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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emissions from biomass burning in the Yucatan" *by* R. Yokelson et al.

Anonymous Referee #2

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This paper presents a detailed and thorough analysis of emissions of a wide range of trace gas and particulates from fires in the Yucatan peninsula region of Mexico during the MILAGRO study. The study presents a detailed analysis of the emission ratios of a wider range of species than had previously been measured and also provides valuable additional information on emissions of previously measured species. An analysis of the development of several trace gases and aerosol components in the plume of a fire as it advects away from the source is discussed. Changes in chemistry and composition are observed, though the changes appear rapid and non linear this not appear to be recognised in the text and the impression is given that changes occur throughout the age of the plume. I see no evidence that this is the case. Regional emission estimates are made and compared with emissions from other major sources in the region. This is a very valuable paper and should certainly be published as it contains a wealth of information. I do, however, have some comments that I would like the authors to

address before the paper is accepted for ACP.

The AMS Collection efficiency has not been stated nor have the assumptions on which it is based.

It would be useful to incorporate details of the formation processes of the species for which emission ratios are discussed where they are known and highlight where they are not rather than just discussing the ratios. This is done for some species but not for all. This will give the reader an understanding of the co-variability and changes in emission ratios that are likely as fuel type and burn condition vary.

To what extent is it useful to report emission factors of species that are produced as secondary products via photochemistry in the plume? For example, H2O2 and H2SO4 EF are reported, are the authors suggesting that these are emitted by the fire or formed in the plume? If the latter then what is the value of an EF?

I see little evidence for the validity of trend lines and regressions for many of the changes in concentration ratios shown (in particular HCOOH/CO, fig 7a; PM2.5/CO, fig 9a; aerosol component/CO except for possibly NO3/CO, fig 10). Clearly there are marked changes between the measurements made at the point of emission and those in the advected plume and these changes are clearly important. However, thereafter there is much scatter in the data and as far as one can see there is a lack of observable trend in most of the plots. The trend line has been forced by fixing the line at the intercept based on the increased number of observations in the near field of the fire. It is important to recognise that more reliance can be placed on these data (I suggest the variance in the measurement is also shown, see below). I do not see how a trend line can usefully be applied as it implies that the changes are continuously occurring on the timescale of a few hours. One could equally argue that rapid changes take place over the first 15 minutes in many of these ratios but thereafter there is no sign of any further significant changes. The authors comment that OH measurements are a factor of 5-20 times greater in the near field plume than the background but thereafter fall to a

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factor 2. This supports the hypothesis above that the changes are distinctly nonlinear. The TEM analyses also shows that very significant changes are taking place on time scales of less than 30 minutes. A discussion to reflect this would be useful as it also possibly goes some way to explain other data on regional biomass burning, for example Capes et al. Certainly the data indicate that further work is necessary in this area to understand the apparent non-linearities in the near-field and the authors are right to point this out.

The authors comment that there is marked fire to fire variability and that comparing near and far field measurements of different fires can be misleading. This is done by Capes et al., 2008 in west Africa. In the Capes et al paper many different fires are sampled from close to fire sources to several hundred kilometres from the sources. The striking thing about these results is that the ratio of particulate organic matter to CO shows very little variability over such a wide range of space scales. If the fire to fire variability shown in this paper is as large as the authors here suggest then it would be very difficult to se how a relationship such as that observed by Capes et al could ever be observed. It would be good to see a plot of organic mass to CO for the fire impacted air masses across the whole region in a similar way. This would identify whether there was intristically more variability in the Yucatan fires or whether the differences between fires average out between the fires. Capes et al state that many small fires were present across the region and it is possible that the variability between fires was averaged out.

Page 771 line 19: The authors state that Only a few observations of the chemical evolution of BB smoke have been made. It is also worth citing the measurements of Abel et al., who investigated changes in aerosol properties downwind of fires in southern Africa and Capes et al., who discuss aerosol evolution over the West African Sahel (Capes et al., 2008) at this point.

Pg 773 line 9-11 The authors state that - The nephelometer was not available on the 12 March flight so we used the UHSAS particle counting/size data to indirectly determine particle mass. The UHSAS does provide an indirect measure of particle mass but it is

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hard to see how the assumptions are any more uncertain than those involved in determining mass from the nephelometer, a methodology that also relies on assumptions about the invariance of optical properties and shape.

Page 776 line 9: reflect the degree - should be reflects

Page 783 line 9: Andrea should be Andreae

Page 786: What are the implications of using acetonitrile as a tracer for BB if the emission ratios are indeed varying by more than a factor of 2.

