

***Interactive comment on* “The tropical forest and fire emissions experiment: laboratory fire measurements and synthesis of campaign data”  
by R. J. Yokelson et al.**

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Revisions to address referee comments on: “The tropical forest and fire emissions experiment: laboratory fire measurements and synthesis of campaign data”

by R. J. Yokelson et al. acpd-2007-0658

**1. Comments from R. Holzinger (Referee)**

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**Referee 1.1.** This is a well written (with reservations referring to section 3.3, see below) and important paper well worth being published in ACP. However, many parts of the

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paper are a bit wordy, so my general suggestion to the authors is to shorten where possible and focus to the major messages. In my opinion the strongest point is that the authors took a lot of effort to reconcile emission factors of individual species measured under different conditions (lab, ground based, and airborne). Their analysis is insightful and yields improved recommended overall emission factors that - despite all problems and uncertainties - can be used for more realistic modeling of global biomass burning emissions.

Their study allows new extrapolations to estimate a global NMOC source of ~500Tg/yr from biomass burning. This is a factor 5 higher than the widely accepted value from Andreae and Merlet, 2001! This major conclusion is unfortunately plugged into a section (3.3) containing unnecessary discussions on biogenic versus pyrogenic emissions and SOA formation. I strongly recommend to focus only on the biomass burning extrapolation in section 3.3 and delete all other discussion. This will also require a few adaption in other sections but overall the paper will benefit a lot.

**Response 1.1.** This is likely the final paper in a series. The function of this paper is to present the lab-fire emission factors and synthesize the lab-fire emission factors with a broad range of results from 4 other previously-published, detailed companion papers. All five papers arose from a single project. The foci of the other papers were: (1) detailed campaign overview and airborne measurements of fire emissions, (2) airborne and ground-based measurements of biogenic emissions, (3) ground-based measurements of fires, and (4) a detailed intercomparison of the instruments used in both lab and field. This synthesis portion of the paper is not intended to be a comprehensive review of all the literature, but it is intended, as the probable, final paper in the series, to show how the different aspects of our campaign fit together and highlight the useful results. All the authors feel that the discussion of up-scaling emissions, comparing pyrogenic and biogenic emissions, and secondary processes are the most useful parts of the paper to the broad scientific audience. In fact, the scope of the discussion matches

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well with our original motivation for the project as described in the previously-published detailed overview. The other Referee may also feel that the scope of the discussion is appropriate - though he had comments on our approach that we address further on in this document. Thus, we feel that the proper way to proceed is to modify the introduction so it does a better job of describing the scope of the overall campaign and also the function of this paper within the series of papers. In addition, we now clarify that the function of section 3.3 is to impart the new knowledge gained and lessons learned from the overall TROFFEE campaign. We did delete two paragraphs from section 3.3 that covered only selected aspects of total VOCs and the carbon cycle. They were probably less useful than the more comprehensive discussion on NMOC. We have also split section 3.3 into “regional” and “global” subsections, which should help the key conclusions stand out better. We added two sentences about O<sub>3</sub> formation to the regional section as it was a major motivation for this work that we neglected to include in this section originally. We made other light revisions to section 3.3 as needed.

#### Revisions:

To better describe the scope of TROFFEE in the introduction, we have inserted the following text at line 15 of page 4223:

“Four previously published TROFFEE papers focused on: (1) a detailed campaign overview and the airborne measurements of the emissions from fires (Yokelson et al., 2007a), (2) ground-based measurements of the emissions from smoldering logs (Christian et al., 2007), (3) airborne and ground-based measurements of biogenic emissions (Karl et al., 2007a), and (4) intercomparison of the instrumentation used in both the lab and field (Karl et al., 2007b). This paper completes the series by presenting the lab-fire emission factors and demonstrating how the different elements of the TROFFEE campaign can be synthesized to improve our understanding of the tropical troposphere.”

The revised section 3.3 is shown at the end of the response to Referee 3 since the

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revisions address comments by both Referees.

Because we deleted two paragraphs of section 3.3 a change was needed in the conclusions:

Page 4248, line 16

Old sentence:

“More total NMOC are emitted by plants and they dominate the OH reactivity of the region, but fires contribute a comparable amount of total VOC.”

New sentence:

“Fires are the main regional source of CO, but more total NMOC are emitted by plants and they dominate the OH reactivity of the region (Karl et al., 2007a).”

**Referee 1.2.** Part of the introduction reads as if the authors seek justification for performing lab experiments. There is no doubt that lab- and field studies must complement one another. I recommend re-wording this part by objectively listing the respective strengths and weaknesses.

