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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Field determination of biomass burning emission ratios and factors via open-path FTIR spectroscopy and fire radiative power assessment: headfire, backfire and residual smouldering combustion in African savannahs" *by* M. J. Wooster et al.

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Referee's Report Bob Yokelson

I agree with Dave Griffith's comments and make some additional comments. In general it is a nice piece of work that should be published. However it needs to be carefully proofread and the authors need to focus more on what they actually did, organize their topics, and adopt one conclusion per topic. If I might suggest a good outline for a new

introduction:

1. Biomass burning is important and hard to sample 2. Griffith showed advantages of OP-FTIR 3. Bertschi et al 2003 first showed significance of RSC for some ecosystems 4. Christian et al 2007 then did point source measurements of RSC 5. We now present long path spatially integrated measurements of RSC and other weakly lofted emissions. The point measurements are beneficial in being fuel specific, which may allow scaling. The path integrated measurements are beneficial in that they may be more site-integrated. 6. We disprove the proposal that MCE could be measured from space with current technology. 7. We compare the fuel consumption obtained from FRP to the traditional measurements (if that is what they are) shown in Table 1. Or defer to another paper?

Additional comments:

Format last two page numbers, line number

P31, L1: either "gases" should be "gas" or "releases" should be "released" (Lots of proofreading needed)

P31, L13: I would phrase this as "preferentially from . . ... combustion processes" since the fire emissions are usually not separated into "phases" (Yokelson et al., 1996)

P31, L15, 16, etc throughout: It is physically more accurate to change "stages" or "phases" to "processes"

P31, L24: "vast majority" should be a range of values or an average and standard deviation if possible.

P32, L2: re, "we see no evidence to support suggestions of a major overestimation in the emission factor of ammonia" who/what are they talking about?? The authors or someone else?

P32, L5-6: This could be highly dependent on the field of view compared to the size of the flame fronts so it would be better to change "remotely" into something more specific such as "airborne" or "space-based" if they can support the latter with a rigorous argument.

P32, L7: "EO" needs to be defined

P32, L6-8: The authors say here their data "supports" the idea of classifying fires into phases from space, but in fact their data seems to prove that was impossible even with the dramatically higher resolution available from a helicopter. Further, the chance of fires being classified by "phases" especially when the "processes" are normally thoroughly mixed; and when viewed from space seems extremely "remote." How did the space-based detection using SEVIRI work on the fires the authors measured in this study? My experience is that only about ten percent of the fires we can locate on the landscape show up as MODIS hotspots and that includes fires well over 100 ha.

P32, L9-10 : The authors demonstrated that they could measure EF and ER with an open-path FTIR system AND a helicopter measuring FRE not the OP-FTIR alone as this sentence implies. Also the utility is limited to very short flame lengths and only smoldering combustion if there are no auxiliary fuel consumption measurements. Here is a good place to introduce some of the realistic caveats that the other referee #1 also mentions regarding the applications of this technique.

P32, L25: a more recent compilation of EFs is now available also: Akagi, S. K., Yokelson, R. J., Wiedinmyer, C., Alvarado, M. J., Reid, J. S., Karl, T., Crounse, J. D., and Wennberg, P. O.: Emission factors for open and domestic biomass burning for use in atmospheric models, Atmos. Chem. Phys. Discuss., 10, 27523-27602, 2010.

P33, L1: GFED3 is out now: van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707-11735, doi:10.5194/acp-10-11707-2010, 2010.

P33, L1-6: It is really important not to overstate in vague terms the capabilities of FRP based fuel consumption estimates or the value of daily snapshots of FRP in terms of so-called "operational forecasting." I reviewed another paper where the author (not an FRP person) stated that FRP-based fuel consumption had "largely replaced" the old approach of using actual on-the-ground, measured, ecosystem-specific fuel consumption values. I am not sure where that author got that idea, but nearly all global emissions models still use ecosystem specific measurements of fuel consumption (see Wiedinmyer et al., 2010 and references there-in). So it's possible that portraying a long-term goal as a current reality has a negative side effects on other earth scientists.

P33, L8-9: The authors are correct in saying that there is a need for improved EF. As Bertschi et al., 2003 showed, a major uncertainty in EF is the contribution of residual smoldering combustion, which could increase e.g. the fire average EFCH4 by more than a factor two if RSC of logs accounted for even 10% of the fuel consumption. The authors system is ideal for measurements of RSC and can make a major contribution in that way. But by trying to portray the system as "ideal" for measuring the total fire emissions including flaming, they are getting somewhat off course.

