**Atmospheric
Chemistry
and Physics
Discussions**

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

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We thank the reviewer for their thoughtful and detailed comments on this paper. We have made all of the minor typographic and textual corrections suggested by the reviewer. We address all of the more substantial comments individually below in this document and hope that our additions and changes make the paper acceptable.

P23434 L12 We have modified this statement somewhat with a qualification but would contend that in general terms these species are indeed used by many in the atmospheric science community as anthropogenic tracers. We don't disagree that the biomass-burning source is well documented in literature going back many years. We have also made a qualifying comment in the section 'Model analysis and global impacts. 'We chose these species in particular for study since they are often (if erro-

neously) considered as anthropogenic tracers rather than from natural emissions.'

P23434, L14. We haven't run the model here for ethane since this species has already been evaluated in terms of global sources and distribution using the GEOSCHEM model see: Yaping Xiao, Jennifer A. Logan, Daniel J. Jacob, Rynda C. Hudman, Robert Yantosca,Donald R. Blake, Global budget of ethane and regional constraints on U.S. sources, JGR, 113, D21306, doi:10.1029/2007JD009415, 2008

P23434 L17. Using data from the UK National Atmospheric Emissions Inventory as a reasonable proxy for developed world benzene emissions, there has been a very significant reduction in solvent and production usage of benzene over the past 20 years. It is now approximately 1/3 the size of emissions in 1990. We have added a commentary on this to the text in the section on Evaluation of the model against data.

P23435 L6-9 References added.

P23435. We have changed this sentence to reflect the current position regarding automated measurements, rather than infer that this has always applied for any kind of NMHC measurement. Long-term hydrocarbon measurements are now predominately made in urban and sub-urban settings for air quality reasons, accepting that a small number of research grade observations are made also in more remote environments.

P23435. Global data on emissions, disaggregated by both sector and hydrocarbon species unfortunately does not exist, so it is difficult to make informed comment on trends in developing countries. However at a national level, using the UK as an example, exhaust emissions and solvent usage have decreased to around $\frac{1}{4}$ and 1/3 respectively of the emissions in 1990 (see for example http://naei.defra.gov.uk/). Trends in developing countries are more difficult to estimate given a lack of observations. It may be a reasonable assumption that catalytic controls may not have as effectively penetrated vehicle fleets in these locations and growth in traffic volume may outweigh control measures. It is potentially reasonable however to assume that there has not been an increase in benzene use as a solvent since it has been replaced effectively

by other less harmful solvents. We have added this short discussion to the text in the introductory section.

P23435, L25-29. We have now included the reference to the long-term record reported in Simpson et al. 2012, a paper that was not published at the time we submitted this manuscript. We would contend that just because a very small number of long-term observations of NMHCs have been reported, perhaps four worldwide, that a gap in data does exist. We have however changed this sentence to refer to the limited extent of current measurements and have removed the word 'emerging' as requested.

P23436 L4-6. Our point here was that the sophistication in data analysis is much greater when trying to attribute sources to background locations, as indicated by the methods used in the papers the reviewer refers to. We have added to the text to make this clearer.

P23436 L22 and L23. References added

P23438. L8. A flight sector is defined as one take off and landing cycle, we have clarified text.

P23439. L9. Exact values are given and a clarification.

P23439, L14. Precision and accuracy estimates are now given in the text for ethene, propene, benzene and toluene specifically.

P23440 and other later comments eg 23441 and P23442, L18. We think the reviewer has slightly misunderstood how we have used field data to inform the later modelling. There are indeed contributions from non-combustion sources visible in our data set. These are sometimes seen in the scatter plots as an elevated NMHC with no corresponding increases in CO – n-butane is the clearest case. Our use of an upper threshold value in CO is so that we specifically exclude such data from our assessment of the NMHC:CO emission ratio from biomass burning. By using a high CO threshold we are only including hydrocarbons with a clear combustion source. If we fitted our

slopes to the entirety of the dataset then the non-combustion sources would indeed skew the values obtained, but we don't, we limit ourselves to occasions only when CO is greater than 200ppb and with a qualifying biomass burning tracer from GC-MS. Our later modelling uses this biomass burning combustion source as input data. We DO NOT use other field data to try to estimate, for example non-combustion sources of hydrocarbons. These are included in the GEOS-Chem model, and are taken from the RETRO emissions database and include both combustion and evaporative/solvent sources. We have added detail on the anthropogenic sources of benzene in RETRO.

P23440, L18-21. We have not attempted to 'date-age' our plumes in this study, and based this statement on simple trajectory ages of plumes. We have added to the text to describe this. When flights are plotted individually (reflecting a range of plume ages) the slopes obtained are the same as that obtained from the composite of all biomass burning plume data. We haven't included flight by flight data for reasons of brevity but have described this in the text.

P23441, L11. Our mistake, this is corrected to read half.

P23441, L21-22. The reviewer is quite right that there are some elevated alkanes which do not have CO enhancements. These are indeed most likely to be from non-combustion sources and we have now made reference to these. We have also been clearer that each alkane has slightly different behaviour and accept we overgeneralised their relationship to CO.

P23441, L23-25. We have modified the text to draw attention to the difference in anthropogenic emission sources for the alkanes and alkenes.

P23441, L26. We haven't attempted to place a quantitative value on plume age in this study since it doesn't add to what we are attempting to demonstrate in terms of global distribution.

P23442, L5. The reviewer has a keen eye and is correct that there are a handful of

benzene values with some elevation but without corresponding elevation in CO. We have drawn attention to these and the fact that this suggests a localised evaporative source of benzene somewhere in the working region of Northern Canada.

