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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The Tropical Forest and fire emissions experiment: overview and airborne fire emission factor measurements" *by* R. J. Yokelson et al.

R. J. Yokelson et al.

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Response to Anonymous Referee #3

R3.1. General Comments:

This paper describes an aircraft campaign that took place in Brazil in 2004. The purpose of the field observations was to characterize emissions of biomass burning from tropical forests in Brazil. The authors provide a detailed overview of the field campaign and measurements, quantify emission factors from many important trace gas species, including specific oxygenated volatile organic compounds (OVOCs), estimate

total emissions from tropical forest deforestation based on the results from the study, and present unique observations of a large smoke plume encountered at the end of the study. The authors offer interesting details of the biomass burning that occurs in Brazil and provide a substantial introduction to the paper. A detailed comparison of the measurements from this study is made with other reported observations from the region.

The experimental methods applied during the field campaign and the analytical techniques applied to the data are well done and have been used successfully in past

studies. The results presented provide new emission factors for many compounds of importance to the regional and global modeling communities, and are a great advancement to the existing data. This manuscript is appropriate for Atmospheric Chemistry & Physics, and I recommend that the paper be published after some changes have been made. **In general, I feel that this paper is much too long.** Although extremely interesting, much of the information presented in the manuscript is not entirely necessary. For example, I believe that the Introduction could be condensed. Also, Section 3.3 that describes explicit details about Brazilian biomass burning practices could be summarized or left out entirely.

Au3.1. In revising the paper we have shortened it by three pages while still providing needed clarification. This is described in detail in the earlier part of this response

R3.2. In the Introduction, the authors should point to Figure 1 when mentioning areas of interest (e.g., Page 6906, line 6 and page 6907, line 11). All of the important places mentioned throughout the paper should be included in this figure, for those unfamiliar with Brazil.

Au3.2. This has been done in revised paper.

R3.3. Page 6910: Why was air pumped into the canisters, and why weren't they initially at vacuum? Is anything lost when the air is sent through a pump?

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Au3.3. The canisters were initially at vacuum as is now clarified in revised paper. The pump had a teflon diaphragm (added to text) to minimize losses. Filling the canisters to ~2 atmospheres total pressure provides more sample gas for preconcentration and thus lower detection limits. Also a pressurized can will not be contaminated during storage and shipping if there is a slow leak in the shut-off valve. Unfortunately, our system for pressurizing cans was a bit awkward to operate so we quickly switched to the method where the cans were simply brought up to AFTIR cell pressure.

New text reads: "For TROFFEE, a Teflon valve was added to the AFTIR cell that connected to two options for filling evacuated canisters. For a canister sample of a plume, we used a teflon-diaphragm pump to pressurize the can with gas from the AFTIR cell, which already contained a grab sample of the plume. Pressurizing the cans allows more sensitive and/or a wider variety of analyses and also prevents contamination in the event of a slow leak. Operationally simpler canister samples of background air were obtained by diverting a portion of the flow through the AFTIR cell into the cans."

R3.4. was similar to **R2.4** and both were addressed under **Au2.4** above.

R3.5. Pages 6910-6911: How did the O_3 from the AFTIR compare to the measurements made from the UV absorbance instrument?

Au3.5. O_3 levels were significantly above the AFTIR detection limits (15-20 ppbv) only on 8 September, at which point the UV O_3 instrument had been redeployed at the ZF-14 tower. In general FTIR is a well-established method for measuring O_3. UV O_3 instruments are known to have a positive artifact in very thick fresh smoke due to UV-absorbing compounds that are not removed by the charcoal filter.

R3.6. Page 6913, line 16: Why were only grab samples from the AFTIR made in the background air?

Au3.6. AFTIR collected about 200000 spectra of the background air flowing through the cell in between grab samples, but these spectra have lower signal to noise unless

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they are carefully co-averaged and their analysis is therefore unlikely to occur in the near future.

R3.7. Page 6917, lines 6-7: How do the authors know that the mixed layer air was calculated to come from the northeast? Did they perform trajectory model analysis of this (or did someone else)? If so, please explain or provide the proper reference.

Au3.7. We used the free on-line HYSPLIT model to obtain these results. The revised sentence now reads: "In contrast, HYSPLIT back trajectories show that the mixed layer air came from the . . . " The preceding sentence also cites HYSPLIT

R3.8. Page 6919, lines 2-10: The authors mention their PM10 measurements, but do not include any comparison with other PM observations here (this is done later), as mentioned in lines 9-10.

