Wong and Geddes present work comparing the relative influence of land use/land cover change (LULCC) and agricultural reactive nitrogen emissions on air quality over modern timescales. They carry out this work using the GEOS-Chem chemical transport model along with a variety of updated emissions inventories and satellite products. In the end, the find that both effects can be important for regional air quality and trends in both LULLC and reactive nitrogen emissions should be considered when assessing multi-decadal trends in air quality. The manuscript is generally well structured and describes a thorough investigation. I can likely recommend this paper for publication after the following minor points are addressed.

Major Comments

1. *The large number of specific geographical regions referenced in the manuscript substantially reduces the readability of the work. For example, how do the changes over Myanmar track with the changes across Southeast Asia (Germany and Benelux, Southern Russia and Kazakhstan, Southern Amazonia and Paraguay, etc.)? I suggest the authors standardize locations more consistently if possible.*

   Response: We thank the reviewer for making this suggestion to improve readability. When we describe the geographic patterns of LULCC, there are a lot of detailed sub-regional patterns that we thought might warrant certain specific geographic designation. We then introduced standardized definitions of regions in Table S2 for describing the area- and population-weighted average changes in O_3 and $PM_{2.5}$. We understand that in many occasions this choice could have reduced the readability of the manuscript where it is unnecessary to include specific geographic descriptions. In our revised manuscript, we increase the use of the standardized definitions of regions in Table S2, and include more specific regional descriptions only where necessary. Furthermore, we remove the use of unnecessary regional abbreviations (e.g. "SEA", "Cam", "WAf") and replace them with the full names of each region, which we feel also improves the readability of the manuscript.

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

   L 162: …providing basis for our subsequent analyses. We also provide definitions for geographical regions, which largely follow Integrated modelling of global environmental change (IMAGE) 2.4 classifications, in Table S2.

   L 168 – 174: …cover types. Expansion of agricultural land at the expense of broadleaf forest coverage is most notable in ~~southern Amazonia~~ South America and Southeast Asia, which is well-documented…The expansion of agricultural land over this time period is also observed in central Asia, ~~African savannah, southeastern~~ Australia, ~~South~~ southern China and ~~North~~ Africa…built-up areas is observed in northern China and ~~eastern~~ Europe, consistent with the findings of Potapov et al. (2015) and Lai et al. (2016).

L 176 – 181: Figure 2 shows the global changes in 3-year (2012-2014 minus 1991-1993) annual mean LAI calculated from the GLASS LAI data set. Over ~~South~~ southern China and South America~~, Paraguay and northern Argentina~~, the area with regionally consistent deforestation experience general increase in LAI, while the opposite effect is observed in Sahel and Former Soviet Union ~~African savannah and central Asia~~. In Europe, LAI ~~decreases in Ukraine, Poland, Germany, but~~ increases in most ~~other~~ parts despite a fairly consistent retraction of agricultural land is observed over the whole Europe…

L 190 – 194: …local reductions in isoprene emissions are observed in parts of South America ~~southern Amazonia, Paraguay and northern Argentina~~…We note that the decrease of isoprene emission simulated in Southeast Asia does not agree with the result from Silva et al. (2016)…

L 202 – 204: …The magnitude of changes in soil NO emission induced by LULCC is comparable to that in agricultural NO emissions inventory (see below) over certain regions (e.g. South America, Australia, ~~Sub-Saharan~~ Africa). Relatively large increases in soil NO is simulated over western Africa …

L 207 – 212: Slight increases of v_d are observed in China, India, Southeast US, ~~Mexico most of~~ central America, ~~northern Amazonia~~South America, Europe and southern Africa. In Southeast Asia v_d decreases concurrently with deforestation and reduction in LAI…Likewise, despite deforestation observed further south ~~in the savannah and grassland over Rio de la Plata basin~~, these losses are offset by strong increases in LAI so that v_d increases by up to 0.1 cm s^{-1}.

L 221 – 224: The increases in agricultural emissions are most substantial over ~~the Indian Subcontinent~~ South Asia, followed by China, parts of ~~Egypt~~ Middle East, Southeast Asia and ~~Brazil~~ South America, and to a less degree in ~~other Latin American countries~~ central America, North America~~, West Asia~~ and ~~African savannah~~ Sahel. The sharpest decline of agricultural emissions is observed in ~~eastern~~ Europe ~~and Benelux~~ and Former Soviet Union, followed by milder declines ~~across central Europe and~~ over Japan and Korea...

