

***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 #2

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Heald and Geddes “The Impact of Historical Land Use Change From 1850 to 2000 on Particulate Matter and Ozone”

GENERAL

This paper examined the impacts of historical land use change (LUC) and the associated agricultural emission change (AEC) on ozone and secondary particulate matter between preindustrial and present day. The main conclusion is that LUC+AEC result in increased burden of nitrate but decreased burden of BSOA and ozone. Such changes further induce radiative perturbations which present a strong cooling forcing since 1850. This is a fantastic work and analyses are comprehensive. Some minor revisions are required before the publication.

1. Some results presented in the study may be model dependent. The authors applied GEOS-Chem (GC) model in their study. Although the GC is a widely used and validated CTM, some inherent characteristics may definitely affect the changes in atmospheric chemistry. For example, to explain why the surface nitrate shows large deviations but tropospheric nitrate burden shows small differences between simulations using 1850 and 2000 anthropogenic emissions, the authors claim that “the increase in surface nitrate from pre-industrial to present-day is controlled more by the rise in anthropogenic NO_x emissions than the rise in agricultural ammonia emissions, while the increase in the burden of tropospheric nitrate is driven primarily by the increase in ammonia”. Are there any observations supporting such conclusion? Similar problems exist for ozone changes (shown in the detailed comments below). The authors need to discuss the possible uncertainties of these responses and remind readers that the predicted changes in atmospheric composition is somewhat model-dependent.

2. The authors performed sensitivity experiments to isolate the impacts of LUE and AEC (Table 2) but did not present those results in their analyses. Based on the qualitative explanations, we can understand that the large enhancement of nitrate is mainly attributed to AEC, the reductions in biogenic secondary organic aerosols (BSOA) is dominantly driven by LUC, and the decline of ozone burden is a compound result of AEC and LUC, and the impacts of LUC seem to outweigh that of AEC. However, without quantitative numbers, we do not know the individual contributions of LUC and AEC. I suggest that the authors add a new Table to summarize changes in atmospheric composition due to different drivers (LUC, AEC, and LUC+AEC) as indicated in Table 2.

3. Definition of LUC is confusing. Sometimes, LUC refers to LUC+AEC: “The global annual mean tropospheric burden of aerosol nitrate increases almost 4-fold due to historical LUC (Table 4)”. In the following sentence, however, LUC refers to land use change alone: “This increase is almost entirely the result of ammonia emissions increases; land use change alone (simulations 1 vs 2; see Tables 1 and 2) increases the

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tropospheric burden of nitrate by only 1.1%”. In addition, the phrase “land use change” is used frequently after the definition of abbreviation “LUC” in the paper. Similar problem exists for ‘DRF’ and ‘BSOA’. Some clean-up work is required for the clarity.

SPECIFIC

1. The title of the paper may be more appropriate as “The Impact of Historical Land Use Change From 1850 to 2000 on Ozone and Secondary Particulate Matter”
2. Page 9 Line 2: “where soil NO_x emissions increase due to land use change”, here NO_x emissions are due to AEC instead of LUC. Similar statement in the paper needs to be clarified.
3. Page 9 Lines 2-5: “Ozone production is widely NO_x limited under 1850 anthropogenic emissions, and thus the ozone production efficiency of additional soil NO_x emissions is considerably higher, and outweighs the impact of elevated deposition velocities for ozone due to LUC” This cannot explain why the burden of ozone is still decreased due to LUC with 1850 anthropogenic emissions.
4. Page 9 Line 19: “DRE” means “direct radiative effect” or just typo for “DRF”?
5. Figure 5 caption: Changes of soil NO_x and ammonia are caused by AEC instead of LUC.

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