Au3.8. To avoid lengthening the paper, but point the reader to the previous PM observations in haze layers we added the following text in line 3: "; similar to observations in previous years (Pereira et al., 1996; Reid et al., 1998)."

Pereira, E. B., Setzer, A. W., Gerab, F., Artaxo, P. E., Pereira, M. C., and Monroe, G.: Airborne measurements of aerosols from burning biomass in Brazil related to the TRACE A experiment, J. Geophys. Res., 101, 23983-23992, 1996.

R3.9. Page 6919, lines 23-27: The inclusion of CO from MOPITT here is interesting. However, I am not clear what the authors mean to say here. Is it that the observations do not match the MOPITT retrievals because the vertical profile used in the MOPITT inversion is not correct? I suggest rewriting these sentences to be a bit more clear.

Au3.9. To quote from the MOPITT validation paper cited: "The radiances measured by MOPITT are not sufficient to uniquely determine the atmospheric vertical distributions, therefore the inversions must be constrained with a priori information about the concentrations and variability in the atmosphere. A single a priori profile and covariance matrix are used for all locations and seasons in the MOPITT retrievals and were com-

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piled from numerous aircraft observations, representing our best estimate of a global average mixing ratio profile and the correlations between altitudes (see Deeter et al. [2003] for details)." The global average profile shown in their Figure 1 appears to be essentially a smoothly varying profile with no strong enhancement near the ground and no sharp drop-off in mixing ratio at the top of a typical boundary layer height. Further, the sensitivity of MOPITT is greater to CO present at higher altitudes. The uncertainty in profile shape and the lower sensitivity at lower altitude where most of the CO is may not be severe problems: we are not remote-sensing experts. However, the comparison shown in their figure 5 indicates that reproducing CO profiles measured during SAFARI 2000 may be more difficult for MOPITT than at sites less impacted by biomass burning. Biomass burning is a major global source of CO, so we speculate that, if possible, a different a-priori assumption about the shape of the vertical profiles in biomass burning areas might improve the retrievals. Since our paper is long and our response will be published, we have lightly revised our original text to read as follows: "The African and the Brazilian CO vertical profiles are not shaped like the a-priori CO vertical profile used for MOPITT CO retrievals (Emmons et al., 2004). We speculate that consideration of the actual profile shapes might enhance CO retrievals from space-based instruments."

R3.10. Section 3.3: Again, I think that this section is too long and contains too much detail that is not directly relevant to the rest of the paper.

Au3.10. This section was shortened significantly and moved in response to multiple comments.

R3.11. Also, I think it is worth mentioning here (when it is first brought up) that the surface measurements made at the ground level could have observed such high CO concentrations since it was measuring the emissions and plumes from RSC. (I don't think this is mentioned until later?)

Au3.11. As was stated on line 15, the ground-based measurements were not in or

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near smoke plumes.

R3.12. Page 6922: the authors explain a lot of historical changes in biomass burning in the Amazon. Could this be another reason for the differences between the observations from the late 80's and those given here?

Au3.12. Possibly, but the 30 ppm increase in ambient CO_2 that we observe comparing 2004 to 1987 is 1.76 ppm/year, which is consistent with the conventional wisdom that global CO_2 is increasing about 2 ppm year.

R3.13. Page 6924-6925: What is the mean of the annual average of acreage burned? (They only present the two highest year numbers).

Au3.13. Fixed as described under Au2.19.

R3.14. Page 6926: this is just a note: AM includes the Ward et al. and Ferrek et al. papers in its compilation of emission factors.

Au3.14. That is correct and so the AM value is not completely independent of the others. However, since it is probably the most widely used value we also compare to it directly.

R3.15. Page 6930-6931: Although interesting here, there is a lot of text about cooking and other fires that may not be necessary for this paper.

Au3.15. We have shortened the paper elsewhere and retain this section since reviewer 2 actually wanted a few more things added.

R3.16. Section 3.5.3: This is a very important section and I think could be made a bit more clear. First, the fact that much of the emissions comes from RSC, but do NOT get lofted is very important, especially to modelers. It is not clear where the authors got ˜ 240 Tg of biomass burned. Is this from the Brazilian database? Is this a 10-year average?

Is all of this burning assumed to be from deforestation? It is also not straightforward

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to arrive at the numbers 2148 Tg CO and 24 Tg PM10 from the text. This needs to be made much clear. (I see an average emission factor of 101.4 g/kg CO and a total tropical forest biomass burned 1330 Tg yr-1. Did I miss something?)

