

## ***Interactive comment on “Spatial and temporal variability in the ratio of trace gases emitted from biomass burning” by T. T. van Leeuwen and G. R. van der Werf***

**Anonymous Referee #2**

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Spatial and temporal variability in biomass burning emissions, by van Leeuwen and van der Werf

Reviewer #2

I will not have the time to produce a more concise or polished review at this time, but hopefully this draft will be useful. This paper is from a group that is in the forefront of quantifying the amount and type of biomass that burns globally and in this paper they explore optimal classification schemes for specifying emission factors (EF) for trace gases. They also present a worthwhile argument that more work needs to be done measuring EF over a broader range of environmental conditions and geographic re-

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gions; accompanied by better documentation of the environmental conditions. The work is valuable and should be published, however, I feel the paper could be improved by expanding the discussion of the new work done in the core area while reducing other sections that are mainly background info and less conclusive and less relevant to the authors main goals. I describe the problem the authors tackle in some detail to provide context for my specific comments. Fire to fire variation in the relative amount of pyrolysis, glowing, and flaming is large and leads to large natural variation in the EF for the major gases emitted such as CO and CH<sub>4</sub> on any given day within a coarse ecosystem classification (e.g. savanna, forest, etc). Less abundant species that are nonetheless atmospherically significant also vary depending on fuel chemistry. This is especially true for those emissions containing non-C atoms such as N, S, or Cl; or those emissions that are produced by direct volatilization of plant-specific precursors. Many factors can influence fuel chemistry and the mix of combustion processes that ultimately lead to variation in EF. It is an n-dimensional problem, with n being a large number. Examples of individual factors include: fuel chemistry (influenced by phenology, deposition, soils, agricultural chemicals, etc), fuel spacing (which affects heat transfer), slope, fuel moisture, precipitation, wind speed and direction, temperature, relative humidity, etc. Whereas the latter 6 can change temporally on daily or shorter scales, the first 3 are more constant in time, but can vary significantly over small spatial scales. They are not all independent of each other and some are tightly coupled in nonlinear ways (such as precipitation, temperature, and fuel moisture). The earth's fire-prone ecosystems can be subdivided based on vegetation or other observable variables with various levels of detail. It is a reasonable hypothesis that a characteristic average and range of the environmental variables may exist at some level of stratification and that this combination of variables may correlate with the average EF.

A common approach has been to partition EF into fairly simple, 3-8 category vegetation schemes. Following in the footsteps of Ito and Penner, this work explores new stratification schemes for EF based on several observable global environmental parameters available to them at various spatial and temporal resolution namely: temperature, pre-

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precipitation, NDVI, and fractional tree cover (FTC). As a result of their investigation, the authors derive useful new results that determine the fraction of observed variability in EF that correlates with the above parameters at various temporal and spatial scales. In particular, they focus on whether EF stratification with detailed vegetation schemes, FTC, or climate windows (operationally defined as restricted temperature and precipitation ranges that further subdivide broadly defined traditional biomes) leads to more precise emissions estimates.

The suggestions to improve the scope of future campaigns are good, but some may not be feasible, such as field campaigns that last the entire dry season. The discussion of field platforms has some limitations I point out. That section could be eliminated or shortened. The material in the introduction on the combustion processes is already ubiquitous in the literature and does not need to be repeated in detail here. The attempt to give mechanistic reasons for variation in EF is based mainly on one older lab study that has been since been contradicted by other work and the situation could just be summarized as inconclusive in a few sentences. Sections 2.1 and 2.2 delay the reader from getting to the new results and the variables discussed there-in are not understood well enough to warrant a lengthy discussion.

Some of the major relevant questions related to this investigation are: 1) Is there enough sampling to support robust relationships for a new, detailed classification scheme; or are the detailed categories un or under sampled? (See my detailed comment later on figure 4.) 2) When there are a lot of EF data for a few narrowly defined ecosystem conditions, is the range in EF about the average small enough so that the dependence on the classification categories (the slope) is significant? 3) How do the author's new classification schemes affect both the total emissions and the distribution of the emissions? For some applications only the global totals matter. For other applications the global totals could be essentially unchanged, but a redistribution would be significant.

