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

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The authors thank Referee #1 for their detailed and extremely informative comments and suggestions.

A. GENERAL COMMENTS:

1. The literature cited has been widened, using many of the studies suggested by Referee #1. The Introduction has been considerably extended and now includes: a general introduction (virtually unchanged from the original P24859, L1-23, other than to address points raised in B and C below), 1.1 Biofuels (incorporating and expanding on P24859, L24 – P24860, L4), and 1.2 bVOCs and LUC (describing previous studies C13000





on the impact of LUC on isoprene (and monoterpene) emissions and their effect on ozone and aerosol concentrations. This is further covered in the extended Discussion section (see point 6 below).

2. The changes in surface ozone and bSOA concentrations have been put clearly into context with background concentrations and % changes given. Two tables have been added to the results section (one for the oil palm scenarios and the other for the SRC scenario) in which annual mean and monthly mean changes are shown (averaged across the relevant regions), together with peak changes in each region.

The changes in isoprene emissions, and ozone and bSOA concentrations that we have simulated are a systematic change from the control run, whereas interannual variability is random (and small for the decade 2000-2009 in HadGEM2 – annual mean surface ozone varies by less than 2% of the decadal mean). Our projected changes would be expected to scale with interannual variability as they are relatively small. Therefore we do not believe it to be relevant or meaningful in this context and we have not included a discussion of interannual variability in the revised manuscript.

The word "significant" has been replaced where used inappropriately.

3. We have included a section on uncertainties in the discussion section of the paper. As we explain in this section, the results and conclusions drawn from our simulations represent best understanding at present. The two chemistry models used agree qualitatively on the changes in isoprene emissions and surface ozone concentration, giving greater credence to our findings. We have demonstrated that changes in dry deposition rates would tend to enhance the effects on ozone concentrations and therefore our findings (rightly given the uncertainties involved) give conservative values. While we agree with the referee that our results show that realistic LUC for biofuel cultivation has negligible impacts on atmospheric composition on a global basis, we believe that we have shown that local to regional scale impacts can be important (increases of 10%+ in surface ozone concentrations and 5%+ in bSOA).

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4. We are not able to re-run the HadGEM2 simulations, hence we have conducted sensitivity studies using the CTM. The results of the sensitivity studies show that the dry deposition impact enhances rather than diminishes the effect of increased isoprene emissions for the SRC and PALM_NOx scenarios. However, the enhancement is very small in both cases. Given that the CTM does not currently include SOA formation from biogenic precursors, we argue that the HadGEM results are (a) more interesting, as they have a wider scope; (b) validated (at least qualitatively) by the addition of the CTM sensitivity studies; (c) conservative (which given the uncertainties involved seems appropriate). Hence we have maintained the overall structure and weighting given to the respective model simulations. As requested, we have included a figure showing the before and after ozone increases in the CTM study (i.e. with and without changes in dry deposition).

5. Figures: The font sizes have been increased and the appearance of the figures improved following the referee's suggestions as far as possible.

6. The Discussion/Conclusions sections have been re-structured. The CTM sensitivity studies have been moved to a stand-alone section (Section 5) to give them greater emphasis. The Discussion section now includes: 6.1 Other bVOCs (incorporating and expanding upon P24873 L12-20), 6.2 Non-bVOC impacts of LUC (unchanged other than to address the points raised in B and C below), 6.3 Comparison with previous work (expanded as suggested in General Comments 1), 6.4 Uncertainties (as suggested in General Comments 3), and 6.5 Future work (incorporating P24873, L21 – P24874, L2). The Conclusions section is now much more concise.

B. SPECIFIC COMMENTS:

P24858 The abstract has been reduced by removing references to very small (i.e. global) changes, other than a statement to say they are negligible. % changes have also been included.

P24858, L10-12. Text altered to read "are substantial at the regional scale, with impli-

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cations for air quality standards." While we acknowledge the referee's argument that the use of monthly and annual mean data is less than ideal for a precise determination of air quality impacts, changes of up to 2 ppbv in monthly mean surface ozone concentrations over Europe in July will result in a deterioration of air quality and is likely to result in additional excedences.

P24858, L15-. % changes have been included in brackets after each absolute change shown in the abstract.

