Response to Reviewer #1 for 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"

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 these 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.