Response to Reviews

We would like to thank the reviewers for their comments and suggestions. We have addressed these below (in blue below original comment).

REVIEWER #2

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.

We thank the reviewer for their positive comments.

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 NOx 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 reminder readers that the predicted changes in atmospheric composition is somewhat model-dependent.

The reviewer makes a good point that all modeling results are, to some extent, model dependent (hence the value of multi-model assessments). To our knowledge, none of our results are exceptionally dependent on the use of the GEOS-Chem model, however we allow that such dependencies (on the specific chemistry scheme, on the GMAO meteorology, etc.) may exist. We add a sentence to acknowledge this. Unfortunately we do not have observational constraints over the pre-industrial to present-day to verify our results, and this must therefore be considered purely a modeling study based on our current knowledge of biosphere-atmosphere exchange and atmospheric chemistry.

Modification:

k. Page 11, lines 19-23: text added: "The simulations analysed in this study were performed with one chemical transport model (GEOS-Chem); the degree to which model-specific treatments of chemical oxidation, aerosol formation, and meteorology may impact the results cannot be assessed here. Thus, additional modelling investigations using alternate model schemes are required to better characterize the uncertainty surrounding the impact of land use change on air quality and climate forcing."

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 overweigh 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.
We have expanded Table 3 to include the quantitative differences in the simulations as requested.

Modification:

l. Table 3 now separately specifies emissions changes due to LUC, and LUC+AEC

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 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.

We appreciate the reviewer's suggestion. We defined LUC to incorporate the net results of land use change and the associated agricultural emissions changes (and present only those results in the paper). We have clarified this in the text.

Modifications:

m. Page 5, lines 15-16: added text: "We focus our results on the net impacts of land use change along with the associated changes in agricultural emissions (which we collectively refer to as LUC), unless otherwise specified."
n. We have replaced most usages of "direct radiative forcing" with DRF and most usages of "land use change" with LUC in the text.

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"

We agree, and have made this change.

2. Page 9 Line 2: "where soil NOx emissions increase due to land use change", here NOx emissions are due to AEC instead of LUC. Similar statement in the paper needs to be clarified.

In fact the changes to soil NOx are due to both LUC and agricultural emissions (as shown in Table 3). As we have defined LUC to include both land use change and the associated agricultural emissions, this sentence remains unchanged.

3. Page 9 Lines 2-5: "Ozone production is widely NOx limited under 1850 anthropogenic emissions, and thus the ozone production efficiency of additional soil NOx 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.

We believe the reviewer may have misinterpreted the sentence. The purpose of this sentence is to explain the contrast in surface concentrations when using 1850 anthropogenic emissions (vs. 2000 anthropogenic emissions) NOT the difference in the burden. The changes in surface concentrations are modest and localized and translate in both cases to a very small decrease in burden. This is consistent with Figures 7, 8, 9, and 10.

4. Page 9 Line 19: “DRE” means “direct radiative effect” or just typo for “DRF”?

Thank you for catching this. We have added text to define DRE as “direct radiative effect”

5. Figure 5 caption: Changes of soil NOx and ammonia are caused by AEC instead of LUC.

In fact the changes to soil NOx are due to both LUC and agricultural emissions. We have clarified the caption.