

Review of paper acp-2021-132: On the competing effects of contemporary land management vs. land cover changes on global air quality by Wong and Geddes

Dear author, co-authors,

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 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 be 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.

Lines 39:40; remove the double point at begin of sentence there and also refer consistently to NO_x (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"

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 NO_x). 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.

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 NO_x 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 NO_x emission inventory but how did you handle this for NH₃? And how for the fertilizer-application driven NO_x 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 NO_x 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 O₃ due to LAI also imply that vd of other relevant trace gases (e.g. NO₂, SO₂)”; how is the deposition of NO₂ being treated in GEOS-CHEM, e.g., does it consider a significant N-compensation point for ecosystems prone to high N loading?

Line 305: “have declined”

Lines 323-325: “the impacts of agricultural emission changes on O₃ (“ Δ O₃, agr_emis”) is the difference in ~~PM_{2.5}~~ O₃ predicted by Simulation 3 and Simulation 2; and the impacts of these combined (“ Δ O₃, LULCC+agr_emis”) is the difference in ~~PM_{2.5}~~ O₃ predicted Simulation 3 and Simulation 1”

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.

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?

Line 341: “NO_x -> NO_x” 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 in LULCC and agricultural emissions contribute to changes.

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.