L 244 – 248: Over parts of South America and Southeast Asia ~~southern Amazonia and maritime southeast Asia~~, where isoprene emissions drop significantly due to deforestation… Indeed, over ~~northeastern~~ India and ~~eastern~~ China, where our model suggests high levels of SNA aerosol precursors…

L 255 – 257: … $PM_{2.5}$ concentrations are also observed in the Middle East, North America, central America and South America. ~~West Asia, Egypt, eastern US, Mexico, Peru and southeastern Brazil.~~

L 258 – 264: The largest decreases (up to 2.1 µg m^{-3}) in annual mean $PM_{2.5}$ due to changes in agricultural emissions are simulated in central and eastern Europe and Former Soviet Union. ~~eastern Europe, particularly over Ukraine (-2.1 µg m-3),~~

~~Bulgaria (-1.5 µg m-3) and Romania (-1.2 µg m-3). In Russia, similarly large decreases are observed in the western and south Siberian part of the country.~~ Despite comparable reductions in agricultural NH_3 emissions, decreases in $PM_{2.5}$ over western Europe ~~the Benelux region~~ are smaller…agricultural emission changes simulated over western Europe are weaker than over central and eastern Europe and Former Soviet Union. ~~eastern Europe~~.

L 268 – 269: …due to agricultural emissions changes over ~~eastern~~ China and ~~northeastern~~ India…

L 273: …We find that surface $PM_{2.5}$ over US, Europe and Former Soviet Union ~~(FSU)~~…

L 283 – 284: …observed over less populated areas. ~~The country definitions of each region in Table 2 are provided in Table S2 (and largely follow IMAGE 2.4 classifications).~~

L 298 – 290: … Regionally, the largest impact of land change ($\Delta PM_{2.5, LULCC+agr\_emis}$) on population-weighted annual mean surface $PM_{2.5}$ is simulated over central and eastern Europe (CEU, -1.01 µg m^{-3}), Former Soviet Union (FSU, -1.00 µg m^{-3}), South Asia (SAs, +1.71 µg m^{-3}) and China (+1.45 µg m^{-3}) ~~CEU (-1.01 µg m^{-3}), FSU (-1.00 µg m^{-3}), SAs (+1.71 µg m^{-3}) and China (+1.45 µg m^{-3})~~…

L 292 - 298: The only exception to this occurs over North America ~~(NAm)~~ where anthropogenic NOx and SO_2 emissions have declines, but agricultural emissions have increased. This suggests that the increase in agricultural emissions over North America ~~NAm~~ has partially…on the order of 5% to 12% of changes due to direct anthropogenic emissions (e.g. in ~~CEU~~ central and eastern Europe and western Europe ~~WEU~~). Notably, over Former Soviet Union ~~FSU~~, the Middle East ~~(ME),~~ and central America ~~(CAm)~~…

L 310 – 311: Particularly, over the regions experiencing rapid change in land use intensity (e.g. Former Soviet Union ~~FSU~~) or slow change in anthropogenic emissions (e.g. central America, the Middle East ~~CAm, ME~~)…

L 317: …Over parts of North America and central America ~~eastern US and central Mexico~~, the increase in dry deposition velocity…

L 320: …modelled surface ozone increases by up to 1.2 ppbv further south ~~over Bolivia and northern Argentina~~…

L 323 – 324: …However, in other parts of ~~sub-Saharan~~ Africa, up to 0.6 ppbv of surface ozone...

L 329 – 333: An exception to this is observed in the large increase in agricultural NOx emissions ~~in parts of Asia~~ which reduce surface O_3 by up to 0.6 ppbv over

NOx-saturated ~~northern~~ India and ~~eastern~~ China, but increase surface O_3 in NOx-limited parts of Southeast Asia ~~Myanmar~~ by similar magnitude. Slight increases in surface O_3 level due to increased agricultural NOx emissions are also simulated over parts of ~~East~~ eastern Africa and South America ~~southern Brazil~~. Whether the effect of agricultural emissions strengthens (e.g. ~~eastern~~ China and Sahel) or offsets (e.g. over southern Brazil and ~~northern~~ India) the effect of LULCC is largely region-dependent.

