Apologies to the late reply. These are the comments to the revised version
https://doi.org/10.5194/acp-2021-132-AC2
The scope of this manuscript is broad, as it is about the relationship between land use change and air pollutants emission, it is relevant in a global scale. The research question is important for interdisciplinary interest and has real world implication. The research has a high potential for societal and policy impact.
Wong and Geddes use a factorial experiments approach to investigate the relative impact of Agricultural emissions, LULCC and Anthropogenic emissions. Wong and Geddes found that agricultural emissions have strong effects on PM2.5, while LULCC has strong effects on O3.
Thanks a lot for consolidating the comments into a revised manuscript. It is much better now especially after addressing the comments from Reviewer 2.
Here are some specific comments for your consideration.

Comments

1. *I feel that the title of the paper does not sound right. Perhaps it could be "Study on the competing effects …" Instead of starting with "On" .*

   Response: We thank the review for this suggestion. We change our title to:

   Examining the competing effects…

2. *There are many complex sentences that I think it is too long. Try split those long sentences into several short one, it would improve the readability.*

   Response: We thank the review for this suggestion. In response to this reviewer's comment, we have made the following changes to the manuscript:

   L 76 – 77: ~~The recent availability both of~~ Consistent long-term ~~land~~ records of land cover derived from satellite remote sensing observations and global anthropogenic emission inventories have become readily available. This~~,~~ opens an opportunity…
   L 173 – 174: …have increased mainly at the expense of forest coverage. ~~, indicative of~~ This is consistent with a global trend in deforestation over this period…
   L 203 – 204: … The largest local reductions in isoprene emissions (up to 30%) are observed in parts of South America, where deforestation from highly isoprene-emitting broadleaf forests is most strongly observed. ~~, causing decreases in isoprene emissions by up to 30%…~~
   L 406 – 410: …We model the effects of contemporary LULCC and agricultural emission changes, individually then in combination, on surface O_3 and $PM_{2.5}$ using the GEOS-Chem CTM. With a uniquely consistent framework, ~~that is~~ we are able to integrate direct information from global emission inventories (CEDS) with updated land surface remote sensing products (ESA CCI land cover and GLASS LAI). ~~on surface O_3 and $PM_{2.5}$, allowing~~ This allows us to avoid invoking extra

assumptions on land management practices (e.g. constant Nr input, emissions or emission factors over time) and biophysical properties of PFTs (e.g. constant PFT-specific LAI over time).

3. *The Introduction is a bit short; it would be helpful if you could include more detail about how agricultural emissions lead to the formation of secondary PM2.5. Under what conditions that would affect its formation. Same for O3 as well.*

   Response: We thank the review for this suggestion. We agree that this will increase the readability of our manuscript. In response to this reviewer's comment, we have made the following changes to the manuscript:

   L 36 – 42: The reactive nitrogen oxides emitted from soil, NOx (≡ NO + NO_2), ~~is a key component of O_3 photochemistry~~ enhance O_3 production when volatile organic compounds (VOCs) are relatively abundant (i.e. NOx-limited regimes), but suppresses O_3 production when the concentration of VOCs is relatively low (i.e. VOC-limited regimes) (Sillman et al., 1990). Reactive nitrogen also contributes to aerosol formation. Ammonia (NH_3) can combine with the nitrate and sulphate ions to form secondary inorganic aerosol, while the emissions of NOx can oxidize further and contribute to particulate nitrate formation (Ansari and Pandis, 1998). Indeed, agricultural emissions are the dominant global anthropogenic source of ~~ammonia (~~NH_3~~)~~ (Hoesly et al., 2018)…

4. *L 67: Any better word usage other than "contemporaneously"? How about simultaneously?*
5. *L 89: "an" instead of "a"*
6. *L 93: Why use "fifth" here instead of "(5)", be consistent.*
7. *L 145: "in"*

   Response: We have made all revisions above as suggested.

8. *L 179: area with regionally consistent deforestation experience increase in LAI? I thought it would decrease LAI when turning forest into grass, please explain.*

   L 183: LAI increases in northern China is not because of decrease of agricultural land but could be afforestation and turning desert to farmland? You can search for the keywords: Green wall of China. And this Nature paper *https://www.nature.com/articles/s41893-019-0220-7*

   L 235: Could be explain by afforestation. See point 9.

   Response: We thank the reviewer for the three related comments above. We appreciate the suggestion of an excellent reference about LAI changes in general, particularly about India and China. After careful studying, we find the paper suggesting in addition to reforestation, greening (increase of LAI) within cropland and forests also contributes to overall greening.

In response to this reviewer's comments, we have made the following changes to the manuscript:

L 188 – 192: …For example, the general increase of LAI in China is not only driven by changes in biome types, but also the greening within cropland (mainly attributable to agricultural intensification) and forests (mainly attributable to ambitious tree planting programmes) (Chen et al., 2019). Similarly, some deforested land in South America might have been cultivated intensively, resulting in an increase rather decrease in LAI. We also note that since the…

L 245 – 246: …over this same period. Such agricultural intensification in turn contribute significantly to the positive LAI trend over the above regions (Chen et al., 2019). Similarly, agricultural emissions…

9. *L 227: what causes the sharp decline of agricultural emissions in Europe? Reduce farming activity? Implementation of clean air policies?*

Response: We thank the reviewer for their questions. We agree that the decline of NH_3 emissions in Europe is indeed very significant, meanwhile well-documented and analyzed, which warrant deeper discussion.