P23442, L18. Yes the model includes all anthropogenic sources of benzene and toluene, both combustion and non-combustion, as defined in the RETRO methodology. We don't disaggregate them in our modelling however, hence throughout we refer to them as simply 'anthropogenic'. We have made this clearer in Model analysis section.

P23442, L22-25. This was poorly worded on our part. We have changed to read 'The substantial emissions from biomass burning are potentially of growing significance in those locations (particularly in developed countries) where such species are on downwards emission trajectories, a result of control technologies, reformulation of gasoline composition and reduced solvent usage.' It is very difficult to make any firm statements about impacts on developing countries since we have so little data to work with, however we have added: 'In those locations with rapidly growing vehicle fleets in developing countries it remains an open question as to whether benzene is on an increasing or decreasing trajectory.'

P23443, L12. Changed as suggested.

P23443, L15. Changed as suggested.

P23444, L2-6. We have re-written this section to improve clarity and included the toluene values.

P23444, L14. The variability in benzene at the Cape Verde observatory is not so great, and the grey boxes represent the standard deviation values. The data we use here is however filtered to remove all measurements made under the influence of local emissions, and represents only background oceanic benzene variability. We have clarified this in the text.

P23444, L25-27. As referred to earlier we have added new material providing some

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quantitative estimates of reductions from various sectors for benzene. A lack of accessible data hinders us, but we have used the UK emissions inventory as a proxy for other developed countries. Some sectors, such as evaporative emissions of benzene are estimated to have fallen by even more than our 0.33 factor used here. The reviewer rightly points to papers that shown that ambient levels have fallen more modestly. Again using data we have access to, we now include detail that suggests that UK ambient benzene is now around half the value in 1990.

We have also added 'We would stress here that we are not suggesting our model proves that RETRO should be adjusted by a multiplier of 0.33, but rather that this is the level of change that is required to make a remote observation match with our model.'

P23445, L5. We think the text is clear that it refers to the RETRO 2000 scenario. We have reiterated this in the text and also the figure caption.

P23455, L9. We have looked at this suggestion seriously however it is not so straightforward as first appears. The measurements for example in Baker et al are not seasonally disaggregated and cover measurement periods in different years for different cities over the period 1999 to 2005. It is perhaps also important to note that we are modelling here on a global scale whereas the observations in Baker et al and also Dollard et al., are in some cases roadside. We are not therefore comparing like with like in way that adds much value. At best one might conclude that our model result and those of the two studies highlighted both indicate urban benzene concentrations greater than 100 ppt but less than 500 ppt in general terms. We added a short discussion of this nature to the text.

P23445, L17-21. We would agree fully that inter-annual variability is another important factor to consider and we have added a comment on this to the text.

P23446, L7. Now clarified.

P23446, L8-9. We have made it clear that we are referring to our model that shows

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the largest benzene source is from biomass burning, and indicated that, in this context, very low means atmospheric concentrations that approach the detection limit of current instrumentation.

P23446, L15. We have added some text earlier on the potential impact of inter-annual variability, but it is beyond the scope of this paper to study multiple years of potential emissions. To do this in a meaningful way would require simulations of perhaps 10-20 different years. We have commented that increasing observations would inform greatly this assessment of variability.

P23446, L17-19. We have added some examples as suggested.

P23446, L28. Numerical resolution now added to the text. The model handles concentrations down to 0.01 ppt, but a further issue here is that we are calculating a ratio of two small numbers, which can tend to a value below the model resolution.

P23447, L3. We have reworded this section to highlight some of the uncertainties the reviewer raises and to stress the uncertainty of using a single background measurement location to constrain emissions.

P23447, L20-22. We have clarified that whilst there are research grade measurements made throughout the troposphere, most routine and long-term datasets exist for urban centres, made as part of air quality activities.

P23448. The plot in Figure 8 uses the original RETRO estimates and we have clarified this. We have also added some text highlighting that reality is likely to fall somewhere between the two scenarios. We show the impact of moving to a 2/3 reduction in tabular format in table 4.

P23448, L15. It is an interesting suggestion to compare Barrow CO to the model, although we are limited only to that data which is publically available. Using information from the NOAA ESRL website we observe that the 2009 maximum in Barrow in CO was 678 ppb, compared to an estimated values of between 600-800 ppb from the model.

We include this comparison in the text.

P23448, L22-25. We have added some further text highlighting where the major sources of uncertainty arise from in this study.

P23448. Changed as suggested.

P23449, L14-16. Changed to 'We chose these species in particular since are often controlled in an air quality context and are species with generally declining urban concentrations in developed economies.'

P23449, L19-20. The largest sources of model uncertainty are in the emission estimates. We show however the effects of a substantial range of anthropogenic emissions using the RETRO and 0.33 x RETRO scenarios. We do not model different biomass burning scenarios in this study, but note that our global CO value is at the lower end of suggested values. This large range of emissions does not however change the broad conclusions of the paper, that in the Southern hemisphere biomass burning hydrocarbons are a very significant source and that in the Northern hemisphere, in some background locations, biomass burning can be important. We have added text to this effect in the conclusions. We agree fully with the reviewer that a comparison with more measurements would be very useful in helping narrow uncertainty. But as discussed previously there is very limited value in attempting to compare city centre benzene from automatic air quality networks with a 2.5 degree global model. We have compared against the only background benzene data we have data access to, but of course would like to see additional similar locations developed through GAW.

P23449, L20-22. Reference now added.

P23449, L27. We have clarified this as: 'Very limited experimental data exist for the long-term trends in hydrocarbons in the background atmosphere, however comparison of model against benzene in the tropical Atlantic shows that the annual cycle can be reasonably captured, although suggesting that the model emissions database is over

estimating anthropogenic emissions.'

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