P24859, L13. While we acknowledge that the Arneth paper was making the case against over-confidence in the value of 500 Tg(C) y-1, it also shows that 15 different modelling studies had arrived at a figure of between 460 and 600 Tg(C) y-1. While there are many reasons not to be convinced that 500 Tg is correct, it is the current consensus and represents the biogenic modelling community's current best estimate. We have included an additional citation here (to Guenther et al.'s 2006 MEGAN paper).

P24860, L23. The bSOA mechanism applied by the HadGEM2 model is a two-product approach, with methodology as described in Derwent et al, 2003, resulting in a molar yield of 3% for isoprene and 13% for monoterpenes (Mann et al, 2010). The mechanism has now been described more fully and the appropriate citations given.

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

P24861, L7. This sentence now reads "In addition to the control run (CTRL) described above that is..."

P24862, L3. Citation (Guenther et al.'s 2006 MEGAN paper) added.

P24862, L7. The standard emission factor for broad-leaf tropical trees is \sim 4x too high for Borneo; oil palm emission are \sim 1.5x that for tropical broad-leaf trees. See next comment.

P24863, L5-. The scaling factors were calculated by replacing the isoprene emission factors for a percentage of the appropriate current vegetation type (broadleaf trees in

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the oil palm scenario, and C3 and C4 grasses in the SRC scenario) with isoprene emission factors for the biofuel crop. The full method for calculating the isoprene scaling factors has now been added as an Appendix (which includes the emission factors used for each biofuel crop type), referred to in the text.

P24863, L13-. L15-L24 have been removed. The description of the response of "NOxsensitive" and "VOC-sensitive" regions has been retained as this nomenclature is referred to in the Results section. Uncertainties and isoprene-specific issues are now discussed in the section on uncertainties in the Discussion section.

P24864, L5-. The discussion on SOA and isoprene here has been kept short (for balance with the now reduced discussion of ozone formation). More details of the SOA formation mechanism in the UKCA chemistry scheme in HadGEM2 have been included in the Model approach section (P24860-24861). The uncertainties involved in the formation of organic aerosol from isoprene (and monoterpenes) are now discussed in the section on uncertainties.

P24684, L9-10. The mechanism in the UKCA chemistry scheme in HadGEM2 has been extended beyond MIM1. The full isoprene mechanism implemented here was developed from MIM for use in LMDz. This has been clarified in Section 2 and a citation added (Folberth et al. 2006).

P24684, L10. The increased competition for oxidants is due to an increase in isoprene concentrations. This sentence has been altered to clarify this. It now reads: "As well as the direct increase in SOA from isoprene and its reaction products, higher isoprene concentrations result in higher yields of SOA from monoterpenes in the model, due to increased competition for OH."

P24684, L15-. The "Global" heading has been removed and the text considerably shortened. A table showing annual changes (global and for SE Asia, South America and Africa) and maximum monthly changes (for SE Asia, South America and Africa) has been added. The table also gives the % difference for each change to provide

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context.

(P24864, L22. The reference to numerical noise has gone as this section has been cut, as outlined above.)

P24865, L6. We would expect NOy effects on ozone production to occur downwind, as the formation of NOy enables long-range transport of reactive nitrogen. We do see and report this effect over SE Asia. We have re-phrased this sentence to make it clearer that in a low-NOx environment removal of ozone through direct ozonolysis outweighs any enhancement in the production rate. It now reads: "as expected in a low-NOx environment where the increase in destruction of ozone through direct reaction with isoprene dominates."

P24865, L14. Although there are considerable differences between the regions of oil palm planting, the responses of the different regions are qualitatively similar as stated. However, in response to the referee's comments we have included a section presenting and discussing the results for South America. Headline figures for this region have been included in Table 2. We have also done the same for Africa.

P24865, L21. Added "(from 41 to 43 Tg y-1)". The same detail has been included for all regions.

P24866, L3. Changed to "... a negligible overall impact on ozone concentrations across the region (annual mean reduction of 67 pptv)".

P24866, L5. The following lines (L6-8) give annual and monthly changes in ozone concentrations to quantify this statement. Table 2 also provides context for the changes.

P24866, L11. Is it appropriate to reference cities/small countries when the grid boxes are so large? While we acknowledge the point that the referee is making here, the NOx emissions in these grid boxes are higher than the surrounding areas due to the elevated emissions from these cities/small countries. This leads to a different response in these boxes and although the emissions are smeared across the whole grid box it is

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the presence of the cities that is causing it. Hence, we have kept the reference.