L 348 – 353: Over Eastern Africa ~~(EAf)~~, Western Africa ~~(WAf)~~ and Southern Africa ~~(SAf)~~, area-averaged ~~$\Delta O_{3,\ agr\_emis+land\_cover}$~~ $\Delta O_{3,\ LULCC+agr\_emis}$ generally has similar magnitudes to population-weighted ~~$\Delta O_{3,\ agr\_emis+land\_cover}$~~ $\Delta O_{3,\ LULCC+agr\_emis}$. In other regions, the differences between area and population-weighted $\Delta O_{3,\ LULCC+agr\_emis}$ are more substantial. The largest discrepancies between area and population-weighted $\Delta O_{3,\ LULCC+agr\_emis}$ is found over China, where increases in surface O_3 are predicted over less populated western China, while reductions in surface O_3 are simulated over more densely-populated eastern China. In South America ~~(SAm)~~…

L 356 – 360: ~~Over China, FSU, ME, WAf, EAf and SAf,~~ Over China, western Africa, eastern Africa, southern Africa, Former Soviet Union and the Middle East, the magnitudes of population-weighted $\Delta O_{3,\ LULCC+agr\_emis}$ are more than 20% of that of $\Delta O_{3,\ anth}$, implying that contemporary land system changes could be a regionally important component in contemporary trends of surface O_3. The effects of agricultural emission changes and LULCC can either noticeably enhance (e.g. over the Middle East ~~ME~~, Japan and Korea ~~JK~~, China) or offset (e.g. over South Asia~~, SAs~~) each other because…

L 380 – 388: The increase is mostly simulated over the Americas, Africa, the Middle East ~~ME~~ and China, which is partially offset the large decrease over Former Soviet Union ~~FSU~~. Meanwhile, despite agricultural changes that lead to notable ΔN_{dep}, over most of Europe, ~~eastern US, China~~, South Asia and Southeast Asia, nitrogen input from other sources are large enough that this signal alone does not lead substantial changes in N_{dep} exceedances of 5 kgN ha^{-1} yr^{-1}. However, over parts of North America, South America, Africa and China, ~~the periphery of North American Great Plain, southeastern part of South America, Nile River Delta, western China and African Savannah,~~ agricultural changes are simulated to increase N_{dep} from below to above 5 kgN ha^{-1} yr^{-1}. This implies these natural ecosystems ~~at the edge~~ of these areas are at risk of nitrogen exceedances due to agricultural changes. In contrast, the substantial reduction of N_{dep} in parts of Former Soviet Union ~~southern Russia~~ may have significantly reduce the risk of nitrogen exceedance of natural ecosystem from agricultural sources.

L 400: …LULCC over ~~northeastern~~ India and ~~eastern~~ China…

L 406 – 411: …Noticeable changes (> 1 µg m^{-3}) population-weighted $\Delta PM_{2.5,\ LULCC+agr\_emis}$ are simulated over China (+1.45 µg m^{-3}), ~~SAs~~ South Asia (+1.71 µg m^{-3}), ~~CEU~~ central and eastern Europe (-1.00 µg m^{-3}) and ~~FSU~~ Former Soviet Union (-

1.01 μg m^{-3}), indicating the potential impact of land change on long-term public health through modulating $PM_{2.5}$ level at regional scale. Our results suggest that contemporary (1996-2014) changes contribute to changes in $PM_{2.5}$ at regional and global scales that range from on the order of 5 to 10% of changes in $PM_{2.5}$ resulting from direct anthropogenic emissions over the same time period, and up to ~25% or more in Former Soviet Union, the Middle East and central America ~~FSU, ME and CAm~~ specifically.

