**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Impacts of near-future cultivation of biofuel feedstocks on atmospheric composition and local air quality" *by* K. Ashworth et al.

Anonymous Referee #1

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This study describes the impact of changes in isoprene emissions that follow from imposing land use changes appropriate for near-future biofuel production, using a tropospheric chemistry-climate model. The authors construct three emissions scenarios: 1) short-rotation coppice (SRC) scenario, where non-isoprene emitting crops are replaced with isoprene-emitting broad-leaved trees in parts of the US, Europe and Australia; 2) PALM scenario, where relatively high isoprene-emitting rainforests are replaced with very high isoprene-emitting oil palm plantations in large areas of Central and South America, Africa and Southeast Asia; and 3) PALM_NOx scenario, which is the same as 2) except with additional NOx emissions representing the processing of the oil palm into fuel (as seen by the OP3 field experiment in Malaysian Borneo). A simulation with

vegetation determined by a dynamic vegetation model (2001-2010 avg) serves as the control.

The chief impacts focused on are ozone and biogenic secondary organic aerosol (bSOA) concentrations. The authors note that the impacts of the scenarios are very small in global quantities (e.g. OH concentration, ozone burden), but are more appreciable close to where the emissions changes are made, and also depend on the season (e.g. > 1.5 ppbv ozone increases in Europe during the summer in the SRC scenario). It is argued that these "significant" impacts underline the need to consider changes in emissions of reactive compounds (such as isoprene and NOx) when assessing the overall life-cycle of biofuels.

In general, the manuscript is clearly written and includes valuable, well-researched biofuel emission scenarios, based on real-world plans and projections. This nicely builds on the more idealized studies that have preceded this one. However, I also think that there are major flaws. The paper lacks appropriate reference to the previous studies; it could give better context for its results (e.g. are pptv changes in ozone worth highlighting?); and it would seem that the most interesting results come from their ancillary CTM study, where the impact of land use on dry deposition is also included. I have provided more detailed comments below, together with some minor line-by-line comments. Overall I do think that a revised version of the manuscript might be suitable for publication in ACP, and the study could contribute to the land use-composition literature.

A. GENERAL COMMENTS

1. The literature cited is too narrow, and the introduction and comparison sections need to reference additional studies relevant to land use/cover change and atmospheric chemistry, which will better put this study in context of those that have preceded it (e.g. simple emissions perturbations, idealized land-use studies, dynamic vegetation models, anthropogenic land-use scenarios). In addition, while it has made an improve-

ment in using well-researched emission scenarios, this study essentially boils down to looking at the model sensitivity to changing isoprene emissions – an area that has been the subject of many modeling studies. So, as well as broadening the review of land-use/cover change in general, I would urge the authors to investigate these other studies (i.e. expanding on the discussion in Section 5.3).

A non-exhaustive list of suggestions is below (in alphabetical order). In reference to the above paragraph, I would urge the authors to read Fiore et al. (2005), Young et al. (2009) and Steiner et al. (2010), which all discuss how isoprene impacts ozone chemistry – i.e. both through altering production (via NOy) and loss (e.g. through ozonolysis). Young et al. (2009) would be a good reference – and good for comparison – since I believe the chemical mechanism is similar to that in the authors' model.

Arneth et al. (2009) – Isoprene emission estimates for European forest, including land use/cover change (and CO2 inhibition).

Bell and Ellis (2004) – Sensitivity study looking at impacts of (separately) doubling eastern USA isoprene and vehicle emissions.

Chen et al. (2009a, b) – Future composition over the US, including land-use/cover changes for particular scenarios.

Civerolo et al. (2000) – Land use/cover change and air quality impacts.

Fiore et al. (2005) – Sensitivity of composition to isoprene emission model. Good discussion on isoprene impact on the ozone budget.

Ganzeveld and Lelieveld (2004) – Impact of Amazon deforestation on atmospheric chemistry.

Ganzeveld et al. (2010) (and refs. therein) – Global modeling study on land use/cover change.

Heald et al. (2008) – future land use change and biogenic SOA.

Jiang et al. (2008) – land use change and climate and AQ in Texas.

