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***Interactive comment on* “The influence of boreal forest fires on the global distribution of non-methane hydrocarbons” by A. C. Lewis et al.**

Anonymous Referee #2

Received and published: 12 October 2012

General comments:

The paper presents airborne observations of non-methane hydrocarbons in Eastern Canada, and derives biomass burning emission ratios for 29 species. The paper then uses the GEOS-Chem model to investigate the impact of northern biomass burning emissions on global distributions of benzene, toluene, ethene and propene, in particular the relative influences of biomass burning and anthropogenic sources at various WMO-GAW observation stations.

The paper presents a fine data set then gives an interesting analysis on the implications for understanding NMHC signals both at background monitoring stations as well as elsewhere in the global environment. A nice feature of the study is applying results from a regional airborne study towards understanding global hydrocarbon measurements.

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There are however several areas where the arguments need to be strengthened, in particular by recognizing and tying the paper into the substantial existing literature, and by including a quantitative discussion on model error and the limits to which the model results can be extrapolated. For example, whereas the model needs to reduce its anthropogenic source by 2/3 to agree with the observations, from the literature it seems highly unlikely that reduced NMHC emissions via catalytic converter use can account for this large discrepancy. A second point is that non-combustive emission signatures are evident in the data for many of the observed NMHCs, but these have not been recognized and it's unclear whether or not the model addresses them, and what impact this may have on the analysis.

Specific comments:

P23434, L12: I would disagree that benzene, toluene, ethene and propene are generally considered to be indicative tracers of anthropogenic activity, since all four are also well-known to be emitted by non-anthropogenic combustion processes such as biomass burning. Same comment on P23442, L22.

P23434, L14: It may not be feasible, but it would be interesting for you to run your model on ethane. Ethane is one of the better-studied NMHCs and has a fairly simple source budget that has been quantified in previous work (Y. Xiao et al., JGR, 2008; A. Pozzer et al., ACP, 2010). The biomass burning source causes significant variability in ethane's global abundance (I. J. Simpson et al., GRL, 2006), and it would be interesting for your research here to build on this existing work.

P23434, L17: I agree that tailpipe emissions of benzene have declined; is there evidence that solvent emissions are also declining, especially on a global basis? It is also worth discussing the competing effects of catalytic converters reducing benzene emissions in some urban areas versus the increasing use of vehicles on a world-wide basis. What might the net effect be?

P23434, L23: Change “parts per trillion range” to “parts per trillion to parts per billion

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range”?

P23435, L6-9: Reference this statement.

P23435, L16: I'm not sure I agree with this statement. There's a large body of literature in non-urban settings dating back to the 1970s, from routine long-term monitoring to biomass burning to oceans, etc.

P23435, L21: Please reference the statement about industrial/domestic solvent reductions. Also these statements seem to apply to the situation in developed countries. What about developing countries?

P23435, L25-29: It would be appropriate to mention the long-term global NMHC record from UC-Irvine dating back to the 1980s, which includes the target compounds listed on P23435 except ethene (M. L. Gupta et al., JGR, 1998; I. J. Simpson et al., Nature, 2012). Because long-term global NMHC records already exist, “gap” may not be appropriate and on P23436, L9 you could remove “emerging”.

P23436, L4-6: “Observations of NMHCs in remote terrestrial or oceanic environments are less easy to categorize in terms of contributing sources than comparable urban measurements.” What is the basis for this statement? For example A. Pozzer et al. (ACP, 2010) successfully modeled NMHC concentrations at a number of remote sites, and I. J. Simpson et al. (GRL, 2006) broadly quantified the influence of biomass burning on fluctuations in background ethane concentrations.

P23436, L22: “Boreal forests...account for roughly one third of the total global forested area...” This statement needs to be referenced.

P23436, L23: “CO₂, CO and CH₄ are the largest emissions by mass with CH₃OH and HCHO the most dominant organic compounds.” This statement needs to be referenced.

P23438, L8: What do you mean by “flight sector”?

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P23438, L9: Somewhere in this paragraph you need to state the dates of this project, e.g., August 8-22, 2012.

P23439, L7: Instead of “approximately 50:50” state the exact split ratio, to show it is precisely known and reproducible.

P23439, L14: The target operating limits need to be specified here, at least for benzene, acetylene, ethene and propene. What are the precision and accuracy of the reported measurements?

P23440, L5: This classification scheme doesn't allow for anthropogenic signatures without a combustive source (see below).

P23440, L18-21: “A range of plumes of different ages are encountered in this study, from overhead passes to several days downwind, however in this study general plume age does not appear to substantially change the slope obtained of any given NMHC against CO.” Do you have a figure showing this? Also by “overhead passes” do you mean “plume encounters”?

P23441, L11-13: “Notable outliers include propene, which is observed in this work to have an ER around double that reported in Simpson et al. (2011)...” This appears to be stated backwards: “double” should read “half”.