Specific comments follow: Page (P), Line (L)

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P23560, L15: I suggest replacing "datasets thought to drive" with "measurements of environmental variables that may correlate with"

P23560, L19: I suggest replacing "driver datasets" with "remotely sensed data"

P23560, L20: I suggest inserting "for EF" after "techniques"

P23560, L21: I suggest inserting "EF" before "measurements"

P23560, L21-24: The comparison of consumption weighted EF to simpler arithmetic means is worth presenting, but the difference is not large compared to typical global uncertainties. It would be useful to also note in the paper (and summarize here) how much the climate-window-specific EF depart from the arithmetic means (and how much data these departures are based on).

P23561, L26: I would replace ", or alternatively" with "and/or"

P23561, L3: change "on" to "to" or "for" or "in"

P23561, L23: delete "campaigns of"

P23561, L26: Could delete one of two overview references for SAFARI 92 and add one for SAFARI 2000: Swap, R.J., H.J. Annegarn, J.T. Suttles, J. Haywood, M.C. Helminger, C. Hely, P.V. Hobbs, B. N. Holben, J. Ji, M.D. King, T. Landmann, W. Maenhaut, L. Otter, B. Pak, S. J. Piketh, S. Platnick, J. Privette, D. Roy, A.M. Thompson, D. Ward, and R. Yokelson, The Southern African Regional Science Initiative (SAFARI 2000): Overview of the dry season field campaign, South African J. Sci., 98, 125-130, 2002.

P23562, L1: "southern Africa"

P23562, L14: Could add: ~"Akagi et al., (2010) produced a compilation and averages for EF by fire type that is weighted toward field measurements in fresh smoke." 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,

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doi:10.5194/acpd-10-27523-2010, 2010.

P23562, L24-25: Many small fires do not show up as hotspots or burned area and remote sensing products often have low sensitivity to CO and other BB products in the boundary layer, thus agreement between estimates using the two different approaches suggests, but does not prove, that they are converging on the right number.

P23564, L18: suggest changing “gassed out” to “outgassed” Section 2.1: I think it could be cut by a factor four and still get the solid, main ideas across. Examples of some possible changes if the section is retained are given next.

P23565, L6-7: Suggest replacing “Several different compounds, with theoretically every possible molecule,” with “Many different compounds”

L9: I think K and C are being confused here. 450K (150C) sounds too low for pyrolysis to be exothermic. I think it's closer to 700K, but it is a known number that can be looked up.

L10: Glowing is from ~800-1000K.

L10-11: The complex, flammable mixture (known as “pyrosolate”) is produced even at 500K; it does not begin at 800K as might be inferred from this sentence. I would delete this sentence as the release of flammables was already introduced on lines 6-7. Just a general statement that the gases evolved from the solid biomass by pyrolysis are flammable and get oxidized if they enter a flame could suffice.

L14-16: I would omit this sentence as the CO is partly from glowing and the CH<sub>4</sub> is partly from “aromatization.” Not sure it's known (rather than speculated) how important the flame chemistry is.

L26: “often referred to as” should be “which is related to” and combustion efficiency should be defined.

P23566, L6: It would be difficult for Oxygen to be in short supply in an open fire. I

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think the key to extinction is that, for the fire to propagate, the fuel geometry has to be such that about 5-10% of the energy released from a unit amount of burning fuel element is transferred to (and ignites) a unit amount of nearby fuel. The most common causes of extinction are a physical gap in the fuels that prevents sufficient heat transfer to additional fuels, rainfall, or fire spread into wet fuels.

Section 2.2. General: this section might be shortened a great deal and left rather vague. Some speculation in older papers about the impact of environmental variables has been proven wrong. Other older papers reached conclusions based on a limited amount of data that are in conflict with subsequent studies. I suggest redoing this section with one summary sentence per variable in a short paragraph. I also make a few comments on the existing text.

