Review of Andreae 2019 BB EF

By Bob Yokelson

My colleagues Christine Wiedinmyer and Kelley Barsanti contributed helpful suggestions to this review.

This paper performs a service to the community by copying and sorting large amounts of biomass burning (BB) emission factor (EF) data from other sources into a single document. It also provides a long list of original research papers. The work was done by a top-notch scientist who was among the pioneers of BB research and who, in addition to having a prodigious publication output, has served the fire community as chair of BIBEX and in producing earlier compilations of literature EFs. It should be published after a few straightforward improvements. First, transcription of a multitude of numbers is actually extremely difficult to do 100% error-free. Second, the manuscript is not comprehensive, nor an educational review, and has just minimal assessment at the global uncertainty level as shown in more detail below. Thus, a more appropriate title would be something like: "Selected literature-average EF for simple biomass burning categories"

Biomass burning is a huge, complex topic and so a full review (or Referee comment) would be equivalent to writing several papers. Due to time constraints, I will only make a quick partial audit of some values that yields some lessons learned and general conclusions, and then point out readily apparent technical issues in the order they appear in the draft.

I will note here that many updated averages calculated now will likely soon be superseded by large-scale recent (WE-CAN, https://www.eol.ucar.edu/field_projects/we-can) or planned work (FIREX-AQ, https://www.esrl.noaa.gov/csd/projects/firex-aq/) of unprecedented scope.

As a reviewer, I thought it incumbent to spot-check the accuracy of at least some numbers from recent important work since data harvesting was the main activity of the paper. I chose to briefly examine selected peat fire entries because the vast majority of the new data are from our papers, papers we contributed to, or a paper I reviewed; so I knew the backstory (and I'm reviewing another peat fire paper also).

The major new work is by Jayarathne et al, Stockwell et al (2 papers), Hatch et al., and Smith et al. All of these papers are used in this review (hereinafter A19) and are also on the Akagi 2011(A11) update website. Thus, one general point is this paper should mention that the A11 assessment has an update website as a community service (http://bai.acom.ucar.edu/Data/fire/). No new global averages for peat fires are computed in A11 primarily because >600 compounds are now identified from peat fires, tropical and temperate peat may burn differently, and a global average is not the only type of desired input. A19 does compute new "snapshot" literature average EF, but based only on tropical peat data, which may or may not be similar to true global averages, but in any case a quick accuracy check was in order.

A number of entries from Stockwell et al 2016 were copied correctly. I was particularly pleased to see that A19 did NOT quote the PM2.5 from Stockwell since it is clearly stated to be a subset of the more extensive PM2.5 data in Jayarathne. This has escaped some readers, so kudos to A19. Next though, I noted that the "BC" entry is actually the "EC" from Jayarathne. EC measurements can be inflated by charring of OC, and the BC by photoacoustic spectroscopy in Stockwell was 0.0055 or ~35 times lower. Also, there was no entry for SSA for peat despite the data in Stockwell et al allowing a reasonable SSA estimate at any wavelength. The EF for SO2 from Tab S3 of Stockwell et al 2015 is probably too high for a global average because it is the only EFSO2 in the study, and SO2 was below detection for most peat fires as revealed by consulting two other tables in the paper. (Factoring in below detection limit data to "averages" is tricky and I will not discuss it in detail here). I checked a handful of NMHCs that were correct, but did note that the sum of 2-methyl-butenes actually included the 3-methyl-butene in "S16", although this is a very minor issue. Is it fair to estimate an error rate from a few spot-checks? I don't know. Overall, this could be a great starting point along with A11, but not using the original material increases the chances of introducing errors!

I also decided to perform a quick check on the formic acid data since the HITRAN parameters for HCOOH were changed by a factor of ~2.2 in 2012, which impacts all orbital and suborbital IR retrievals from before then. In A11 we adjusted all the old data for HCOOH, acetol, and glycolaldehyde based on new IR cross-sections. I randomly chose Yokelson et al., (2003) to see if HCOOH was updated and was surprised to see our formic acid data and nearly all our data from our 2003 paper missing. I found our data in the Sinha et al., (2003) entry where it had also appeared. So I'm glad the data don't appear twice, although it would be easier to trace the source if quoting the original paper. *In any case the old incorrect value is still there.* As an aside, I also noted that Burling et al., 2011 is in the reference list, but the data are not in the spreadsheet, perhaps to avoid duplication?

So again, this is a good resource and a lot of papers were read with some caution per limited spot checks, but users should be encouraged to consult the original work to double-check or trace important values. I think I noted somewhere that A19 has a place to send in corrections so that is a good feature.

