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Interactive Comment

Interactive comment on "Present and future nitrogen deposition to national parks in the United States: critical load exceedances" *by* R. A. Ellis et al.

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Anonymous Referee #2 This is an interesting and well-written manuscript dealing with an important U.S. natural resource planning topic. It is appropriate for publication in ACP. I have only a few comments regarding the present period analysis. My most critical observation is a lack of caveats for GEOS-Chem and the future emission scenarios (see detailed comments below). 1. Pg 9154, lines 9-17: Emission from agricultural soils (driven in most cases by N-fertilizer) is also a major anthropogenic source of NO. Also please clarify that NOx from "fuel combustion" includes mobile as well as non-mobile sources. This becomes important in future scenarios with projected increases in urban areas and population. The largest fraction of NH3 (80% of agricultural emissions with ag emissions contributing 80%+ of total NH3 emissions) is emitted from animal husbandry activities so I would make this more prominent (not just another source). Soil-emitted NH3 may be quickly removed but, like NOx, a portion is transformed into particles, which can then undergo significant downstream transport. This is mentioned briefly later in the paper but should be mentioned here as well.

We have revised the paragraph, “Elevated N deposition from human activity is mainly driven by emissions of nitrogen oxides (NOx $\equiv$ NO + NO2) and ammonia (NH3) (Table 1). NOx is produced in combustion by oxidation of atmospheric N2 and fuel nitrogen. It is oxidized in the atmosphere on a time scale of a day to nitric acid (HNO3), which is removed rapidly by wet and dry deposition. The major source of NH3 is from livestock manure. NH3 is also produced industrially as fertilizer from reaction of N2 and H2 (Haber-Bosch process). Part of this NH3 is lost to the atmosphere upon fertilizer application. Soil cycling of fertilizer N is also a source of NOx. NH3 is removed rapidly from the atmosphere by wet and dry deposition, similarly to HNO3. Both HNO3 and NH3 can partition into particles, promoting long-range transport as particle dry deposition is slow."

2. Pg 9156, lines 7-8: As noted by reviewer 1, additional information is needed to explain the source of the present day emission differences for Zhang et al.

See response to reviewer 1. Differences are small.

3. Figure 2: Agreement between simulated and observed results is quite remarkable, particularly in the western U.S. where you are comparing relatively course model results to point measurements in complex terrain. Your results appear to be better than Zhang et al.. Some improvement is expected given your longer averaging period (annual vs monthly), but please discuss. Also, I assume the model background and observations shown are for 2006 only. If so, please correct the caption. If not, define what you are plotting for background values (e.g., average?). What observations are

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shown in the scatter plots, 2006, 2006-2008, avg 2006-2008? Which Parks lack NADP monitors? Would you care to speculate what the scatterplots would look like if these monitors were available (e.g., same, more scatter, less scatter, etc.).

Caption corrected (annual mean, 2006-2008). Our comparison statistics are comparable to Zhang et al. (2012) and we now say so in the text, “The model shows strong correlation with observations for both species and no significant national bias. Zhang et al. (2012) found similar agreement in their comparisons for the ensemble of NADP sites, with some degradation when considering seasonal variations.” Readers can figure out by comparing figures which 16 parks lack NADP monitors. Listing them in the text would be tedious, and we can’t speculate in absence of data.

4. Table 2: Please include “GEOS-Chem simulated deposition values for 2006” in the table caption. This is important information and should not be in a footnote.

We have changed the column heading to “GEOS-Chem deposition (2006)”.

5. Pg 9158, lines 25-26: “There is little 2006-2008 interannual variability, either in the model or in observations (Zhang et al., 2012).” On page 4547 of Zhang et al., they state that there is little interannual variability in the model results for 2006-2008, but I didn’t see any mention of interannual variability in the observations. One might expect interannual variability to be relatively low for regions dominated by dry deposition, but it can be significant in wet-dominated areas. Also, it is expected that a numerical simulation will underestimate observed variability. Unless I missed the statement regarding observations in Zhang et al., I think your statement in this manuscript is a bit too broad without additional support.

We have removed the line “in observations”.

6. You mentioned in passing that there are few or no dry deposition observations with which to evaluate your model. While correct, this is a major source of uncertainty. Since the western Parks are dominated by dry deposition, what are the implications of this

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uncertainty for your results? This could also be combined with reviewer 1's observation regarding the underestimation of current deposition totals in the west.

We have added to the final section, "Dry deposition usually accounts for most of total N deposition and this represents an important source of uncertainty, particularly in the west."

7. Insufficient caveats are provided for GEOS-Chem, but in particular caveats are missing for the RCPs. Caveats are needed to allow the reader to place the projected results in a meaningful application context. Below are a few caveat-related questions that come to mind: a) The future emission scenarios include land use change in response to socioeconomic and energy policy changes. Did you change the land use driving the future GEOS-Chem simulations to match the RCP changes? This may not be a serious issue in the eastern U.S., where total N deposition it is usually dominated by wet removal, but underlying vegetation is a critical driver for dry deposition processes, which dominate the West. If you did not make this adjustment, what are the implications for your results?