**Response 1.2.** We originally wrote this section thinking that some readers would assume that measurements on lab fires were of no value since field fires have the overwhelming advantage of being “real” and the actual phenomena of global importance. We have revised the text to stress the ultimate advantage of field fires better in the introduction. We have also revised the text to no longer say that lab fires offer “advantages” so there should be no unintended connotation of lab fires being “better.” (This may have already been obvious with continued reading, since throughout the remainder of the paper we questioned the representativeness of the lab fire emissions and their weight in the final recommendations is very limited.) Finally, we decided to split our motivation for carrying out the lab fires into general and specific reasons. To accomplish these three objectives we reorganized the second two paragraphs of the introduction, which follow the change described above and now read as follows:

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“Field measurements can probe the actual fires that are of global significance and they have obvious priority in developing recommended emission factors, but measurements of lab fires can also be useful. The TROFFEE lab experiment was conducted for a number of reasons - both general and specific. In general, it is often possible to quantify more species from lab fires because smoke concentrations tend to be higher and it is easier to deploy more extensive instrumentation. Also, in the lab, one can capture and probe all the smoke from a whole fire, while the vast majority of the smoke from field fires must go unsampled. In the field the possibility exists for over estimating the relative importance of strongly lofted flaming emissions from airborne platforms; or under estimating their importance from ground based platforms. Measuring the elemental composition of the fuel that actually burned is easier for laboratory fires, which then facilitates mass-balance studies that account for the fate of various elements in the fuel. A very serious disadvantage of laboratory fire simulations is the possibility that the lab fire emissions are different from fire emissions produced in the field. This is especially critical for tropical forest fuels as it is impractical to burn a diverse suite of large diameter tropical logs in the lab.

Reasons for carrying out our laboratory component that were specific to TROFFEE included: (1) Determine the proton-transfer reaction mass spectrometry (PTR-MS) sampling protocol for the field campaign (by identifying the significant mass/charge ( $m/z$ ) ratios observed by PTR-MS in smoke). (2) Employ techniques in addition to those used in the field including particle collection on filters, ash analyses, open-path Fourier transform infrared spectroscopy (FTIR), and gas chromatography coupled to PTR-MS (GC/PTR-MS). (3) Intercompare PTR-MS with open-path FTIR and GC/PTR-MS. The intercomparison showed good agreement in most cases, but also revealed important biomass burning emissions that are difficult to measure by FTIR (due to interference by water lines) or PTR-MS (due to low proton affinity or sampling losses). (4) Use GC/PTR-MS and FTIR to measure the fractional contribution for fire-emitted species that appear at the same  $m/z$  in the PTR-MS (Karl et al., 2007a). (5) Measure the emissions from burning sugar cane, which are important, but not accessible in our field

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study.”

The final paragraph of the introduction was revised to better reflect the paper contents:

“The laboratory experiment involved measuring the emissions from 32 fires that burned tropical forest fuels and a few other fuels (e.g. sugar cane, pine needles, and savanna grass). In this paper we present and discuss: (1) a partial accounting of the fate of the nitrogen, chlorine, and potassium in the biomass fuel in our lab fires, (2) a synthesis of the lab, ground, and airborne EF to derive recommended EF for primary tropical deforestation fires and tropical pasture maintenance fires, (3) new estimates of the vegetative and fire emissions of NMOC at the Amazon-basin and global scale with comments on the significance of the new information, and (4) excess emission ratios and emission factors for sugar cane fires.”

Note: because the introduction has been heavily modified here and in response to the other Referee, the revised introduction is presented in its entirety at the end of the response to Referee 3.

**Referee 1.3.** page 4224, line 6: briefly explain following statement: "Also, in the field the possibility exists for over-estimating flaming emissions from airborne platforms or underestimating them from ground-based platforms."

**Response 1.3.** Field fires can have 1-100 meter flame lengths and produce convection columns 2-10 kilometers high, but they can also produce large amounts of unlofted emissions. Thus, it's very difficult (if not impossible) to adequately sample the flaming from the ground or the smoldering from the air. This sentence has been moved in the revised introduction (see above) and has been modified to read:

“In the field the possibility exists for over estimating the relative importance of strongly lofted flaming emissions from airborne platforms; or under estimating their importance from ground based platforms.”

**Referee 1.4.** Table 2: the difference between units "g/kg dry weight" and "g/kg dry fuel"

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needs explanation.

**Response 1.4.** We have revised what is shown in Table 2 to make it easier to understand. Originally, we showed the g/kg dry weight of plant tissue and ash. There is no difference between g/kg dry weight plant tissue and g/kg dry fuel. However, for the ash, the g/kg dry weight of ash has to be multiplied by the fractional conversion of fuel to ash (which was not shown) to compute the ash yield in g/kg dry fuel. We had shown the raw ash analysis so the reader could see that the chlorine concentration in ash is about 5 times higher than in the original biomass fuel. We decided it would be simpler for the reader if we just show the grams of chlorine in the ash per kilogram of dry fuel burned. The revised footnote should make this clear. "Units: plant tissue, ash, and PM2.5 g/kg dry fuel." Then we revised Figure 1b) so it shows that conversion of fuel chlorine to ash chlorine using the concentration units in Table 2 (rather than on a mass basis as was done originally). The different weighting in the linear regression then caused a non-significant change in slope from 0.48 to 0.51. This new value was updated in the text. Also, the fractional conversion of fuel to ash we used is that measured for each lab fire as described on line 23 of page 4225 and not the conversion percent from the CHN analyzer shown in Table 1. Thus we revised the header and footnote of Table 1 to make it clear that that % ash shown there is the value applicable only to the CHN analyzer. We also lightly revised part of section 2.2 to make this more clear and the new text reads as follows:

"Table 1 also shows the dry weight percentage of carbon, hydrogen, and nitrogen in the bulk plant tissue; the percent ash residual in the CHN analyzer; the percentage of carbon in the organic (burnable) plant material (ash-free %C); and the fuel moisture at the time of the fire ( $100 \times (\text{wet-dry})/\text{dry}$ ). We determined the production of ash and partially burned fuel for each fire by manually weighing these residuals."