In response to this reviewer's comment, we have made the following changes to the manuscript:

L 235 – 237: The particularly sharp decline of agricultural emissions in Europe is mainly attributable to the implementation of emission control protocols (National Emissions Ceilings (NEC) and Integrated Pollution Prevention and Control (IPPC) directives) within the European Union (Skjøth and Hertel, 2013). According to the CEDS inventory…

10. *L 272: I still do not understand clearly how you calculate the population-weighted average. Do you mean per capita? Where do you get the population data from?*

Response: We thank the reviewer for pointing out the ambiguity. In response to this reviewer's comment, we have added description of our method of calculating population-weighted average in supplemental material:

Text S2. The population-weighted averaged changes surface O_3 (ppb) or $PM_{2.5}$ (μg m^{-3}) ($\Delta[X]_{\text{pop\_weighted, Y}}$) for region Y is calculated as follow:

$$\Delta[X]_{\text{pop\_weighted,Y}} = \frac{\sum_i^{gridcells\ in\ Y} \Delta[X]_i Pop_i}{\sum_i^{gridcells\ in\ Y} Pop_i} \text{(S2)}$$

where $\Delta[X]_i$ is changes in surface concentration of concerned chemical species, and Pop_i is the population count for individual gridcell i. The global gridded population is from the fourth version of The Gridded Population of the World (GPWv4) (CIESIN, 2018), and remapped to match the resolution of GEOS-Chem output.

We also make this change in our main text to reference the supplemental text:

L 282: …so that the effects on population-weighted average (method described in Text S2)…

11. *L 281: "land change phenomena", do you mean land use change? Or the area change as what Table 3 suggests? Please use a word that would not confuse the readers.*

    Response: We thank the reviewer for pointing out the ambiguity. In response to this reviewer's comment, we have clarified our wording:

    L 291: Table 3 summarizes the simulated effects of ~~these land change phenomena~~ LULCC and agricultural emission changes on $PM_{2.5}$…

12. *Conclusions: It is good that you mentioned about the limitation in the study. You could also add several sentences to discuss about the implication on policies. What policies could help reduce or mitigate the impact of LULCC, agricultural and anthropogenic emissions? How do you prioritise it? It will increase the impact of your paper to policymakers.*

    Response: We thank the reviewer for this constructive comment. We agree that some discussion about potential policy priorities to mitigate air pollution from land system is a valuable addition without sacrificing the scientific rigor of our manuscript.

    In response to this reviewer's comment, we have made the following changes to the manuscript:

    L 488 – 496: Incentivizing these and other practices that improve agricultural nitrogen use efficiency (e.g. including livestock production with cropping, synchronizing nitrogen supply with crop demand) (e.g. Fageria and Baligar, 2005; Langholtz et al., 2021) can be one of the keys to mitigate the air quality impacts of reactive nitrogen input without compromising agricultural productivity (e.g. Guo et al., 2020). Furthermore, as increasing reactive nitrogen input and land use change are the two of the main strategies to meet the global demand for biomass-based products in the future (Foley et al., 2011), the distinct yet significant impacts of agricultural emissions and land use change on O3, PM2.5 and nitrogen deposition should be investigated as part of the overall environmental impacts of land system changes, especially when tradeoff between increasing land input and cropland expansion exists (e.g. Lotze-Campen et al., 2010; Mauser et al., 2015). This could benefit agricultural policy activities by appropriately considering all in the externalities and socioeconomic costs of different options and scenarios for agricultural expansion.

13. *The caption of the figure should allow readers to understand the figure without looking at the main text, self-explanatory. If possible, write the complete form of acronyms that are not used frequently in the main text, for example CEDS in Figure 4. If space allows, I will write "Leaf Area Index" instead of LAI in Figure 2. Same for Figure 5b and 6b, you could write "Agricultural Emissions effect" instead.*

We thank the reviewer for this suggestion. In response to this reviewer's comment, we have made the following changes to our figure captions:

L 824 – 825: Figure 2. Global Land Surface Satellite (GLASS) ~~GLASS~~ derived changes in 3-year mean annual ~~LAI~~ leaf area index (LAI) (2012 to 2014 average minus 1991 to 1993 average).

L 830 – 831: Figure 4. Changes in agricultural NH_3 and NO_x emissions (2014 – 1992) as implemented by the Community Emissions Data System (CEDS).

L 832 – 833: Figure 5. Simulated changes in annual mean surface $PM_{2.5}$ due to (a) LULCC, (b) agricultural emission ("Agr Emis") changes, and (c) the combined effects of agricultural emissions and LULCC.

L 834 – 835: Figure 6. Simulated changes in annual mean surface O_3 due to (a) LULCC, (b) agricultural emission ("Agr Emis") changes, and (c) the combined effects of agricultural emissions and LULCC.