Lathière et al. (2006) – isoprene emission changes from idealized European afforestation and deforestation.

Sanderson et al. (2003) – Use of a dynamic vegetation model to project isoprene emission changes, and the subsequent impact on ozone.

Steiner et al. (2006) – AQ effects from future isoprene changes in California.

Steiner et al. (2010) – Isoprene, PAN, ozone and AQ

Young et al. (2009) – Change in isoprene emissions from including CO2-inhibition, and the composition impact.

2. Although there are a couple of exceptions, on the whole, the changes in ozone and bSOA concentrations are small with the different scenarios. Can the authors please provide greater context for these changes? E.g., how do the changes compare to interannual variability (IAV)? Changes of +/-10% about a multi-year mean are not unusual, which could well mean that the impacts here are lost in the noise. Most ozone instruments would find it difficult to resolve changes of a few pptv!

Also, the word "significant" is used several times in the manuscript. I'm sure that the authors are aware that this has a reserved meaning in science writing, implying a statistical test has been conducted. I'm not sure how one can determine significance if each simulation has only been run for 1 year, but is there a longer control simulation to get a handle on IAV in the model?

3. A follow on from (2), to consider for the discussion section. When there are so many uncertainties related to isoprene, what can we really conclude about these relatively small changes in SOA and ozone? E.g. uncertainties in SOA yield, emissions, and HOx chemistry, let alone the ability of a global model to simulate regional AQ. I realize that there is an impetus to report "positive" results, but perhaps this study is suggesting that – based on these scenarios – an increase in biofuel cultivation generally has a

minor impact on atmospheric chemistry. . . .Or am I missing something?

4. It would seem that the results from the HadGEM2 experiments would have been more notable if the dry deposition impact had also been included. Is it not possible to include the deposition impact in HadGEM2? I realize that the idea is not to "contaminate" the signal with the climate impact, but this would seem an important chemical impact. To play devil's advocate, would it have been better to focus on the results from the CTM, where things can be controlled more easily, and leave HadGEM2 for the discussion on the climate effects? At the very least, I think a figure from the CTM simulation(s) should be shown.

5. Figures. Please increase the font sizes for the color bars – you could have them spanning two panels in Fig 1 (look at the "ppl shakey" documentation, assuming that you are using ferret). Adding titles to the columns (Jan, Jul) and rows ($\triangle$C5H8 etc.) would make reference easier, and would also remove the need for the titles under each panel. Also, "Latitude" and "Longitude" are obsolete as the scales are labeled with "E", "N" etc.

6. A suggestion on structure: I would rename Section 5 as something like "Non-BVOC land use effects", and move Section 5.3 to be the Discussion. This would include an expanded comparison section as well as discussion of the uncertainties, incorporating much of the content of the current Conclusions (e.g. the paragraph starting P24873, L12). This would then allow a more concise closing section.

B. SPECIFIC COMMENTS

P24858. Abstract could be shorter if the less notable results are excluded. (Unless very few of the results are notable. . ..)

P24858, L10-12. As is, I don't think that the study can say much about AQ standards, especially with the small changes and when looking at monthly-mean results (more interesting with the daily max, length of exposure to >70 ppbv etc.)

P24858, L15-. Give the percent changes in the values as well, for context.

P24859, L13. The Arneth et al. reference actually argues against settling on the 500 Tg C yr-1 emissions of isoprene, and suggests that the value is really pretty uncertain. (See point 3 in general comments too).

P24860, L23. Please provide details/reference for SOA parameterization.

P24860, L25. SSTs *and* sea-ice fields?

P24861, L7. Need to be clearer what the CTRL run is (I take it you mean the simulation described at the end of the previous paragraph. . .?)

P24862, L3. Citation for "significant emitters of isoprene".

P24862, L7. ". . .scaled in HadGEM2. . ." Scaled by 7?

P24863, L5-. What are the scaling factors used here?

P24863, L13-. The basics of the chemistry (NOx/VOC-sensitive etc.) could be shortened, but more isoprene-specific discussion should be brought in, e.g. as mentioned in point 1 in the general comments, as well as perhaps something on the HOx chemistry uncertainties.