L 417 – 421: The increase in agricultural emissions reduces O_3 over NOx-saturated parts of China and South Asia ~~SAs~~ by… enhancements of dry deposition reduce O_3 over parts of China, North America and South America ~~Rio de la Plata Basin, eastern China, and eastern US~~ by up to 1.2 ppbv. Overall, the largest population-weighted $\Delta O_{3,\ LULCC+agr\_emis}$ is simulated over western Africa ~~WAf~~ (+0.42 ppbv) and eastern Africa ~~EAf~~ (+0.47 ppbv)…

2. *Following the text on lines 152-162 and the supplement, the GEOS-Chem model appears to have reasonably large issues in the simulation of SNA. Annual mean biases of 30-50% are not necessarily consistent with the model capturing "the present-day annual means of surface SNA" as stated on Line 161. The authors should describe how these biases influence the interpretation of the results in this work (e.g., are biases in annual magnitudes sufficiently unimportant for the simulation of changes in SNA?).*

   Response: We thank the review for raising the important issue of model performance. From Fig. S1 we can see that GEOS-Chem reasonably captures the global geographic distributions of individual SNA species, but with biases in the absolute magnitudes.

   As discussed in line 272 – 276, we find that the sensitivity of SNA to agricultural emissions does vary with anthropogenic emissions, and, by extension, to background SNA concentration. Therefore, against observations in US, China, and EANET-covered regions, we recognize that the model may have underestimated the sensitivity of SNA to agricultural emissions in our scenario. The effect of biases against the Canadian network data are harder to interpret, but the changes in $PM_{2.5}$ over Canada are generally small. To address the reviewer's concern, we have rewritten line 155 – 159 of our main text as follows:

   L 155 – 159: … the same time period. ~~In general, the model captures the regional annual means of individual SNA species reasonably (Fig. S1 and Table S1), especially over US and Europe, where the bias is within ±30%. The model underestimates all SNA species over China in a relatively uniform fashion (36 – 55%). Over the region covered by Acid Deposition Monitoring Network in East Asia (EANET) (Japan, Korea and southeast Asia) the model underestimates the negatively charge ions (36% for sulphate and 16% for nitrate) while overestimating ammonium by 14%.~~ In general, the model captures the spatial distributions of individual SNA species reasonably well (Fig. S1). The model is able to capture

regional annual means of individual SNA species (Table S1) over Europe. Over the US and China, where annual means of all SNA species are underestimated by 21 – 55%, and in regions covered by Acid Deposition Monitoring Network in East Asia (Japan, Korea and southeast Asia) where SO_4^{2-} is underestimated by 36%, we expect the model mayunderestimate the sensitivity of SNA concentration to NH_3 emission perturbations. This may imply that results from our study should be interpreted as conservative. Figure S2…

3. *Despite the nonlinearity in the response of atmospheric composition to changes in surface fluxes, the changes in ozone and PM due the combined effects of agricultural emissions and LULCC (Tables 3 and 4) are nearly linear with respect to the individual process changes. Do the authors have any hypotheses as to why this might be?*

   Response: We thank the reviewer for noticing this fundamental question. We had hypothesized that the changes in BVOC emissions due to LULCC, and NH_3 emissions due to agriculture, might be strong enough to change the chemical regime of O_3 and SNA production. Therefore, we would not have ruled out the possibility of non-linear interactions between these two factors, which may indeed be present over different time scales. Yet as the reviewer notes, the effects of agricultural emissions and LULCC are approximately linearly additive over our period of interest. We expect this is mainly because of two reasons: (1) the changes in surface fluxes on this time scale are not large enough to change the O_3 and SNA chemical production regimes; and (2) LULCC mainly impacts O_3 precursors, while agricultural emissions mainly impact SNA precursors, which are in many cases spatially segregated. The interaction between these two factors in space turns out to be relatively small on the timescales investigated.

   In response to the reviewer's observation, we make note of this result in our revised manuscript:

   L 442: …despite the relative small signal that we obtain here.

   We find the effects of agricultural emissions and LULCC to be largely linearly additive over contemporary timescales, which may be attributable to two factors: 1) LULCC mainly impacts O_3 precursors while agricultural emissions mainly impact SNA precursors, and these are often spatially segregated; 2) LULCC and agriculture-related changes in surface fluxes of O_3 and SNA precursors are not large enough to change their respective chemical production regime. At longer timescale when land change signals are stronger, the effects of LULCC and agricultural emissions may be non-linear.