Page 786: The emission ratios of HCN from the two aircraft are a factor of 3 different yet the variability within the fires sampled by each aircraft is much less than this difference. Both estimates are a factor of 2 different from that in Brazilian deforestation fires. What is the value in simply averaging two different estimates to get a result closer to that of the Brazilian fires? Are the data from the Twin Otter different for DF and CR fires? What might explain these differences especially as the suggestion from the particulate data is that the two aircraft sampled a similar mix of fires (pg 787 line 16)?

Page 786: line 22-25: To what extent does the temperature of the fire impact on the NOx emission and does this account for some differences or mean that the N content must be considerably higher in the Yucatan than in Brazil?

Page 789 line 20: This is not true if cloud is present as is the case on several flights. Is the sulfuric acid EF consistent with this finding?

Page 789 and 790: The discussion of organic mass emissions should include the study of Capes et al in the west African Sahel who also used AMS measurements to derive OC:CO. The numbers presented appear to be largely consistent with their estimates though the west African emission ratios appear a little higher.

Page 794 line 25: which of the points are cloud impacted?

Page 794-795: The intercept in some of the regressions (figures 5, 7, 9 and 10) are

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forced to zero on the grounds that the near field was sampled multiple times and so the authors place greater emphasis on the representative nature of the average of the near field samples than the points obtained in the ageing plume. This is a reasonable thing to do as long as the trends appear linear but is not always the case. Moreover, it would be good to see the variability of the source data possibly expressed using an uncertainty bar that reflects the variation in measurements at source. This would give some idea of the overall variability of the fire at source and allow the reader to interpret the trend data more easily.

Page 795 lines 11-14 The authors state that - The NEMR reached in 1.4 h is as large as the NEMR observed in smoke from Canada that was 8 days old during NEAQS (F. Flocke private communication). This demonstrates the large variability in both initial emissions and photochemical rates that are associated with BB plumes. In what way do near and far field observations demonstrate that there is wide variability from one region to the other. Many of the plumes seen from biomass burning fires from Canada observed during ICARTT and related studies show that there is a marked lack of photochemical processing of fire plumes after long range transport. Could the NEMR have been generated rapidly but then remained unchanged during transport?

Page 797 lines 17-26: This discussion is somewhat convoluted and also illustrates my concerns over using trend lines and regressions on this type of data. The authors compare the BC/CO at source and at the end to state that the burn conditions were similar but elsewhere have clearly shown that the burn conditions change in the middle period. Yes there is a change between the source and the most aged part of the plume but there are a number of different factors that affect the data in the middle portion making it difficult to conclude whether the changes are continuous or more rapid and mostly in the near field.

Pg 800 lines 3-5: It is interesting to speculate why these differences might arise. The extremely high levels of OH in the near source plume in this study are indicative of very fast processing and have the potential for rapid processing to occur. One wonders

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whether the levels of pollution surrounding the fires studied have any influence as well as the conditions of the burn, for example the background NOx or VOC levels. No OH measurements are available in the BB plumes studied by Capes et al. but in the Sahel there are no other sources of pollution to influence photochemical oxidant levels appreciably. Clearly there are differences and it would be interesting to hear ideas about what these differences might be.

Pg 800 final paragraph: This discussion also appears to support a rather rapid initial processing of the plume.

Pg 802 lines 10-18: The Capes et al analysis used a similar approach to probe the deltaOA/deltaCO relationship to that discussed here. However, as Capes et al point out, the region is impacted by many small and widely distributed fires and there may well be some averaging associated with such an analysis. Despite this the variability reported in this paper would have been seen in the Capes et al analysis as the approaches are similar. It is unclear at the present time why these differences are present – clearly an understanding of what drives such variability and the rapid changes in chemistry over short time is required as most regional and global models require inputs on spatial and time scales that are of the same order if not larger than the changes observed in this ageing study. This represents a significant challenge.

Pg 807: It is not at all clear to me what section 4.3 adds to the already very long paper. I suggest removal of this section.

Pg 808 line 5: There are several papers on BB in the Sahelian tropical region in Africa Haywood et al (2008) and other papers in the same issue.

Pg 809: Capes et al does not appear in the reference list

References Abel, S. J., et al. (2003), Evolution of biomass burning aerosol properties from an agricultural fire in southern Africa, Geophysical Research Letters, 30, (2003)

Capes et al., (2008), Aging of biomass burning aerosols over West Africa: Aircraft

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measurements of chemical composition, microphysical properties, and emission ratios, JGR, 113, D00C15

Haywood J. M. et al. (2008) Overview of the Dust and Biomass-burning Experiment and African Monsoon Multidisciplinary Analysis Special Observing Period-0, JGR, 113, D00C17

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 767, 2009.