Dear Handling Editor,

Many thanks for your careful review of our manuscript. We appreciate your time, and value your concerns. We believe we have addressed all your concerns in our revised manuscript. Below, we respond to each of your comments individually, followed by the appropriate changes to our manuscript.

We look forward to your timely response to this revision, and hope that you will agree our manuscript has improved in response to your input.

First of all, sorry for the delay in my decision on your manuscript having received already some of the last reviewers' comments quite a long time ago. Anyhow, I didn't want to take a hasty decision and needed to the find the time to carefully check once more again the reviews and your response to the shared feedback. This was a good decision since in doing so, also reading over again the revised ms I came across a number of more minor issues, that should be anyhow resolved. But there are also still some more major issues that anyhow must resolved and some that you could potentially consider in a further revision. This mainly refers to some specific features of the representation of atmosphere-biosphere exchange in the GEOS-CHEM modelling system you applied for your analysis and which have not been raised by the reviewers.

Comments

Lines 39:40; remove the double point at begin of sentence there and also refer consistently to NOx (subscript x).

Line 53: "in land cover classification.". By putting the term classification you make it appear more that the classification of land cover is the main issue where it is simply "no changes in land cover"

Line 305: "have declined"

Lines 323-325: "the impacts of agricultural emission changes on O_3 ("$\Delta O_{3,\ agr\_emis}$") is the difference in $PM_{2.5}$ O_3 predicted by Simulation 3 and Simulation 2; and the impacts of these combined ("$\Delta O_{3,\ LULCC+agr\_emis}$") is the difference in $PM_{2.5}$ O_3 predicted Simulation 3 and Simulation 1"

Line 341: "NOx -> NOx" and check this for consistency.

Line 367: "but these there the positive and negative largely offset each other"

Line 395: "which partially offsets"

Line 397: "does not lead to substantial changes"

Line 423: "Our results suggest that contemporary (1996-2014) changes <u>in LULCC and agricultural emissions</u> contribute to changes.

Response: We have made all revisions above as suggested.

Lines 70:74: I do really appreciate your point about the fact that we need to consistently consider the combined effect of both LULCC and changes in agricultural emissions also since here we might see some compensating effects. This was actually one of the main take-home messages also of the Ganzeveld et al. 2010 study in which we included in the most consistent manner these combined effects but then in a study on the anticipated future changes in LULCC and agricultural N-emissions (only NOx). I am very much *aware that by sharing this comment at this stage that 1) I should have done this in an earlier stage and 2) that I am really in doubt making this comment since I don't want to leave any impression of "pushing" my own papers. But now having read again this particular statement making a strong point about this consistent representation of all involved processes, I bring it up since I also see that this is actually a shortcoming of many other studies. This is also further stressed in the follow-up statements in lines 76-79.*

Response: We agree with the editor that the conclusion of Ganzeveld et al. 2010 study is highly relevant. We have included a more detailed discussion in our manuscript:

L 70 – 84: While changes in land cover and agricultural emissions actually occur simultaneously across the globe, they are rarely considered together in simulations of air quality from chemical transport models. The importance of studying these combined processes at the same time was highlighted by Ganzeveld et al. (2010) in their analysis of air quality impacts from future land use and land cover changes. In this study, for example, opposing effects on O_3 were simulated with decreases in tropical forest soil NO_x emissions being compensated by increases in soil NO_x emissions associated with agriculture. Still, this work did not explore the concomitant changes in ammonia emissions that would be expected with the changes in agricultural activity. It remains unclear to what extent LULCC… This opens an opportunity for a more holistic and observationally-constrained assessment of the impacts on global O_3 and PM air quality from contemporary changes in LULCC and agricultural emissions simultaneously, which has been advocated by Ganzeveld et al. (2010), and a comparison…

Chapter 2: Methods; The description of all the steps to consider the dependence of dry deposition and emissions on LULCC and agriculture makes clear that you made a large effort to consistently consider the impact of this on these two processes. However, it triggers the question to what extent your results might then be missing one specific aspect of atmosphere-biosphere exchange that might be quite important for the overall/compensating effects; canopy interactions; e.g., how much of the emitted NOx and NH3 is really escaping the vegetation canopy (especially relevant for large LAI's) and how a decoupled treatment of soil-canopy N-emission and deposition would further effect your results. The first feature, also referred to as the canopy reduction factor is considered in the Hudman et al. soil NOx emission inventory but how did you handle this for NH3? And how for the fertilizer-application driven NOx emissions in your approach (reading that those were removed from the Hudman inventory to avoid double counting). In addition, there would be some other aspects of atmosphere-biosphere exchange of relevance for your study, and that should be included in the discussion: how does the deposition representation in GEOS-CHEM consider the dependence on stomatal exchange and soil water status (an important feature of LULCC). You refer in the discussions shortly to the fact that e.g., the coupling with latent heat exchange and boundary layer dynamics has been ignored. I am very much aware that some of these features (and uncertainties) in LULCC and agricultural management are likely much more important but not having considered these additional dependencies of the system in a consistent manner is important to indicate already at an earlier point in your ms. In addition, there are other aspects of (N) atmosphere-biosphere exchange that have not been mentioned at all and might be quite relevant, existence of NOx and NH3 compensation points. Properly discussing these potentially important features is required also reading lines 211-213: "represent the change in soil emission driven purely by LAI and land cover changes" The same holds for the statement in line 223-224: "Significant changes in the vd of O_3 due to LAI also imply that v_d of other relevant trace gases (e.g. NO2, SO2)"; how is the deposition of NO_2 being treated in GEOS-CHEM, e.g., does it consider a significant N- compensation point for ecosystems prone to high N loading?