P24864, L5-. Discussion on isoprene and SOA is a bit light on references (e.g. Carlton et al., 2009 and refs. therein), especially on the uncertainties.

P24684, L9-10. My recollection of the MIM is that methacrolein and MVK are lumped together as "MACR". Does your text mean that the mechanism has been revised to separate them? If so, this should be described in Section 2.

P24684, L10. What is meant by "increased competition for oxidants"? Do you mean from increased isoprene?

P24684, L15-. I would remove the "Global" heading and just summarize that the changes are very small when averaged this way. There is too much here on a not

very interesting result.

(P24684, L22. I would presume that "numerical noise" was extremely tiny (e.g. the difference in running the model on another computer). As stated in the general comments, the interesting thing is how the change compares to IAV, i.e. more relevant noise.)

P24865, L6. What about the impact on ozone production via NOy changes? Else, do you have ozone budget output to determine whether it is a P or L effect?

P24865, L14. Is SE Asia really all that similar to South America ("highly polluted urban areas")? As there are only 2 figures currently, I'd consider showing the results from the other areas too.

P24865, L21. What are the before/after emissions? (i.e. X Tg -> Y Tg)

P24866, L3. Change to "...a very modest reduction of 59 pptv..." (?)

P24866, L5. Can you quantify the "strong increase in ozone production"?

P24866, L11. Is it appropriate to reference cities/small countries when the grid boxes are so large?

P24866, L25-26. Can you explain what you mean by the reference to the effects of "OH depletion"?

P24867, L3. Do you mean 40% of the annual mean *change*?

P24867, L7-. As for the previous section, I'd drastically shorten the global discussion. Also, as mentioned above, I'd consider showing a figure for Australia and the US, as it will give something for future studies to reference.

P24867, L16. Citation for high NOx and ozone. (Else, are you describing the case for this model?)

P24868, L17-20. This is interesting – does it suggest that the NOx locked up in the

isoprene nitrates is not being recycled, which is what we might expect from a low VOC:NOx regime? (See Poschl et al.'s MIM paper for more on this).

P24869, L6. FRSGC/UCI model – does this include SOA?

P24869, L16. 7-9% above the concentrations in PALM/PALM_NOx? Please clarify.

P24869, L19. Smaller in magnitude than PALM?

P24870, L2-3. Citation for climate effects of land use (see typographical suggestion below as well).

P24870, L15. Citation for isoprene/temperature effect.

P24871, L9-13. Figure(s) for your US results would help the discussion here.

P24872, L4. From what base is the 4 ppbv increase? (Ditto for SOA)

P24872, L16. Isn't HadGEM2 an ESM? What is lacking in the model's capabilities?

P24872, L26. "key to atmospheric composition..." Not sure that the magnitude of the impact really demonstrates something this forceful. (Ditto for P24873, L15: "It is apparent from this work")

P24872, L29. Citation for rising NOx emissions in the tropics? (Do we know about any trend? Satellite?)

P24873, L15. What is meant by "robust global simulation"?

C. TYPOGRAPHICAL CORRECTIONS/SUGGESTIONS

Throughout. Check for consistency of "land-use" versus "land use".

P24860, L15. "SRC" is not defined yet (define L7?).

P24860, L16. Watch for consistent tense ("uses" -> "used")

P24861, L20. Is "fast low-growing tree species" really "fast-growing short tree species"?

P24861, L28. Comma after "achieved".

P24862, L2. Spell out "EIA".

P24863, L4. Units spacing/clarity: "0.34 L (ethanol) / kg (biomass)"

P24863, L20. "equilibrium" -> "balance".

P24864, L11. "...in the model. However, note that recent studies..."

P24866, L13. "...to net ozone destruction."

P24866, L24-26. "...mixing ratios generally decrease...of nitrates, although..."

P24870, L2-3. Suggestion: "Direct climate impacts from land use changes result from...[what's in your parentheses, but with a citation]. In the two palm oil scenarios, the impacts of replacing native forest.. ...

P24870, L7. Comma: "2010)),"

P24870, L9. "SE Asia" (for consistency)

P24870, L14. "in response to..."

P24872, L19-20. "Even when NOx emissions from biofuel processing are included..."

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