Au3.16. In reverse order:

(1) Section 3.5.3 was estimating the CO_2 emissions, not the CO emissions. The 1330 Tg of biomass burned in tropical deforestation fires globally per year is 1.33×10^{12} Kg of biomass burned. Our CO_2 EF is 1615 g/kg. The product of these two numbers is 2148 Tg. The global CO would be $101 \times 1.33 \times 10^{12}$ or 134 Tg.

(2) We changed our fuel loading mass unit from tons (t) to Mega-grams (Mg). The new section on characteristics of Brazilian fires (section 2.3.2) shows that 120 Mg/ha is an average fuel consumption for Brazilian deforestation fires. That section and the new section 3.3 give the average annual deforestation as 20000 km2 - or 2-million ha. In the revised text of the "bottom-up estimates" section, we clarify this and also include a separate estimate for the additional biomass burned in pasture fires and an estimate for the biomass burned by RSC in pasture fires. These are important improvements that we planned to make voluntarily if the reviewers had not commented. The new text reads:

" 3.3.3. Regional-global bottom-up emissions estimates. About 2 million ha of tropical rain forest are burned in an average year in Brazil and $\sim$120 Mg/ha of fuel is consumed in these fires (Sect. 2.3.2). Thus, $\sim$2.4 $\times$ 1011 kg of biomass are burned annually in primary deforestation fires. The last value can be multiplied by any EF in Table 2 for a bottom-up estimate of annual emissions from Brazilian tropical deforestation fires. For instance, 388 Tg and 4 Tg are crude estimates of the average annual CO2 and PM10 emissions from Brazilian deforestation fires. The amount of biomass burned and the total emissions for each species approximately double if pasture fires are included, although 20-50% even larger emissions than predicted by this type of estimate are warranted for several VOC to account for RSC in Brazilian pasture fires (Christian et

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al., 2007b)."

R3.17. Section 3.6: This section is very interesting, especially since the measurements made during this plume intercept were so much higher than the other observations. I think that this entire section could be condensed some. Also, it is a little unclear in general. The authors talk about a plume that covers a huge expanse. It appears that the last flight of the campaign caught the beginning of this event. However, despite its size and duration, the authors talk about identifying the source of the "white ocean" to a few particular fire points detected by satellite? It sounds like the observations included a combination of aged smoke and relatively new plumes. Therefore, I am not entirely convinced that the smoke that was measured was relatively young (0-4 hours as stated on page 6935, line 6).

Au3.17. We have condensed this section. We no longer say we were trying to "pinpoint" the source of the smoke. The word "pinpoint" was replaced by the word "identify." The 3 groups of hotspots we are proposing as the source of our mega-plume sampled numbered in the 100s-1000s each. The original text signaled this with phrases such as "very substantial hotspot activity" (line 25, page 6934) and "numerous hotspots" (line 5 page 6935). The original text also suggested that the hotspots were an under-estimate. For the revised text we actually counted the hotspots and give our estimate rather than using qualitative language. We have rephrased the revised text to indicate that we think there were at least three major source areas for the portion of the mega-plume we sampled on 8 September: (1) smoke less than one-day old from fires near 50W that burned during the afternoon of 7 September and (2) smoke less than four hours old from fires that burned during the morning of 8 September near 56 and 55W. Without a dedicated study of all available hotspot and burned area products it doesn't seem worthwhile to estimate the fraction contributed by each source area. Further, since we state that many of the fires near 56W were absent from the NOAA-12 product, the obvious implication is that many more fires, perhaps in other areas, could have contributed as well.

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R3.18. Page 6935, line 27: The authors state that the plume went a "Little further north than usual." What's usual?

Au3.18. We now refer the reader to Figure 7c where the usual exit point of the emissions (S of São Paulo) is shown. New text in context: "During most of September, the mega-plume exited South America well south of São Paulo (e.g. Fig. 7c). However, the TOMS aerosol index (AI) images for 16 and 17 September show the mega-plume (with AI $\sim$3.5) exiting South America over São Paulo (Figs. 8a and b)."

R3.19. Page 6936, line 15: I can't remember where in the paper the speculation that large fires may be becoming more common in the Amazon. A reminder would be helpful.

Au3.19. This is in the new section 2.3.2. A reminder has been included in parentheses.