We added the following text, "The RCP scenarios also include land use change in response to socioeconomic and energy policy changes, which would affect dry deposition but we do not consider that effect here."

b) Figure 1 and text discussion. After reading the RCP documentation in van Vuuren et al. (2011a & b) and Riahi et al., 2011, the series of NOx emission maps seem reasonable. I am more curious about the NH3 maps – assuming that the majority of NH3 is associated with agricultural production. .... -The RCP scenarios project land use change using transition rates determined from historical data (1500-2000) (Hurtt, et al., 2011). These rates are then projected forward in time to 2100. Is the use of these historical transition rates, particularly in the western U.S. and the southern and western edges of the Midwest that have only recently been opened to extensive irrigation (post 1950), reasonable for projections to 2050?

We take the RCP scenarios at face value. They are widely used community scenarios. Assessing their reliability seems outside the scope of the paper.

-Land use transition rates are a function of agricultural productivity, proximity to existing agricultural areas, proximity to current water bodies and cities and a random factor. Your graphs of both RCP 8.5 and 2.6 suggest agricultural expansion/ intensification in Arizona, New Mexico, Wyoming and Montana in 2050. Is this reasonable considering water supplies are currently limited and may become more limited under future climate conditions?

See above.

-What is the source of the ammonia increase in RCP 8.5? Riahi states "aggregate arable land use in developed countries slightly decreases," and that bioenergy expands in the developing world and is focused primarily on forests biomass. He goes on to say that agricultural residues are used for bioenergy where cost effective, i.e., no dedicated bioenergy crops to drive agricultural production. Further, RCP 8.5 energy system moves toward coal-intensive technology and unconventional natural gas and oil extraction and urban population expansion in the west. The summaries in Hurtt and van Vuuren suggest that while biofuel expansion would explain NH3 increases in RPC 2.6, there is no such driver in 8.5. It is a little difficult to tease out U.S. trends from these broad global discussions (Riahi, et al. does not explicitly address NH3 at all). Perhaps a Table that summarizes regional changes in agricultural lands for the US would be helpful. Regardless, you need to help me to understand the source of the increased NH3 emissions in RCP 8.5.

We have added the following text, "The increase in the US is mainly due to increases in fertilizer use and livestock driven by a growing population and thus higher food demand (Riahi and van Vuuren, personal communication)."

c) Future NOx emissions: van Vuuren -Overview (pg 21) states that all the RCP scenarios assume NOx emissions will continue to decline in the future in response to "rising

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income levels." Particularly in RCP 8.5 – this is driven by policy intervention. Is the assumption of no technical limitation on NOx reductions reasonable? This assumption is particularly important for the western U.S., which is projected to have rather large increases in urban land use (Hurtt et al., 2011). What are the implications if these reductions are not realized?

Again we take the RCP scenarios at face value.

d) This paper addresses the response of N deposition to future emissions. What do you think would happen to your results if you included future weather as well, i.e., warmer temperatures and changing precipitation patterns?

We have added the following text, "We do not consider effects of climate change on transport and precipitation patterns; these effects are uncertain including in their sign (Jacob and Winner, 2009)."

e) Finally, bidirectional ammonia flux approaches are becoming widely accepted (Fletcher, et al., 2013, Massad et al., 2010; Bash et al., 2013; Dennis et al., 2013). Adoption of this approach would likely change both the ratio of wet to dry removal and N deposition in the western U.S. under current conditions. What are the potential implications for your study?

We have added the following text, "Better understanding is also needed of N cycling between the atmosphere and terrestrial ecosystems, as increased N deposition is expected to drive increased re-emission (not accounted here) that would extend the range of influence from a particular anthropogenic source."

f) Given these considerations, how do you see this work evolving in the future?

We have added a paragraph at the end of the section on policy implications: "There are several priority avenues for future work. Better understanding of NH3 dry deposition and its coupling to the terrestrial N cycle is critical. In our work we viewed deposition as a terminal sink for anthropogenic N, effectively assuming eventual removal from

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the receptor ecosystem by denitrification or runoff. However, some of the N will be re-emitted as NH3 or NOx. Improved model schemes have been recently developed to account for bidirectional fluxes of NH3 (Massad et al., 2010; Bash et al., 2013; Flechard et al., 2013) and to relate soil NOx emissions to N deposition (Hudman et al., 2013). A major development would be to use a fully coupled atmosphere-land model to track the fate of anthropogenic N as it cycles between the atmosphere and terrestrial ecosystems. Such a model could eventually lead to a better understanding of critical loads for ecosystems as related to atmospheric deposition. Better information is also needed on the factors expected to determine ammonia emission in the future including projections of agricultural activity and fertilizer use in different sectors.“

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9151, 2013.