Finally, we have revised the paper so that "g/kg dry fuel" is the terminology used consistently throughout the paper and tables wherever it is appropriate.

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**Referee 1.5.** Section "2.4 PTR-MS" can be shortened. Just give details that are different from standard operation and specific to this experiment. The principal functionality of PTRMS does not need to be reproduced here.

**Response 1.5.** We eliminated lines 5-13 of page 4227 including equation 1.

**Referee 1.6.** Table 3: I suppose MCE, CO<sub>2</sub>, CO are no FTIR species and PM2.5 is not a PTRMS species. Units are indicated to be g/kg - is this kg dry weight? For all species listed?

**Response 1.6.** The Referee is correct. We used the real-time instruments to measure CO<sub>2</sub>, CO, and MCE and the PTR-MS did not measure PM2.5. Table 3 was revised to reflect this. Also, we revised the table header to define the emission factor units as g/kg dry fuel to be consistent with the rest of the paper and our responses above.

**Referee 1.7.** Section 3.2: On the discussion of HCN and acetonitrile it would be interesting to show a plot (of) EF versus nitrogen fuel content. At least for acetonitrile I expect a good correlation. Biomass burning estimations using CH<sub>3</sub>CN as tracer could probably be improved when the N-content of biomass is considered.

**Response 1.7.** It is a good idea to explore this issue. We have made two changes in response to the Referees suggestion.

(1) The ease of measuring the fuel chemistry and performing a mass-balance was one of our stated general reasons to conduct laboratory fire work. We have renamed section 3.1 to "The fate of nitrogen, chlorine, and potassium in the fuel." We now lead off that section with a very brief discussion of the N balance for these fires and a reference to a more detailed discussion in a previous lab-fire paper.

We added at line 18 of page 4229:

"The percentage of the fuel nitrogen that ended up in each measured, emitted nitrogen-containing trace gas is listed next in order. Ammonia ( $16 \pm 8.9$ ), NO<sub>x</sub> ( $5.4 \pm 2.8$ ), HCN ( $1.3 \pm 0.6$ ), CH<sub>3</sub>CN ( $1.1 \pm 0.5$ ), propanenitrile ( $0.9 \pm 0.5$ ), acrylonitrile ( $0.5 \pm$

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0.3), and pyrrole ( $0.07 \pm 0.04$ ). The sum of  $\text{NH}_3 + \text{NO}_x$  is within the range of previous observations and these data are potentially consistent with an accounting for all the fuel nitrogen (when  $\text{N}_2$  emissions and ash N are also considered) as discussed in detail by Goode et al., (1999).”

(2) In section 3.2, page 4234, line 9: where we raised the topic of variability in EFHCN and  $\text{EFCH}_3\text{CN}$ , we now show the affect on the variability of normalizing by fuel N. We do not show a plot because the magnitude of the normalization effect did not warrant this. However, there was a modest, but significant, reduction in uncertainty as the new text now clarifies:

“For the laboratory fires we have total fuel N data and can examine the dependence of the EF for N-compounds on total fuel N. We start by noting that, for the lab fires, the standard deviation in the EF divided by the mean EF is  $0.69 \pm 0.03$  for HCN,  $\text{CH}_3\text{CN}$ , and  $\text{NH}_3$ . If we divide each EF by the total fuel N, this ratio decreases to  $0.51 \pm 0.04$ . Thus there is a significant almost 30% decrease in variability when normalizing these EF by total fuel N. The fact that considerable variability remains may indicate that the emission of these N compounds may depend less on total fuel N than they do on the fuel content of specific N-containing molecules that naturally occur in quite variable amounts.”

**Referee 1.8.** Figure 5: no unit is given for the y-axis

**Response 1.8.** Thank you. We added y-axis units to Figures 2-5.

**Referee 1.9.** The last paragraph of section 3.2 (“We next compare...”) is redundant in my opinion and can be deleted.

**Response 1.9.** This paragraph highlighted the significant differences in how well the emissions from the major fire types are characterized. This strongly impacts the accuracy of emissions estimates and models at all scales. However, we also made this point (less explicitly) starting at line 24 of page 4244 in the global pyrogenic NMOC

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estimate. Thus, we have eliminated the paragraph as recommended by the Referee.

**Referee 1.10.** Page 4244 line12 : what is EFNMOOC?

**Response 1.10.** The sentence referred to "... the sum of our EFNMOOC ...". It has been revised to refer to "... the sum of the EF for all our measured NMOC ...".

**Minor voluntary addition by authors:**

We added a range in parentheses after our estimate of global biogenic and pyrogenic NMOC in the abstract and conclusions.

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