P33, L11-12: "maturity" and "accuracy" are not the same thing, which should be recognized here. E.G. using a combination of space-based products Kopacz et al determined that many current estimates that agree well with each other substantially underestimate (i.e. on the order of 100% error) biomass burning. The new MOPITT5 retrievals may also change our view of how much biomass burns. It's important to cite a variety of conclusion to discourage unwarranted confidence in numbers that are controversial to say the least.

P33, L22: virtually all studies that show EF as a function of MCE show that nearly all species vary by a factor of at least 500% sometimes a factor 20. Also "emissions factors" has too many "s"

P33, L30: As the authors undoubtedly observed during their field measurements, once a flaming front begins to move across the site, the emissions from flaming and smoldering are mixed. Thus, I am not sure why they keep invoking the idea of flaming and smoldering phases as separate entities??

P34, L2-6: Exactly! Yes! Good stuff! This is important. I would make this the main point of abstract and intro. What is also important is better estimates of the fuel consumption by RSC and the authors provided a new measurement of that for an important ecosystem that confirms previous speculation by Bertschi et al (2003) and that should also be highlighted.

P34, L9: parentheses around "1993"

P34, L16-18: The sentence should be eliminated as Griffith et al (1991) already used OP-FTIR to measure fire emissions and further, the authors work was undertaken in 2007 well before the 2011 paper of Fenandez Gomez appeared and not because of their suggestion. I would eliminate all mention of this paper throughout as it is basically just bench-scale TG-FTIR of biomass combustion, which has been done since the 1980s. See e.g.

Lephardt, J. O., and R. A. Fenner, Characterization of pyrolysis and combustion of complex systems using Fourier transform infrared spectroscopy, Appl Spectrosc., 34, 174 - 185, 1980.

DeGroot, W. F., W. Pan, M. D. Rahman, and G. N. Richards, First chemical events in pyrolysis of wood, J. Anal. Appl. Pyrolysis, 13, 221-231, 1988.

P34, L23: "demonstrate" should be "confirm" as this is not a first and Griffith et al., 1991 should be referenced here as the first to do this in smoke.

P35, L4-5: "southern" - and to get 25% of global BB you need to include the distinctly different more humid wooded savannas.

P35, L13-16: The carbon-mass balance method should also be used in lab studies because the fuel mass changes in response to evaporation of fuel moisture. Phases

are not normally observed in lab studies and lab studies show that trying to separate lab fires into phases or stages introduces errors (Table 3 in Yokelson et al., 1996; Burling et al., 2010).

Burling, I. R., Yokelson, R. J., Griffith, D.W.T., Johnson, T. J., Veres, P., Roberts, J.M., Warneke, C., Urbanski, S.P., Reardon, J., Weise, D.R., Hao, W.M., and de Gouw, J.: Laboratory measurements of trace gas emissions from biomass burning of fuel types from the southeastern and southwestern United States, Atmos. Chem. Phys., 10, 11115-11130, doi:10.5194/acp-10-11115-2010, 2010.

P35, L18: Lab-field differences in EF are discussed in the most detail in Yokelson et al. (2008). Yokelson, R.J., T.J. Christian, T.G. Karl, and A. Guenther, The tropical forest and fire emissions experiment: Laboratory fire measurements and synthesis of campaign data, Atmos. Chem. Phys., 8, 3509-3527, 2008.

P35, L21: at least second case of "maybe" should be "may be"

P35, L23-24: Omit "logistics and costs hindering the deployment" doesn't make any sense when the authors also used a helicopter.

P36, L23-4: "Fernandez-Gomez et al. (2011) suggest the time is right for a re-appraisal of the approach."I don't think the authors cited have any experience at all with measuring real fires so not sure if they would be the authorities to cite here. This whole section could be replaced along in accordance with this outline. Griffith et al tried it first and it showed sensitivity simultaneously to a lot of difficult to measure important gases. But it was hard to do weighting for total emissions. Since then Bertschi et al showed even the RSC important and we demonstrate here a novel way to weight the spectra dominated by individual processes into an overall average.

P36, L27: "to" before "similar" proofread please, I have 5-6 papers of my own I could be working on instead.