L14: I suggest changing: “In general, the four most important factors controlling EFs of CO, CH<sub>4</sub>, and CO<sub>2</sub> are vegetation characteristics, climate and weather, topography, and fire practices” to: “Important factors influencing EFs of CO, CH<sub>4</sub>, and CO<sub>2</sub> likely include vegetation characteristics, climate and weather, topography, and fire practices”. I am not sure if the true importance of the many possible factors influencing EF has been determined and also, in some sense, five factors were listed.

L19: “burning efficiency” is not defined. Is it combustion completeness (the fraction of available biomass that burned) or combustion efficiency (the fraction of fuel C burned converted to CO<sub>2</sub>)? The role of fuel moisture is not really known yet. For instance, crown fires spread at high rates with huge flames burning fresh foliage of extremely high moisture. Wet fuels can ignite if a sustained ignition source is applied. So this is an example of these issues being far more complex than scientists guessed they would be years ago.

Another general issue affecting section 2.2, other sections of this paper, and science in general is worth mentioning here. Often, a relationship that holds true in initial work is noted, which makes sense, but then future work does not support the relationship.

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An example of that involving fires follows. In Yokelson et al., (1996) and Goode et al., (2000) good correlation for NH<sub>3</sub>/NO<sub>x</sub> vs MCE was found for all studies published at that time. However, McMeeking et al., 2009 constructed a similar plot (their Figure 10) with the available studies at the later date and there was no useful level of correlation. Similarly, Figure 4 of McMeeking et al shows a very weak dependence of MCE (a function of CO and CO<sub>2</sub>) on fuel moisture that contrasts the more limited results of Lobert et al. Thus I think that mentioning the results of Lobert et al at the top of page 23567 – as well as a detailed discussion of how environmental variables affect EF - is not that useful given the limited amount of current knowledge. In general, there is very little hard data on how environmental variables effect the emissions from real fires and the limited amount of lab data is often conflicting and inconclusive. That limited amount of lab data is hard to find because it is buried in papers mainly focused on other topics. For example, in addition to the studies quoted above, there is information suggesting that fuel consumption is effected more by fuel spacing than by fuel moisture (Bertschi et al. 2003). McMeeking, G. R., et al. (2009), Emissions of trace gases and aerosols during the open combustion of biomass in the laboratory, *J. Geophys. Res.*, 114, D19210, doi:10.1029/2009JD011836.

P23567, L4-14: Efficient heat transfer between fuel elements is very important and not mentioned (see last reference above). In deforestation fires, huge logs can be consumed mostly by flaming combustion (Christian et al., 2007). Again, I think a summary statement that these variables impact emissions in complex poorly understood ways is sufficient at this point.

P23567, L10: “oxidized” should be “pyrolyzed” Section 2.2.2: Just saying that vegetation and the length of the fire season are influenced by climate should suffice. Not sure what is meant by “settlement and ecological support for” vegetation. The seasonal trend in emissions for savanna fires in Korontzi et al was actually just for Dambo fires and no trend was seen for Miombo fires, which account for a larger amount of biomass burned.

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P23568, L2: fuels are not always “oxidized” – should be “consumed.”

L4 “burning efficiency” = ?

L3-6: Lobert had a good idea to do this, but it was a very limited investigation of this in the lab, I don’t know of any field verification of these effects.

Section 2.2.3 Again a few simple lab trials may not be relevant. The key variable is that heat rises and an upslope fire therefore achieves better heat transfer from the burning fuels to the unburned fuels, which makes it spread faster if all else is equal.

P23568, L17: suggest replacing “has become for a large part” with “is mainly” since new analysis of the Antarctic ice core indicates that historical burning by humans may have been higher hundreds of years ago.