P24866, L25-26. The phrase "OH depletion" has been replaced with "enhanced removal of the OH radical". In these areas the reduction of OH radicals due to increased isoprene emissions lead to a decrease in the rate of conversion of NO2 to HNO3, resulting in higher NOx concentrations. The sentence has been altered to make this clearer.

P24867, L3. Yes – but this section has now been removed.

P24867, L7-. The global discussion section for the SRC scenario was only a single paragraph and has been retained, although the text referring to global annual mean changes has been removed. As in the oil palm results section a table has been inserted showing annual changes (global and for Europe and N America) and maximum monthly changes (for Europe and N America). The table also gives the % difference for each change to provide context.

A figure and section has been added for N America as suggested. A brief paragraph has been inserted stating that for Australia the impacts were negligible at all temporal and spatial scales.

P24867, L16. HadGEM2 characterises Europe as hi-NOx, hi-O3 – this has now been made clearer.

P24868, L17-20. We have re-structured this paragraph for greater clarity. While it is likely that some of the effects observed are due to the treatment of isoprene nitrates within the model, we cannot easily quantify this. Isoprene nitrate formation and recycling is an important source of divergence between isoprene chemistry schemes, and has been included in the section on uncertainties included in the Discussion section.

P24869, L6. The FRSGC/UCI CTM does not include formation of SOA. The additional sensitivity study performed with the CTM considered changes in ozone concentrations only – this has now been made clear in the text.

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P24869, L16. The increases are 7-9% above the increases in PALM_NOx. The sentence has been altered to make this clearer: "The increases in ozone simulated by the FRSGC/UCI CTM for the PALM_NOx scenario are around 7-9\% higher when deposition is allowed to change than the increases projected in simulations in which deposition does not alter."

P24869, L19. The phrase "than PALM" has been added for clarity.

P24870, L2-3. Citations of biogeochemical and biogeophysical effects of LUC added (Bathiany et al., 2010 and Davin et al., 2007 and references therein).

P24870, L15. Citation added (Guenther et al., 1995 and Arneth et al., 2007)

P24871, L9-13. A figure showing the changes in annual mean surface concentrations of ozone and bSOA for the US has been included in the results section to permit comparison of the spatial distribution of the changes.

P24872, L4. The base concentrations have been shown for all changes in ozone and bSOA concentrations reported in this section.

P24872, L16. HadGEM2 is an ESM. However, in this study its atmosphere-biosphere couplings were not implemented. The initial description of HadGEM2 has been altered to better reflect the configuration used. Section 2 now reads "In this study HadGEM2 was run in its climate configuration, so the changes in atmospheric composition and climate do not feedback onto biogenic emissions."

P24872, L26. "will be key to atmospheric composition" had been altered to "play an important role in". (P24873, L3 "It is apparent from this work" has been replaced with "This work suggests").

P24872, L29. The reference to rising NOx levels in the tropics has been replaced with "if background levels of NOx rise in the tropics as a result of increasing industrialisation"

P24873, L15. We intended "robust global simulation" to mean a simulation in which we

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could have some confidence in the results. The entire sentence has been re-worded to make this clearer. It now reads: "While a robust simulation of the emissions and atmospheric reactions of these compounds is not possible at present, due to the large uncertainties involved, ..."

C. TYPOGRAPHICAL CORRECTIONS/SUGGESTIONS:

Consistency of land use vs. land-use - We have adopted land use throughout.

P24860, L15. SRC replaced with "short rotation coppice" here.

P24860, L16. "uses" replaced with "was performed with"

P24861, L20. This phrase has been changed to "fast-growing tree species"

P24861, L28. Comma inserted.

P24862, L2. Citation altered to read "Energy Information Administration" rather than "EIA".

P24863, L4. Units altered as suggested.

P24863, L20. "equilibrium" replaced with "balance".

P24864, L11. Sentence split as suggested.

P24866, L13. "leading to a shift to ozone destruction" replaced with " leading to net ozone destruction".

P24866, L24-26. "mixing ratios decrease over most of the region due to increases in the formation of nitrates although" changed to "mixing ratios generally decrease due to increases in the formation of nitrates, although"

P24870, L2-3. Text amended as suggested, and citation added.

P24870, L7. Comma inserted.

P24870, L9. "South East Asia" replaced with "SE Asia" as suggested.

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P24870, L14. "to" added.

P24872, L19-20. "NOx processing emissions" changed to "NOx emissions from biofuel processing" as suggested.

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