P37, L14: the authors say here there "substantial" RSC, but it appears to be only 1% of fuel consumption on all four fires in Table 1, which apparently confirms the hypothesis of Bertschi et al., (2003) regarding grass fires. Much more RSC may occur in wooded savannas or in Brazil pasture fires as note earlier.

P37, L21-22: give wavelength range or define SWIR and TIR

P38, L11: "stacked" usually refers to a display option whereas what the authors mean is usually called "co-added" or simply "averaged together"

P39, L5-6: This is different from the Bertschi et al definition, which includes unlofted emissions that could be produced while flaming continues too far away to entrain the said RSC. Again a difference between process and stage. That may contribute a bit to the lower fuel consumption by RSC than Bertschi et al obtained.

P39, L24: maybe should be "may be" throughout??? I found at least 4 instances of this error.

P39, L25, One caveat: is not clear to me how to handle differing degrees of non-linearity in different portions of the path without using some sort of layered approach. The authors should mention if their model allows for different temperatures and a varying concentration over the path.

P40, L4-8: Interesting, we found the opposite effect. Of the compounds they measured the only one we find affected by the updates to HITRAN is HCHO, where the new parameters imply a decrease of 30% percent in mixing ratios rather than an increase. We have corrected the EF for changes in HITRAN and new PNNL reference spectra for all the older FTIR-BB studies that we know of in the review by Akagi et al., (2010).

P40, L9: The MALT code can deal with different temperatures along the optical path. It sounds like the Burton code can as well, but that should be specified here since flaming combustion could certainly cause great temperature variation along the path. Minor point: if a single path length is used the emission factors are independent of the path and depend only on the ratios between compounds and to CO2

P40, L25-26: I prefer by... processes" rather than "during .... phases" as explained above

P41, L26-P42, L1: we don't usually see Co2 that high but within 5 ppm of LiCor is OK. We find it more challenging to match a large range of CO2s, but that is hard to test here.

P42, L22: The inability to get a background outside the plume is more of an issue in a ground-based experiment than an airborne experiment. We have found that plotting excess and absolute amounts is generally comparable, but the excess approach is less susceptible to the following. Variation in the ER during a fire can cause the slope of the absolute plot to extrapolate to an unrealistic background and then distort the slope, but the authors do not seem to have encountered this on the plots shown, which look great. Also in many cases, the plume mixes with a background that is actually aged smoke and so not subtracting the background can return an ER reflecting both aged and fresh smoke.

P43, L10-12: if you use column amounts it is true the subtracting the pre-fire amount is incorrect. If you use path average mixing ratios then subtracting the prefire mixing ratio is OK. Also if the in-plume-path is changing spectra to spectra its only meaningful to compare gases made in the same spectra and a time series using column amounts is more accurate. The non-plume portion of the path is likely getting more and more polluted by smoke generated earlier as the experiment continues.

P44, L19-21: If the fuel consumption implied by FRE is independent of F/S, then how do the authors get F/S? If they have to do phases, Yokelson et al., 1996 showed that phases don't work. In general there is insufficient detail on the fuel measurements.

P44, L25: "plot" should be "path"

P45, L16-18: to get to 99% you have to at least include CH4, which they did and should be noted.

P45, L18-19: The statement “The majority of the remaining carbon is emitted as aerosols” is wrong. Most the rest is NMOC. A typical EFNMOC is 30 g/kg including all the species that can be measured and twice that including the unmeasured species (Akagi et al., 2010) whereas EFPM for savanna fires is only 5-7 g/kg.

P46, L9-10: You can get ER and MCE in real time as long as the plume is all wind-driven and there is not a vertical velocity profile across the path as would be the case on most larger fires except during RSC.

P48, L17-26: Somewhere in this paragraph would be a good place to compare to the RSC results of Bertschi et al (2003) Table 3 and Christian et al. (2007) Table 3 and Figure 2. There is likely an EF difference for RSC in grasses (or dung?) and logs.

P48, L23-26: Koppman et al never passed review. Yokelson et al 1996 identified CH2O as a product of the pyrolysis process and there is no pyrolysis phase since both flames and glowing during smoldering drive pyrolysis. CH2O clearly is negatively correlated with MCE in numerous studies.

