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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Impact of Historical Land Use Change From 1850 to 2000 on Particulate Matter and Ozone" *by* Colette L. Heald and Jeffrey A. Geddes

Anonymous Referee #1

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This fascinating paper applies a systems-engineering modeling approach to assess the impact of 1850 versus 2000 land cover fraction changes on short-lived precursor emissions and the resultant atmospheric ozone and aerosol effects. The study examines the chemical sensitivity to the land cover change fractions under both 1850 and 2000 chemical background states (in terms of anthropogenic short-lived precursor emissions not the physical climate), in part to offer a useful uncertainty range estimate. The NH3/NOx/nitrate results are especially interesting and consistent with the recent Bauer et al., GRL 2015 results, also the Lelieveld et al., Nature, 2015 finding that agriculture is the main driver of particulate-related human health impacts in Europe. The paper is an important contribution to emerging land-chemistry-climate science and de-

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serves to be published in ACP once the following technical issues are addressed:

1. The main concern is that all simulations apply year 2010 meteorology. The underlying assumption is that surface albedo, energy and water changes caused by different land cover types have zero impact on atmospheric chemistry and aerosols. The manuscript needs to include clear statements about the missing meteorological feedbacks. Perhaps a more appropriate title would be something like: "..Impact of Historical Land Use Change Emissions. . .."

2. There is an extensive existing literature and multiple international assessment programs on the climatic effects of human land cover change through biophysical, albedo and meteorological changes. This paper essentially has pre-concluded that these water/energy/radiative changes have negligible impact on chemistry and aerosols, and only the short-lived precursor emissions changes are important. In fact, the meteorological/biophysical effects are apparently so unimportant to chemistry that they are not even mentioned. What is needed is a clear discussion of why albedo/biophysical/meteorological feedbacks have not been included in this analysis and how their inclusion would impact the results.

3. It is an excellent and efficient strategy to archive gridded radiative efficiencies that can be used in conjunction with global CTM-derived ozone and aerosol burden changes to assess radiative forcing impacts. However, the radiative efficiencies applied in this study are based on present day surface albedo and atmospheric water and cloud content etc. This seems to be wrong. The scattering aerosols (nitrate and BSOA) are quite sensitive to underlying surface albedo. For example, whether the underlying surface is covered in dark forest or bright crop/pasture can have a large quantitative impact on the local/regional aerosol radiative forcing. Atmospheric water content and cloud cover will also impact the aerosol radiative forcing results. There may be some effects on the SW ozone forcing too.

4. Assessing the impacts of land cover change on climate is a challenging multidisci-

plinary field. Therefore, it is critical for the chemistry-climate community to be extremely clear in their novel assessments that may be viewed by scientists from other disciplines (e.g. carbon cycle, surface biophysical climate communities) who may be less familiar with nuances in the atmospheric chemistry. For this reason, I recommend to modify the paper text carefully such that forcing values from Ward et al., 2014, Unger et al., 2014 and this study are not directly compared. These 3 exciting studies all examine rather different aspects of human land cover change effects on chemistry-climate interactions using completely different experimental design protocols. Indeed, the authors are quick to point out when their own results differ: "This value is smaller than the LULCC change in DRE (+0.034 Wm-2) estimated by Heald and Spracklen (2015). . ..is therefore not directly comparable". Yet, they proceed to compare Ward et al. 2014 and Unger et al. 2014: "We note here that the forcing estimated by Unger (2014) is of the opposite sign of that estimated by Ward et al. (2014)", which is fundamentally misleading to readers. Ward et al., 2014 includes methane, dust, fire and carbon changes, whereas Unger (2014) focuses on BVOC emission changes and physical climate feedbacks. Consider that global chemistry-climate models give a wide spread of ozone and aerosol results even when based on carefully designed harmonized experimental protocols (e.g. ACC-MIP)! Here, the authors are attempting to compare directly quantitative values from totally un-harmonized experiments that address different forcing components. Ultimately, a coordinated LU-AerChem-MIP multi-model assessment is needed if a single attributed LULCC-chemical forcing of global climate is to be determined, (if that exists).

5. Methane. The authors include an interesting discussion of the oxidation capacity consequences of omitting the expanding rice production. In addition, (i) animal husbandry/livestock is a large global source of methane emissions (ii) the land use-induced short-lived precursor emission changes (NOx, BVOCs etc.) would influence the methane lifetime itself too.

6. The authors conclude that the BSOA and ozone global forcing results are qual-

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itatively similar to those in Unger et al., 2014, but that the magnitude difference is mostly caused by differences in estimating the BVOC emissions change due to land use change. Does this mean that global BVOC-ozone and BSOA land use forcing depends mostly on BVOC emissions and is largely independent of the (complex) BVOC photochemical oxidation mechanisms under development? i.e. are current massive uncertainties in isoprene oxidation under different NOx regimes largely irrelevant for BVOC impacts on global climate? Based on current available information/evidence, the answer is yes.

7. In addition to the mapping of human land use onto the x-PFTs in global models, I suggest that the assigned PFT-specific basal emission rate for BVOCs is a large driver of the uncertainty too that needs to be discussed. For example, at least, if this study assigned a zero or v. low basal rate for pasture/grass PFT, would the results be even more consistent with Unger et al., 2014?

8. Does the SOA condensation model here depend on pre-existing OA levels? In which case the BSOA results would be sensitive to the assumed fire emissions that are prescribed to year 2010 in the study. Would burning be higher in some regions in 1850, leading to potentially even higher PI BSOA than reported here?

9. For IPCC-standard radiative forcing results, uncertainty ranges are needed. Naked values like -0.071 Wm-2, and -0.01 Wm-2 seem small and meaningless esp. without uncertainty ranges. Are these numbers statistically significant with 95% confidence relative to inter-annual climate variability in the model? Quantitative information is needed on the statistical robustness of the results. The authors argue in the abstract that these global forcing values are 'substantial'. That is a matter of debate e.g. the CO2 value is 1.8 Wm-2. Are 0.5-2% of the CO2 historical forcing values 'substantial'? Or are they simply lost in inter-annual climate variability?

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