P23441, L21-22: “For alkanes, anthropogenic and biomass burning the NMHC:CO relationships are rather similar, and in isolation are difficult to disentangle.” It seems to me you actually do have clear anthropogenic signals, but these have been incorrectly grouped with the background data. Many alkanes have non-combustive anthropogenic sources, which can be seen as increasing mixing ratios at low CO levels, i.e. the clear wings in all 6 panels of Figure 2a. The presence and contribution of these non-combustive anthropogenic sources needs to be addressed.

P23441, L23-25: “Here the differences between elevation caused by anthropogenic emissions and those caused by biomass burning are much clearer.” Again I would

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suggest it's equally clear in both cases, just the signal you are looking for is different because of the evaporative vs. combustive anthropogenic influences for alkanes vs. alkenes.

P23441, L26: Is there a way to show the reader the older vs. younger plumes?

P23442, L5: Here you also seem to have some non-combustive signal for benzene, toluene, etc., though not as many points as for the alkanes.

P23442, L18 and P23443, L1-2: Following from the above comments, does your model incorporate the non-combustive sources of benzene and toluene? If not, what level of uncertainty does not including these sources introduce?

P23442, L22-25: "Their large emission rates from biomass burning are potentially of growing significance given that all these species are on downwards emissions trajectories in developed countries,...". Again how does an increasing global vehicle fleet potentially offset these downwards trends in developed countries? What is the net effect? It just seems that this sentence is missing a couple of arguments.

P23443, L12: Change "emissions factors" to "emission ratios".

P23443, L15: I would change "excellent" to "good to excellent" or "good to excellent for the four selected tracers" because there is still the factor of two difference for propene.

P23444, L2-6: This sentence is extremely confusing, mostly because the global emissions follow a discussion of ERs. Please rework. Also present the toluene results.

P23444, L14: As you have stated elsewhere, the short lifetimes of your target compounds means that their concentrations in the atmosphere are highly variable. Some discussion of the variability encountered at the remote WMO GAW stations would be helpful. For example in Figure 3 I would prefer to see actual data points together with the average ... there must be data points that fall outside the grey boxes?

P23444, L25-27: Again the discussion would be strengthened if there was some sense

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of the relative effects of emission reductions in developed countries vs. possible increases elsewhere. Instead of “dramatically” can you be quantitative? A two-thirds reduction in the anthropogenic source seems very extreme rather than justifiable, especially if the model does not include the non-combustive anthropogenic component. Have any of the urban areas in developed countries achieved a two-thirds reduction in benzene between 2000-2010? If not then it seems something else is going on with the model that should be discussed. For example G. J. Dollard et al. (AE, 2007) only observed a 20% reduction in benzene levels from 1993-2004 in the UK.

P23445, L5: Is Figure 4 showing the anthropogenic benzene with the two-thirds reduction? Either way this needs to be clearly stated. The sentence seems to suggest not, which would then make the right panels in Figure 4 have very large uncertainties. At very least the text and figure caption need a substantial discussion of the uncertainty in these results.

P23445, L9: The paper would benefit by having the results of Figure 4 being compared to existing measurements from the literature, to see if the model results are reasonable. For example A. K. Baker et al. (AE, 2008) measured benzene levels in 28 US cities from 1999-2005 (summertime measurements); G. J. Dollard et al. (AE, 2007) report benzene concentrations in the UK (annual means); there are studies in Asia in the 1999-2000 timeframe; and so forth. Please fold a comparison with the literature into the discussion.

P23445, L17-21: Another factor to consider is interannual variability in biomass burning emissions, which can substantially affect global NMHC concentrations even for compounds with a large anthropogenic source relative to the biomass burning source (e.g., I. J. Simpson et al., GRL, 2006).

P23446, L7: Clarify “biomass burning is high”.

P23446, L8-9: This sentence needs a reference. On L11 quantify “exceptionally low”.

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P23446, L15: Again it would be helpful to discuss the interannual variability of biomass burning emissions. The contribution of biomass burning to the abundance of various NMHCs can change significantly from year to year.

P23446, L17-19: Can you be more specific? A few examples or references would help.

P23446, L28: How low? What is the numerical resolution of the model?

P23447, L3: I suggest changing “there is some potential” to “it appears that”. I’m also not sure that the use of “now” is correct because it suggests that model used to be accurate (in 2000), whereas the declines of benzene in urban areas do not appear to have been large enough to justify the 2/3 reduction (see comments above). In other words attributing the model’s overestimate to recent emission reductions seems to be an oversimplification that is not substantiated by the literature. Please rework the text to discuss this.

P23447, L20-22: Same concerns as above. Please revise to make sure the sentence accurately reflects existing work.

P23448, L11: Given that the model needed to be adjusted by 2/3 to reproduce the observations, what kind of uncertainty do you expect in the graphs generated in Figure 8? This needs to be thoroughly discussed before presenting Figure 8.