L19-24: Slash and burn fires may be less intense than fires featuring windrows, but it seems to mainly affect the particle emissions? It might be more important to mention that the fuels consumed in understory fires (e.g. shrubs and leaf litter - common in the Miombo) are different from the fuels consumed in deforestation fires (mostly large trunks) and whether or not these different types of tropical forest fires can be identified from space.

L24-26: The key here is variable relative consumption of the peat and forest overstory. In summary I recommend reducing section 2 to a few summary statements showing that both combustion and how it is affected by the environment are very complicated, almost completely unstudied, and poorly understood. Nevertheless, empirical relationships between satellite observables and EF may exist and should be explored.

P23569, L3-4: The more recent compilation of EF by Akagi et al., (2010) likely includes additional data of value to the authors.

P23569, L10-11: I think that the EF values for some categories for some species such as glyoxal in A&M2001 are from lab studies.

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P23569, L11-12 and 22-27: I thought the EF were already calculated in A&M2001. If the authors recalculated them, they may want to explain why they did that.

P23569, L20: The more detailed study of Susott et al., (1996) suggests a global average C fraction for biomass closer to 50%, but with a considerable range. I am not suggesting recalculating the EF again, but acknowledging the uncertainty in %C is relevant.

P23570, L20-27: This is an important point. Indochina and other areas could be added to list.

P23570, L29: change "the three" to "three"

P23571, L8: change "all in-situ" to "all these in-situ"

P23571, L11-15: Here is an example of what I mean by expanding the core discussion. It is interesting that "r" goes down when limiting to extratropical forest. Examining this further may be useful. Also the slopes for the biomes here could be compared to the previously determined slopes for those biomes in the cited studies. That would yield insight into how correlations evolve as more data is added. If section 2 is greatly reduced (as suggested earlier) the implications of this could be explored a bit instead since it is the new work.

P23571, L21-P23572, L4: interesting and worth including any quantitative data that may be in references. L26: add "per unit area" after "consumed."

P23572: general. I am confused by Figure 3. It's a good idea to show the cycle of biomass consumption shifted so the peaks match up and also superimpose the measurement cycle. But why is there one vertical axis? Which line is which? The text seems to say the solid line is measurements, but the figure caption seems to say the solid line is fire emissions. I guess MOM stands for "month of measurement." This discussion is worth including and expanding, but needs clarification. For instance, what is the relative amount of fuel consumption in the off-peak months compared to

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the peak month? Is the fuel consumption symmetrical about the peak so that roughly equal amounts occur before and after the peak? Is the peak month in the middle or near the end of the local dry season? These are important questions that the authors are very well qualified to address. Quantifying the off-peak biomass consumption could help build momentum for an off-peak measurement campaign.

P23573, L2: I would change "explain" to "model" since any reasonably accurate, predictive, empirical relationship is valuable.

P23573, L3: I would change "of the" to "in our"

P23573, L11: I totally agree with the justification given on lines 18-19 for using FTC, but I am not sure I understand what was done here. Many papers give the latitude and longitude of the fires sampled and the observed vegetation type for each fire sampled. It might be useful to compare the FTC at that lat/long to the observed vegetation type and the EF. It might also be interesting to compare mapped to observed vegetation type or the FTC before and after the fire? But was the FTC deresolved first? That may be necessary, but does it also prevent inspecting potentially useful information? In any case it should be clarified.

P23573, L21: Useful correlation may exist between monthly precipitation and EF and should be pursued, but that is different from the precipitation "dependence" of EF. For example, dead fine fuels dry within 1-several hours after rainfall or reductions in RH. Also, in developing the 1978 US National Fire Danger Rating System ([http://www.fs.fed.us/rm/pubs\\_int/int\\_gtr169.pdf](http://www.fs.fed.us/rm/pubs_int/int_gtr169.pdf)), precipitation duration was more important than precipitation amount as excess water simply runs off the fuels during intense storms.