Response: We thank the editor for highlighting the importance of considering other important processes in comprehensively simulating atmosphere-biosphere exchange.

The question of bidirectional exchange and compensation points is a very interesting one to consider, which we should have addressed more explicitly in our manuscript. We note that bidirectional exchange has in some cases been implicitly accounted for in the CEDS inventory for ammonia fertilizer emissions. For example, in CEDS, the agricultural ammonia emissions over the United States are scaled to the NEI emissions estimate, and would therefore reflect some of the assumptions included in the NEI emissions modeling for ammonia (which is based on an implementation of bidirectional exchange in the CMAQ model). However, we cannot necessarily comment with certainty on how this would be treated elsewhere, and this will introduce an element of uncertainty in our simulation. It is also the case that the current public release of GEOS-Chem otherwise does not have online bidirectional exchange parameterizations for any species.

In response to this comment from the editor, we discuss the importance of bidirectional exchange for NH_3, and evidence for atmospheric compensation points for NO_2, in our revised manuscript. We clarify that in some cases these effects may have been accounted for implicitly, but include the caveat that our work cannot account for all these effects everywhere, since it is not yet the state of the science for GEOS-Chem. We discuss how neglecting these effects might contribute to uncertainties in our simulation, by drawing on global studies that have implemented bidirectional exchange for ammonia, and elaborate on the importance of this in future model investigation.

Regarding canopy uptake, we clarify in our revised manuscript that indeed the biogenic soil NOx emissions calculated using the Hudman scheme includes an explicit online treatment of canopy uptake. The implicit accounting of canopy uptake in the agricultural emissions may again depend somewhat on whether CEDS has scaled the emissions to a particular regional inventory that accounts for this (as would be the case over the United States). For simplicity, therefore, we make the assumption that the CEDS inventory provides an estimate of "above canopy" emissions into the atmosphere. We further argue that these agricultural emissions from soil represent only a fraction of the total agricultural emissions considered here (e.g. in addition to manure- and waste-associated emissions), so that the uncertainty introduced by the canopy reduction factor is only applicable for a fraction (~35-50% depending on region) of the agricultural ammonia emissions, reducing somewhat the apparent importance of quantifying this. Likewise, the fertilizer soil NOx emissions represent a small overall fraction of the total global soil NOx emissions (which in our simulation otherwise include a canopy reduction estimate). In response to the editor's comment we clarify these points in our revised manuscript, and include a caveat that this deserves detailed attention in future implementations of globally consistent emissions inventory development.

Overall, we believe the editor has raised excellent points of concern which deserved to be addressed in our manuscript. We feel that the changes we have made draw attention to these concerns, and point to future work that could be done to improve the representation of these complex biosphere-atmosphere exchange processes in GEOS-Chem.

L 115 – 124: Gaseous dry deposition follows Wang et al. (1998) and Wesely (1989), while particle deposition follows Zhang et al. (2001). In GEOS-Chem, the surface exchange modules are uni-directional (which implies that the effects of bidirectional exchanges of trace gases are not explicitly

modelled). In certain regions for which the CEDS inventory scales the calculated emissions to a regional inventory, the extent of accounting for bidirectional exchange may depend on the underlying assumptions in the regional inventory modeling. For example, agricultural ammonia emissions from NEI for the United States includes considering bidirectional ammonia exchange modeling from the Community Multiscale Air Quality Modeling System (CMAQ) (US EPA, 2018) **. However, we cannot comment with certainty how this is treated elsewhere across the globe, so we assume that neglecting bidirectional exchange of ammonia (and other species for which an atmospheric compensation point may exist) introduces some uncertainty in our simulation (which we discuss in a subsequent section).**

L 136 – 144: For this study, "agricultural emissions" specifically refer to NO_x and NH_3 emitted from fertilizer application and manure management, which correspond directly to agricultural nitrogen input. We do not consider the changes in agricultural of other trace species (e.g. CH_4, SO_2, CO). For simplicity, we assume that agricultural emissions from fertilizer application in CEDS represent "above canopy" emissions to the atmosphere (instead of making assumptions about the implicit treatment of canopy reduction over each region). We note that the fertilizer emissions of represent only a fraction of the total agricultural NH_3 emissions we are considering here (e.g. which also include livestock operation), so that uncertainty in a canopy reduction will only affect a fraction of the total. Likewise, fertilizer NO_x emissions are small compared to the total soil NO_x emissions (for which canopy reduction is accounted for online in the Hudman et al. (2012) parameterization).