Minor Comments

1. *L23-25: This statement is sufficiently qualified to be nearly meaningless and could be much stronger. Your work does more than demonstrate possible impacts which imply potential importance!*

Response: We thank the reviewer for recognizing the importance of our results. We make the following change:

L 23 – 25: Our results demonstrate the ~~possible~~ impacts of contemporary LULCC and agricultural N_r emission changes on $PM_{2.5}$ and O_3 air quality, ~~which also implies~~ and the importance of land system changes on air quality over multi-decadal timescales.

2. *L143-145: Are there other LULCC impacts on meteorology which the authors think might important that aren't addressed through changing the roughness length?*

Response: We thank the reviewer for this theoretically important question, which can be clarified in two different directions. In terms of surface exchange schemes for chemical transport models, changes in canopy heights, and therefore displacement height (*h*), might also have an impact on aerodynamic resistance (R_a) in addition to roughness length (z_0) (the latter of which is considered in our study). However, these former effects from canopy height are not considered by default in GEOS-Chem, and therefore neither in our modelling study. We hypothesize the effect of *h* will be small, by considering how R_a is typically calculated in land surface exchange scheme:

$$R_a = \frac{1}{\kappa u_*}(\ln\left(\frac{z-d}{z_0}\right) - \Psi\left(\frac{z-d}{L}\right) + \Psi(\frac{z_0}{L}))$$

Since the middle of the first vertical grid of GEOS-Chem (*z*) is around 60 – 70 meters (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_vertical_grids), which is significantly larger than *d* such that $z - d \approx z$, the changes in *d* are not nearly as important as the changes in z_0. We include this argument in our supplemental material

Another important dimension is how LULCC directly impact mesoscale and large-scale meteorology through changing sensible heat and latent heat fluxes, and its subsequent influence on transport and chemistry. Similarly, agricultural emissions may also perturb meteorology mainly through aerosol and cloud radiative effects. While these effects can be important (e.g. Wang et al., 2020), assessing their importance requires comprehensive climate and Earth system modelling, was not in the scope of our manuscript but deserves to be mentioned.

To respond to the reviewer's comment, we have made the following these changes in our manuscript:

L 143-145: …The dominant surface type can be readily mapped to the 11 deposition surface type in the Wesely dry deposition model. We adopt the approach of Geddes et al. (2016) to replace roughness length (z_0) from assimilated meteorology with that prescribed for each deposition surface type. We ignore changes in in displacement height as they are expected to be much less important than the changes in z_0 (Text S1). To derive…

L 454-455: …computed LAI changes and trends, and these have been shown to be important to changes in simulated O_3 in this study and elsewhere (Wong et al. 2019). Finally, the meteorological feedbacks (e.g. changes in sensible heat, latent heat, air temperature, boundary layer height) and the subsequent effects on atmospheric chemistry and transport from LULCC and agricultural emissions are not considered in our study, which could potentially be important (e.g. Wang et al., 2020)**.**

Text S1. Considering how aerodynamic resistance (R_a) is typically calculated in land surface exchange scheme:

$$R_a = \frac{1}{\kappa u_*}\left(\ln\left(\frac{z-d}{z_0}\right) - \Psi\left(\frac{z-d}{L}\right) + \Psi\left(\frac{z_0}{L}\right)\right)$$

where κ is von Kármán constant, u_* is friction velocity (m s^{-1}), L is Obukhov Length (L) and d is displacement height (m) (Foken, 2006)**. Since the middle of the first vertical grid of GEOS-Chem (z) is around 60 – 70 meters (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_vertical_grids), which is significantly larger than d such that $z - d \approx z$ under most conditions, the changes in d sare expected to be less important than the changes in z_0.**

3. *L183-186: What is the potential size and influence of this effect on the results in this work?*

   Response: We thank the reviewer for the important question. Accurately assessing this questions requires comprehensive comparisons between LAI retrieval from static land cover map versus that from dynamic land cover map. A quantitative study may be outside of the scope of our study, but the question deserves to be addressed in our manuscript. Based on the work of Fang et al. (2013), we provide the following argument for the potential size and influence of such effect, to address the reviewer's question:

