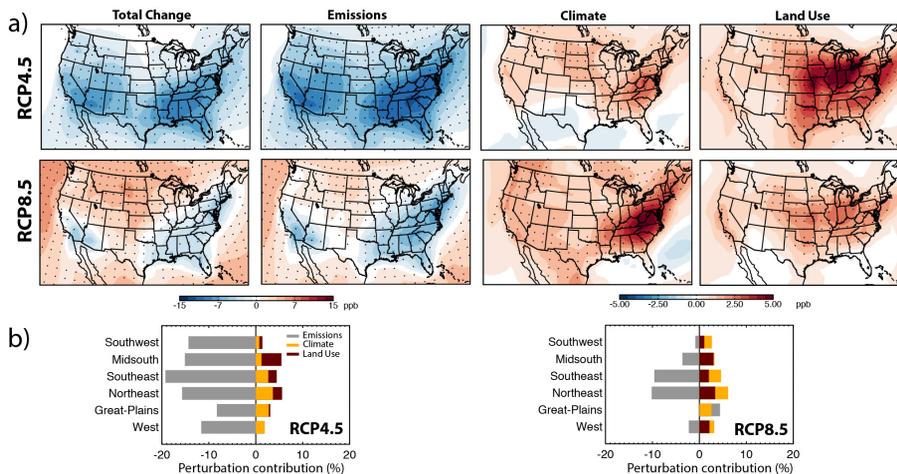
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**Figure 9.** Projected simulated 2050–2000 changes in haze index (HI) as a result of the combination of climate, land use and emissions changes (a) and the cumulative probability distributions of daily mean haze index in the Crater Lake and Acadia NPs (b). The maps 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). 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.

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**Figure 10.** Projected simulated 2050–2000 changes in surface O<sub>3</sub> 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. Regions with changes that are significant at the 95 % confidence level are indicated in the maps with dots, and O<sub>3</sub> concentrations are annual maximum daily 8 h (MDA-8) averages.

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