L 228 – 230: Figure 3b shows the changes in annual mean soil NO emission due to LULCC, which represent the change in soil emission driven purely by LAI (which can also affect canopy uptake) and land cover changes (which affects both biome-based emission factor and canopy uptake) (i.e. without considering the changes in nitrogen input)…

L 491 – 506: Agricultural NO_x and NH_3 emissions estimates also carry large uncertainty due their biological nature and resulting dependence on environmental conditions, which are not explicitly considered in the construction of bottom-up anthropogenic emission inventories (Crippa et al., 2018; Hoesly et al., 2018). Bidirectional exchanges of NO_2 (Breuninger et al., 2013; Chaparro-Suarez et al., 2011; Lerdau et al., 2000) **and NH_3** (Bash et al., 2013; Massad et al., 2010; Wichink Kruit et al., 2012; Zhang et al., 2010)**are not explicitly modelled (although in some regions may be implicitly accounted for in the regional scaling performed by CEDS), which introduces some uncertainty in the accuracy of surface flux modelling. Zhu et al. (2015) implemented a bi-directional NH_3 exchange model in GEOS-Chem, and found no substantial improvement with observations in the modelled NH_3 concentration, NH_4^+ wet deposition and nitrate aerosol concentration compared to the default GEOS-Chem uni-directional exchange framework. This indicates the uni-directional framework may still be sufficiently accurate in simulating global air quality comparing to bi-directional framework, which requires more observations to properly parameterize at global scale. In the case of NO_2, we make the assumption that in most regions we are interested in (fig. S9), the ambient concentrations of NO_2 exceed an ecosystem compensation point (0.05-0.6 ppb) (e.g. Breuninger et al. 2013) so that we can assume deposition would dominate. The simplistic representation of dry deposition in general, particularly the lack of dependence of stomatal conductance on atmospheric and soil water content, may not adequately capture the effects of LULCC, as biomes can have differential responses to meteorological and hydrological conditions. The inherent…**

Lines 333-334: Here there is an apparent flaw: "In contrast, modelled surface ozone increases by up to 1.2 ppbv further south, where strong increases in LAI lead to largely increases vd"; O3 increasing due to enhanced dry deposition? It should also read as "lead to large increases in vd" and what is large? Give a percentage or the absolute numbers.

Response: We thank the editor for pointing out our mistake. We have made the following correction:

L 353 – 354: In contrast, modelled surface ozone ~~increases~~ decreases by up to 1.2 ppbv further south, where strong increases in LAI lead to largely increases v_d (up to 0.06 m s^{-1}).

Line 336: "up to 0.6 ppbv of surface ozone increases are simulated, mainly because of the relatively large increase in soil NO emission". This is an example that triggers the question what happened to the effective emissions into the atmosphere; is it indeed purely the changes in the soil NO emissions (due to temperature or moisture effects, or management) and how much an effect is there by changes in the canopy reduction factor due to changes in LAI?

Response: We thank the editor for raising this interesting question. The relatively strong increase in soil NO emission over West Africa is likely due to the combination of reduced LAI (and therefore lower canopy reduction factor) and cropland expansion. We have made the following changes:

L 232 – 234: Relatively large increases in soil NO is simulated over western Africa due to both cropland expansion and LAI reduction, which leads to smaller canopy reduction factor and larger emission factor.

Lines 428-230; these conclusions are consistent with the findings by Ganzeveld et al. on the small impact of future LULCC and agricultural emissions changes on ozone also due to a number of compensating effects. I think it would be very useful to stress that your findings on contemporary versus future changes in LULCC and agricultural emissions in different modelling systems/approaches come up with such a consistent finding.

Response: We agree with the editor, that the similarity of conclusion under different timeframe and modelling framework may start to indicate certain generality, which is important to note. We have made the following changes:

L 447 – 450: We find that the role of LULCC over 1992 to 2014 is regionally significant enough to induce changes in BVOC emissions and dry deposition which affect surface O_3, but that the overall effects largely offset each other on the global scale, leading to very small population-weighted $\Delta O_{3,\,LULCC+agr\_emis}$. This finding with consistent with that of Ganzeveld et al. (2010), even the timeframe of study (2000 – 2050) is different.