R3.20. Page 6937: The conclusion includes a lot of really interesting and worthwhile topics for further pursuit. Although I think that this is a valuable list, I feel as though a few of these topics weren't addressed in the paper (e.g., 6).

Au3.20. The conclusions are structured as follows: summary of what we did on biomass burning in TROFFE, summary of what we have done globally on biomass burning in recent years, partial list of future research needs. We did not address any of the future research needs during TROFFE. We have revised the sentence before the list to read "More biomass burning research is needed including: (1) . . . "

R3.21. Page 6909, lines 23-25 could be omitted.

Au3.21. This text may have been useful, but it was eliminated to shorten paper.

R3.22. Page 6914, line 5: another ")" is needed after delta CO).

Au3.22. Done.

R3.23. Page 6914, line 15: The parenthesis around the sentence starting with "EFs are combined" can be removed. The sentence can also be moved to line 11.

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Au3.33. Done.

R3.24. Page 6918, line 8: Could be changed to “Most of the lower CO values were observed above the mixed layer.”

Au3.24. Done.

R3.25. Page 6918, line 19: Remove “above.”

Au3.25. We changed “the above” to “our.”

R3.26. Page 6920, lines 11-14: This sentence should be reworded. A suggestion is “A large uncertainty in the estimated area burned is due to uncertainties in remote sensing applications. For example, it is unclear if small fires or understory fires can be quantified from space, and many fires can be missed from space due to cloud cover, which is common over tropical forested regions.”

Au3.26. We have used the reviewers suggestion verbatim.

R3.27. Page 6922, line 6: What is Caatinga? This could be put on the map in Figure 1.

Au3.27. 3-10S and 36-42W. This is hard to indicate on our map since it is not a vegetation map. The text now includes “eastern Brazil” in parentheses.

R3.28. Page 6922, line 23: Where is Mato Grosso? This could also be put on the map in

Figure 1.

Au3.28. The states we sampled in are now labeled in a revised Figure 1.

R3.29. Page 6929, line 12: This is not a complete sentence.

Au3.29. It is now examples enclosed in parentheses.

R3.30. Page 6930, line 14: The text in the parentheses could be removed.

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Au3.30. Done.

R3.31. Page 6930, line 17: Add a ":" after questions

Au3.31. Done.

R3.32. Page 6933, line 4-5: This sentence doesn't make much sense.

Au3.32. When fires are less common and separated by greater distances, the visible smoke plumes are mixing with the dilute regional haze during aging. When fires are more common and closer together, the visible plumes tend to mix with other visible plumes of concentrated fresh smoke during aging. The two scenarios could result in different chemical products. If so, it could be important to recognize in models. This is clarified in the revised paper. New text reads: "Whereas smoke plumes usually age in relative isolation from each other while diluting with regional haze, in the mega-plume scenario, direct mixing of fresh smoke plumes likely dominates. The two chemical processing environments may lead to different outcomes and the latter scenario may be the relevant processing environment for a large part of the total regional emissions."

R3.33. Page 6933, line 16: separate the sentence: "that it was sampled. The EEF are"

Au3.33. Done.

R3.34. Page 6933, lines 27-29: This sentence doesn't make much sense, and could be reworded.

Au3.34. The sentence topic is similar to the topic of R3.32. We have simplified the sentence to read: "The haze was less-concentrated and of even more ambiguous age. Nevertheless, it probably had some chemical processes in common with the mega-plume." We think this will make sense to the reader in light of previous clarifications in response to R3.32.

Additional changes:

1) We originally included a short paragraph about phenol that mentioned possible sam-

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pling losses for the PTR-MS and we flagged the Table 2 values as lower limits. Subsequent work showed good agreement for phenol between open-path FTIR and PTR-MS. Thus we eliminated the flag (footnote) from Table 2 and also the short paragraph in the comparison to other work.

2) As described above, a footnote was added to Table 2 to define "nm" as "not measured." (In most cases this was because a measurement was not attempted.) We also replaced the formulas for acetonitrile and acetaldehyde with their names as was already the case for the other PTR-MS compounds. Finally, the table entry "C6 hydrocarbons" was corrected to "C6 carbonyls."

3) Slightly revised section 3.3.2 and conclusions to reflect the value of acetonitrile for source apportionment.

E.G. New: Still, these results suggest that (with proper attention to the type of fire), PTR-MS acetonitrile measurements could contribute to source apportionment or, with inverse modeling, estimates of the amount of biomass burned.

4) Updated references

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