Again if section 2 is greatly reduced, more time could be spent exploring/clarifying the effect of the resolution used for FTC, precipitation, and temperature. One difference between tropics and temperate zone is that the daily variation in temperature generally exceeds the seasonal variation in temperature in the tropics. Fires are generally lit after

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11-12 AM and out by 3-5PM in tropics. Would fires correlate better with the daily high if available, etc?

P23575, L2: Potentially related to the authors work there was some earlier work relating savanna fire emissions to PGREEN by Hoffa et al that was also used in Ito and Penner. In general there are older papers using non-vegetative classification schemes that could be compared to and/or discussed in more detail.

Hoffa, E. A., D. E. Ward, W. M. Hao, R. A. Susott, and R. H. Wakimoto (1999), Seasonality of carbon emissions from biomass burning in a Zambian savanna, *J. Geophys. Res.*, 104, 13,841–13,853.

Ito, A. and J. E. Penner (2005), Estimates of CO emissions from open biomass burning in southern Africa for the year 2000, *J. Geophys. Res.*, 110, D19306, doi:10.1029/2004JD005347.

Ito, A. and J. E. Penner (2004), Global estimates of biomass burning emissions based on satellite imagery for the year 2000, *J. Geophys. Res.*, 109, D14S05, doi:10.1029/2003JD004423.

Ward, D. E., W. M. Hao, R. A. Susott, R. E. Babbitt, R. W. Shea, J. B. Kauffman, and C. O. Justice (1996), Effect of fuel composition on combustion efficiency and emission factors for African savanna ecosystems, *J. Geophys. Res.*, 101, 23,569–23,576.

Hély, C., P. R. Dowty, S. Alleaume, K. K. Caylor, S. Korontzi, R. J. Swap, H. H. Shugart, and C. O. Justice (2003d), Regional fuel load for two climatically contrasting years in southern Africa, *J. Geophys. Res.*, 108(D13), 8475, doi:10.1029/2002JD002341.

P23575, L4: For length of the dry season it should be the consecutive number of months with low precipitation not the total out of 12. Should specify if this is the case.

P233575, L11: There are many references on the time lag for dead fuel moisture to approach equilibrium in the environment. E.g.: the 1978 Fire Danger Rating System, or Ralph Nelson's chapter in "Forest Fires" by Johnson and Miyanishi.

C11522

P23575, L12: I would change "driver" to "remotely sensed environmental" and then if the authors like, on line 13 explain that they are calling these remotely sensed environmental observables "driver data" or just "environmental data."

P23575, L14: recommend changing "all EF data for the" to "all our EF data for" Section 3.4: The correlation in Table 1 with EF could be compared to the correlation with vegetation type shown in Figure 2. This type of analysis is useful and is the core contribution of the paper. However, the relatively small sample size in the plots in Figure 4 should be noted as well as the need for more data.

P23576, L21: Yokelson et al., (2009) and Akagi et al., (2010) found that EF for tropical forest fires in the Yucatan were similar to those for tropical forest fires in Brazil.

Section 3.5: I think what the authors are doing here is stating that a biome average EF using all EF from that biome may not represent the average smoke for the whole biome. Therefore, they create climate windows and place the EF for each biome within their respective climate window. Then they compute a weighted average EF for the biome where the weighting factor is the GFED fuel consumption for that climate window in that biome. It should be mentioned that the GFED fuel consumption by climate window has some uncertainty and there are also other issues besides relative fuel consumption that affect the representativeness of the sampling. The new weighted-average biome EF are not very different from the straight average biome EF when compared to typical global uncertainties. Therefore, it might be interesting to reveal the range of climate-window-specific EF retrieved for the various climate windows. This could imply significant regional or seasonal variation. It should be recalled that ultimately one is limited by the fact that many compounds were measured in only a handful of studies.

P23577, L12: "a large body of" before "available" as some studies are missing.

P23577, L20: "campaigns" is the correct spelling.

Section 4.1: Is it possible to estimate uncertainties for the space-based products?

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P23578, L7-10: It is not known how wind and topography affect EF for real fires. See my intro for ideas on other variables that influence EF that are not explicitly included among the available remote sensing data.