P49, L27-P50, L3: Table 5 of Christian et al presents more extensive measurements of smoldering dung that should also be compared to here

P50, L10: I would change “wildfires” to “unplanned grass fires” since Christian et al and Bertschi et al show that in wooded areas ignoring RSC may not be valid.

P52, L1-12: This is a good discussion and raises some important issues, but a little more could be added. First a clarification: There is another Yokelson et al 2003 paper that describes post-flight tests in which losses of up to $\sim$50% were seen for NH3 when comparing an open-path FTIR and closed cell system in the exact same smoke. A specific algorithm was developed to correct for those losses and, when applied to the flight data reported in the Yokelson et al 2003 paper already cited, it suggested that those values were low by 5-15%. Those low values were also carried across to the Sinha et al 2003 paper the authors cite. However, the authors study-average EFNH3

(1.3) is about 3.5 times higher (normalized to CO to roughly account for flaming smoldering differences) than the closed cell EFNH3 (0.26) that appears in Sinha et al for fires in the same national park. In fact the author’s data (Table 4) for the combustion type with an EFCO similar to that in Sinha et al 2003; the backing fire of plot 2, has an EFNH3 virtually identical to the value in Sinha et al. This suggests that the difference is largely due to the type of smoke sampled from the two platforms. The authors also got a significantly higher EFCO indicating that their sampling weighted smoldering more than the airborne work, which is acknowledged in their conclusions.

It may be helpful that our website has pictures of one of the fires we sampled in Kruger National Park whose EF are shown in Sinha et al. Following the link and hitting next several times may be informative for the authors. One sees a great deal of smoke that went “straight up” and would be hard to sample from the ground. The final “Madikwe Plume” picture shows a planned fire that jumped the control line, which could have been a serious danger in a ground-based deployment.

http://www.cas.umt.edu/chemistry/documents/yokelson/s2k/jpg_1815b.htm

In addition, as the authors correctly point out the EFNH3 for different fuel elements varied widely in Keene et al (0.03 to 1.37). Another thing to add is that Christian et al., 2007 measured an EF for smoldering dung of 5.55 g/kg. So it would only take on the order of 20 g/m2 of dung consumption to have a fairly large effect on the authors EFNH3. An elephant might be up to that job. One can imagine weakly lofted plumes from smoldering dung finding their way into the author’s optical path throughout the fire, which they say is the case for at least one of the heading fires. If this is the case, it in no way invalidates the author’s data, but instead strengthens the idea that they may be more sensitive to important aspects of the emissions that are missed by aircraft. So likely a combination of interannual variation in fuels, spatial heterogeneity in fuels within KNP, normal fire-fire high variability, imperfect correction factors, but mostly air versus ground sampling biases could all play a role.

Yokelson, R.J., T.J. Christian, I.T. Bertschi, and W.M. Hao, Evaluation of adsorption effects on measurements of ammonia, acetic acid, and methanol, J. Geophys. Res. 108, 4649, doi:10.1029/2003JD003549, 2003.

P52, L15 'suggested" not "demonstrated"

P53, L3-5: The data here doesn't make sense. The MCE starts off at "nearly 1.0," which is what we also see for flaming combustion (Yokelson et al., 1996). The authors get an MCE of 0.76 for their "pure smoldering." This suggests that the head fire smoke with an MCE of 0.86 was mostly (~60%) smoldering. So it indicates a transition from about 60% smoldering to about 100% smoldering, but also seems to confirm the tendency for the ground-based sampling to probe smoldering emissions, which one would also infer from the pictures at the link provided above.

P53, L7-12: A plot of mean BT versus MCE might be useful here, but the fact that the Fire 3 headfire and RSC have virtually the same mean BT ("mean BT: headfire 520±17 vs. RSC 512±18 K") despite the large difference in CO/CO2 ratios (~50%), when viewed from a helicopter, seems to prove that the probability of getting a daily snapshot of instantaneous MCE from space is essentially zero. I would think that this should be stated and in the in the abstract as a major and very useful conclusion that the speculation about those prospects can be put to rest. Or if I have reached the wrong conclusion, then these results need to be explained a lot better. Another major point on this section is that either the quoted MCE in the text or the CO/CO2 ratios in the Tables are wrong. The Fire 1 MCE cannot decrease from the headfire to RSC as stated in the text while the CO/CO2 ratio drops as stated in Table 3. So along with proofreading the text, the authors need to carefully examine their table entries.