P23448, L15: The station at Barrow is equipped with measurements that would let you verify the statement that these sites are subject to significant impacts from biomass burning. For example what does an analysis of the CO data at Barrow reveal? What do the existing measurements and modeling of CH₄ show? Or, what would be the expected impact on CO for a 500 pptv increase in benzene from biomass burning? Has this been previously observed at Barrow, and if so how frequently?

P23448, L22-25: What is the estimated uncertainty in this analysis? This needs to be addressed before the conclusions on lines 25-27 can be drawn. In general the results presented in this paragraph don’t seem very meaningful given that they completely

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change depending on which version of the RETRO model is used.

P23448, L28: Seems like “may be” should be changed to “are very likely”.

P23449, L14-16: Same concern about these statements as before.

P23449, L16-19: A thorough, quantitative discussion of model error needs to be provided before this conclusion can be drawn. Same comment for L22-24.

P23449, L19-20: Again, it seems difficult to make any conclusive statement about the anthropogenic source given its large uncertainty in the model, and without testing the plausibility of the model output with data and analysis from the literature.

P23449, L20-22: “...benzene...is often considered as having no safe lower limit.” This statement needs to be referenced.

P23449, L27: It’s unclear how “generally reasonable” can be concluded when the anthropogenic source needs to be decreased by 2/3 to correctly reproduce the seasonal cycle.

Technical corrections

There are lots of little typos that need to be fixed.

P23434, L14: Change “propene” to “propene levels”.

P23434, L18: Define “WMO-GAW” for the reader and don’t capitalize “Global”.

P23434, L26: Need to define “OH”.

P23435, L17: Change “overwhelming” to “overwhelmingly”.

P23435, L24: Choose whether or not to capitalize the book title and change “, Koppman (2007)” to “(Koppman, 2007)”.

P23436, L7: Consider switching “man-made” to “anthropogenic” (gender-neutral).

P23437, L1-3: There is a mix of commas and semicolons in this sentence; please unify.

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P23437, L2: Change “transport” to “transport by”.

P23437, L13-15: These references seem to be in random order.

P23437, L24: Change “background” to “background concentration”.

P23437, L24-25: Remove the inner set of brackets from the references.

P23437, L27: Change “whether” to “whether or not”; otherwise the nature of the disagreement is unclear.

P23438, L1: BORTAS needs to be defined here, not on L6.

P23438, L13: Define “FAAM”.

P23438, L13: Change “data ... is” to “data ... are”.

P23438, L15: Do not capitalize “whole air samples”.

P23438, L23: “in turn” needs a comma after as well.

P23438, L27: Suggest changing “I” to “L”.

P23439, L15: Define “OVOC” here, not on L16.

P23439, L20: The sentence will read better if you change “over” to “in comparison to”.

P23439, L24: Change “i” to “in”.

P23439, L26: “attributes of plume” is incomplete.

P23440, L5: “GC-MS” and “GCMS” both used in this sentence; choose one.

P23440, L6: “200ppb” needs a space.

P23440, L13: Change “emissions ratio” to “emission ratio”.

P23441, L16: Sentence needs a period.

P23441, L21: Omit “the”.

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P23442, L7: No need to capitalize “emission factor”.

P23443, L11: Change “during experiments” to “during the experiments”.

P23443, L20 and elsewhere: Change “GFED III” to “GFED3”.

P23443, L23: Define “RETRO”.

P23444, L15: Capitalize “watch”.

P23447, L10: Change “is show” to “is shown”.

P23447, L14: Omit “these”.

P23447, L16: Change “will grow” to “will enhance”.

P23448, L4 and elsewhere: Do not capitalize “global”.

P23448, L22: Change “that is biomass” to “that is from biomass burning”.

P23449, L8: Change “emissions estimates” to “emission estimates”.

P23449, L25: Change “data exists” to “data exist”.

P23450, L4: Qualify “are predicted”.

P23450, L5: Change “with significant” to “with a significant”.

Table 1: Sometimes compounds in Table 1 are capitalized, sometimes not. Please review for consistency. Same comment for Figure 2c y-axis labels.

Figure 2a: I’m not sure a Figure with 6 panels can be called “Figure 2a”. It should probably be Figure 2 (a-f) and then “Figure 2b” should be Figure 3 (a-f) and so forth. Also the axis labels look quite scratchy and the figure needs an x-axis in the top four panels (even if it is unlabeled).

Figure 2a: Change “triangle” to “triangles”.

Figure 3 caption: Capitalize “observatory”.

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Figure 3 caption: Change “the estimated benzene reducing benzene in RETRO” to “the estimated benzene by reducing the benzene levels in RETRO”.

Figure 3 caption: Even with the multiplication dots, the wording make it seem like you are reducing the RETRO emissions by 20% and 33%, rather than multiplying them by 0.2 and 0.33.

Figure 6 caption: Change “with fit” to “with the fit”.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 23433, 2012.

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