Section 4.3: I recommend eliminating this section for reasons given below and then expanding section 4.1.

P23578, L17-21: CO<sub>2</sub>, CO, and CH<sub>4</sub> are fairly easy to measure by NDIR, GC, and FTIR, because they are not sticky or reactive. Differences between measurement techniques are more important for sticky or reactive gases. Yokelson et al., 2007 found good agreement between their airborne measurements for these gases and the tower-based measurements of Ward et al., 1992. And both experiments probed the convective phase of the fire. Residual smoldering combustion, which can last for weeks, does contribute emissions that are sometimes significant, but rarely sampled.

P23579, L10: Is there a reference for increasing MCE with sample height?

P23579, L20: The tower based measurements underestimated particle emissions by a factor of 2 in Brazil (Babbitt et al., 1996), but was OK for some gases. Ground-based sampling of savanna fires with long poles does not sound safe. Babbitt, R. E., Ward, D. E., Susott, R. A., Artaxo, P., and Kauffman, J. B.: A comparison of concurrent airborne and ground-based emissions generated from biomass burning in the Amazon Basin, SCAR-B Proceedings, Transtec, São Paulo, Brazil, 1996.

General: Ground-based measurements may probe smoke that has not cooled to ambient levels yet and the effects of this initial cooling are not included in most atmospheric chemistry models. Low-level airborne samples capture the smoke after cooling but before the bulk of the photochemistry, which is simulated in models. Most of the emissions from most fires are entrained in intense convection columns that are difficult or dangerous to sample from the ground. An airplane allows measurements of many more fires and the fires do not need to be planned in advance as is often the case for ground-based measurements. Measurements of both convected emissions in the air

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and residual smoldering on ground is ideal, but requires accompanying measurements of the fuel consumption feeding into each process. This is all discussed in Akagi et al., (2010).

P23580, L11: "fase" should be "phase"

P23581, L6: There is large natural variation in the relative amount of fuel consumed by flaming and smoldering. Also Yokelson et al., (1996) showed that delineating smoldering and flaming phases causes errors in estimating the true fuel consumption by process. It might be interesting to explore if the GFED fuel consumption, which is highly specific, can be used to estimate the flaming to smoldering ratio.

Section 4.5: The same climate window could produce different EF in different locations e.g. Russia and Canada. The weighted means presented here are almost identical to the arithmetic means, thus the range may be more interesting than the difference in average values.

P23582, L10-12: It seems that pine-oak forests high in the mountains of Mexico could be classified as extratropical? However, the measurements available so far were impacted by urban deposition for some nitrogen-containing gases.

P23582, L15: A temperate forest biome was added in Akagi et al., (2010).

P23582, L16-19: Rather than simply pose the question here, perhaps the authors can address/answer it. For instance, how does the correlation with vegetation type shown in Figure 2 compare to the correlation with FTC and other variables they explored. I think they can at least start to answer this question, which could be a useful additional contribution of the paper.

P23582, L26: I am not confident that burned area is better known than many EF; and in any case such a statement should be accompanied by references to that effect.

P23583: L14-17: Excellent point about a standardized, accurate protocol being needed for measurement of fuel and environmental variables and it could be combined with

C11525

more “boots on the ground” satellite validation (hotspot detection efficiency, burned area accuracy, etc). (“campaigns” is the correct spelling.)

Section 4.6: I would just collapse this into two sentences in the conclusions that ideally we need 1) more EF measurements in off peak months and key, un-sampled geographic areas, 2) more measurements of environmental variables accompanying the field EF measurements, 3) more validation of satellite products.

Conclusions may be revised as paper is revised. For now on P23584:

L9: Add 10% for uncertainty in fuel C

L18: There were some available papers that were not included

L21: “uncharted” means “unmapped,” “unsampled” would be better.

L25: EF based on a more uniform and accurate sampling protocol were summarized in Akagi et al., (2010).

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 23559, 2010.