P54, L15: One more fuel type prone to RSC needs to be added here and that is coarse woody debris or "residual woody debris," which can account for a large portion of the total fuel consumption in the important wooded savanna ecosystem and Brazilian pasture fires, as well as many temperate and boreal ecosystems. These "old logs" were

the topic of RSC measurements by Christian et al. (2007) and Bertschi et al. (2003) who predicted that RSC would be less important in grass fires as the authors have apparently confirmed.

P55, L9: I think that a sentence that NH3, CH2O (we normally see negative correlation with MCE for CH2O), and CO EFs were higher than in airborne studies is what one would expect for ground-based sampling and that this technique is well-suited for measuring unlofted emissions would suffice here. And no need to revisit this later or skip it here.

P55, L11-12: I wouldn't go so far as to say that the authors showed this technique can probe "open vegetation fires burning in a manner fully representative of wildfire phenomena." For one thing, the approach requires that the smoke is blown near horizontally towards the authors equipment and wildfires generally propagate in the same direction as the wind, but without control lines. A second issue is that wildfire smoke often rises vertically as is acknowledged later by the authors. The authors need to decide on one consistent assessment and then perhaps an outline of the main points will make the conclusion less prone to revisiting topics multiple times.

P55, L20: can omit the word "potential"

P55, L23-25: Re "our results generally agree well with those from previous airborne campaigns in the same study area which did sample elevated plumes (Sinha et al., 2003a)." So just above this you point out the disagreements with Sinha and now you say you agree with them. I think just one statement will suffice that your results have lower MCE than Sinha and higher EF for species associated with lower MCE and this nicely shows the different smoke that tends to be probed from the two vantage points.

P55, L28: search and replace "maybe" to "may be"

P56, L3 search and replace:)

P56, L9: I would omit the last sentence which adds nothing. Griffith et al already demonstrated the approach 20 years ago, the authors did their own deployment 4 years ago, and Fernandez Gomez have little or no experience sampling real fires.

P63, L27 “?” in references and they need to be checked thoroughly throughout

Table 1: date and lat/long are optional additions

Table 3: There are lots of questionable values, especially fire 1 RSC CO/CO2. Check thru and make consistent with text. Fire 2 and 4 seem most reasonable?

Table 4 compare to Bertschi et al 2003

Fig 2 caption: Re “It should be noted that BTs are unlikely to represent actual fire temperatures since the flames may under-fill pixels (see e.g., Wooster et al., 2005).” Flame temperatures are typically 1100C, glowing combustion occurs at about 800C and temperatures throughout the site of a fire would then range from ambient to $\sim$1100C. The “fire temperature” would depend on what you mean by that: just as humans have different temperatures (oral, anal, armpit, skin, etc). At the authors full scale of 538 C the flames are likely filling half the pixel? Common sense dictates that with a more distant view point the pixel temperatures would go down. We found that the MODIS airborne simulator at 30 Km on the ER2 failed to detect a 200 ha fire; likely larger than the average fire in southern Africa. The fire was also undetected by MODIS itself. Fires detected by MODIS active fire detection in the tropics at “1 km nominal resolution” have a typical 4-micron temperature of 60C, which simply means to me that the “vast majority” of the pixel is not burning at the instant the satellite passes over. Is there some advanced math that I am missing that retrieves information from this seemingly intractable problem? Also, If the authors de-resolve the data shown, what would the brightness temperature be of a 1 km box centered on and containing the whole fire? and would they be able to infer fuel consumption from that brightness temperature (another paper?)

Fig. 5: A very useful result is the dramatic peak in FRP after MCE has already declined,

seemingly showing that spatial extent of the fire affects FRP far more than MCE – even from a helicopter.

Fig 6c seems to show digital limitation on the horizontal axis.

Fig 7: Good figure, but ensure it is consistent with text and tables.

Fig 8: a picture of whatever column may have developed from the helicopter might be useful also. The caption seems to state that emissions from elephant dung are important, while the text may sometimes say they aren’t?

Fig 9: ensure consistency with tables and text.

Parting thoughts: A bit more on the FRE-based fuel measurements –even a paragraph - is needed. A slightly more realistic and consistent assessment of what was accomplished would be useful. One more quick revision where each major topic is visited once in a logical order would help. With these steps the paper could be excellent.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3529, 2011.