   ***L183-186:* … land use change. We note that since the relationship between satellite-derived surface reflectance and retrieved LAI depends on land cover, the use of static land cover map in long-term LAI retrievals (Claverie et al., 2016; Xiao et al., 2016; Zhu et al., 2013) may not fully capture the effect of LULCC on LAI (Fang et al., 2013). In particular,** Fang et al. (2013) **show that LAI could be substantially overestimated when grasses and crops are misclassified as forest. We may therefore overestimate dry deposition velocity over regions with significant deforestation. Such impact on biogenic emissions is secondary as biogenic emissions are typically much more sensitive to land cover type than LAI** (e.g. Guenther et al., 2012)**.**

4. *L195-196: This seems like a bigger issue than just in Southeast Asia as it relates to oil palm plantations. Presumably everywhere that relatively large land cover changes occur that are not represented in the datasets used here will be missed.*

Response: We thank the reviewer for the important question. Southeast Asia is a special case as palm plantations are very widespread in that region, and they have much higher isoprene emission than other evergreen broadleaf trees, creating a systematic bias that contradicts with the common understanding of how tropical deforestation reduces isoprene emissions. This phenomenon is relatively well-documented (Silva et al., 2016; Stavrakou et al., 2014).

It is true that intra-PFT variabilities of BVOC emissions factors can be large, although the use of PFT-based emission factor in regional and global modelling is generally justifiable (Guenther et al. (2012). We suppose this could may be of importance in other examples of LULCC that we may not be aware of, so to address the reviewer's comment, we have added this further caveat in our text:

L 455: …Though the use of PFT-based emission factors in regional and global modelling is generally justifiable (Guenther et al., 2012)**, we cannot rule out the possibility of intra-PFT variabilities of BVOC emission factors affecting the accuracies our results, which is exemplified by the inability of our model to capture the palm-driven isoprene emission increase over Southeast Asia** (Silva et al., 2016) **as discussed in section 3.**

There are minor grammatical errors throughout the manuscript, related dominantly to article use and subject-verb agreement. Some of these are listed below:

L11 "cause reduction" to "cause a reduction", "level" to "levels"

L12 "level India, China and eastern US" to "levels in India, China and the eastern US"

L14 "Across" to "across"

L35 "…introduce an enormous amount"

L340 "likely"

Response: We have made all revisions as suggested

Reference

Fang, H., Li, W., & Myneni, R. B. (2013). The Impact of Potential Land Cover Misclassification on MODIS Leaf Area Index (LAI) Estimation: A Statistical Perspective. *Remote Sensing*, *5*(2), 830–844. https://doi.org/10.3390/rs5020830

Foken, T. (2006). 50 years of the Monin-Obukhov similarity theory. In *Boundary-Layer Meteorology* (Vol. 119, Issue 3, pp. 431–447). https://doi.org/10.1007/s10546-006-9048-6

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., & Wang, X. (2012). The model of emissions of gases and aerosols from nature version 2.1 (MEGAN2.1): An extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development*, *5*(6), 1471–1492. https://doi.org/10.5194/gmd-5-1471-2012

Silva, S. J., Heald, C. L., Geddes, J. A., Austin, K. G., Kasibhatla, P. S., & Marlier, M. E. (2016). Impacts of current and projected oil palm plantation expansion on air quality over Southeast Asia. *Atmospheric Chemistry and Physics*, *16*(16), 10621–10635. https://doi.org/10.5194/acp-16-10621-2016

Stavrakou, T., Müller, J. F., Bauwens, M., De Smedt, I., Van Roozendael, M., Guenther, A., Wild, M., & Xia, X. (2014). Isoprene emissions over Asia 1979-2012: Impact of climate and land-use changes. *Atmospheric Chemistry and Physics*, *14*(9), 4587–4605. https://doi.org/10.5194/acp-14-4587-2014

Wang, L., Tai, A. P. K., Tam, C. Y., Sadiq, M., Wang, P., & Cheung, K. K. W. (2020). Impacts of future land use and land cover change on mid-21st-century surface ozone air quality: Distinguishing between the biogeophysical and biogeochemical effects. *Atmospheric Chemistry and Physics*, *20*(19), 11349–11369. https://doi.org/10.5194/acp-20-11349-2020