This brief dive into the data reveals some general issues that impact the whole paper. A literature average where every study is weighted the same may not be a true global average for many reasons including:

1) No effort is made to weight more modern measurements techniques. For instance A11 preferred thermal optical EC over plain thermal EC. Now we can probably prefer PAS or SP2 BC to any EC or at least be clear about the measurement. E.g. Li, H., Lamb, K. D., Schwarz, J. P., Selimovic, V., Yokelson, R. J., McMeeking, G. R., and May, A.: Inter-comparison of black carbon measurement methods for simulated open biomass burning

emissions, Atmos. Environ., 206, 156-169, https://doi.org/10.1016/j.atmosenv.2019.03.010, 2019.

2) The values are not weighted by available estimates of relative activity within the category. For instance, some examples:
    a) Clean-burning stoves and dirty open-cooking fires are lumped together and not weighted for the greater prevalence of open-cooking. A11 has separate cooking fire categories for this reason and that should be mentioned in the A19 text. Further, cooking fire studies in labs tend to see different amounts of pollutants than in field studies with some very different results (e.g. Coffey et al., ES&T, 2017, references therein, and references mentioned below).
    b) Wildfires and prescribed fires create a similar amount of emissions in the US annually, but wildfire measurements are much less common in the literature. Wildfire emissions were recently found to differ significantly from prescribed fire emissions (Liu et al., 2017). In practice, A19 included two studies with anomalously large EFPM (up to 4 times the average) that seem to have pulled the temperate forest average to a value in between the most advanced measurements of the wildfire and prescribed fire EFPM. However, separate EFs for prescribed and wildfires has potential to significantly improve air quality modeling.
    c) Crop residue burned in piles is lumped together with crop residue burned loose in the field without the detailed caveats provided in A11 about how drastically the emissions differ between the two burning styles. Some recent papers now estimate how the crop residue is burned (e.g. Lasko et al Environ. Res. Lett. 12 (2017).
    d) Grasses and shrubs are combined as “savannas”, but for the dominant moist savanna fires, the fuels are mainly grass and miombo tree leaf litter and then some logs late in the dry season. Some global models assume a pure grassland category. It would be more useful to users of this paper to include separate grassland and shrubland/woody savanna categories.
    e) The studies are not weighted by the amount of sampling: a study sampling 157 whole fires gets the same weight as a study grab sampling one fire (A11 uses weighting).
    f) Some attention is paid to how representative the sampling is, but not a lot.
    g) A global average may be inappropriate for a regional/seasonal application, or regional/seasonal EFs may improve global models. This is alluded to indirectly, but not stressed enough.

Another general issue relates to the most useful reviewer comment we got on A11. What has changed? People are busy and may be curious if changing their model input will matter or which species to double-check in detail. In response we added a figure showing all the large changes for major emissions between A&M2001 and A11. Something similar could be added to this study although the targets are less clear because A11 is updated on web and A&M2001 has been updated by private communication over the years. What has changed between A11 and A19 and the last update widely dispersed by private communication? A11 computed new values for

temperate forest (2014) and savannas (2015), and these are posted on the website. It might be best to compare to the 2015 web update, which includes all the updated averages.

In general A&M2001, A11, and A19 will all be useful resources and highlighting the overlap and complementary strengths will make all these resources more useful to the community. A11 can add A19 to their update page and A19 can do a better job of pointing to A11. Even A&M2001 has some important components (e.g. equations) that are not in A19.

It would be more important to include some assessment of what is new than the global totals in Table 3 if length is an issue. Global totals are/were interesting; especially in the early days of BB research to confirm global importance of BB, but they are less important now. Nearly all fire emissions are too reactive to be well mixed globally and even for relatively inert species such as CO, the location and timing is needed along with amount for inversions. The standard among modelers now is to compare emissions at the regional level.

Another general issue is that the goals and accomplishments of much of the recent BB EF research are not discussed and many new EF results are not included in the tables; even though the papers were used to some extent. At the time of A11 about half the NMOG (by mass) were still unidentified, yet they surely react in real plumes. Tremendous progress has been made in the last 5-7 years with PTR-ToF-MS, 2D-GC-ToF-MS, etc to identify more of the unknowns. In addition, the amount of sampling and especially the sophistication of the instrumentation for sampling of previously undersampled fire types has seen a substantial increase including agricultural fires, wildfires, cooking fires, etc. No amount of measurements can reduce natural variability, but we have nonetheless greatly decreased uncertainty in smoke chemistry, there is an important difference.

Along these lines, no rationale is given for selecting 121 compounds to include out of the 700 plus that have now been measured. No estimate is given of how much additional NMOG is unaccounted for by the A19 tables. These are major issues. The uncertainty in data from carefully-simulated lab fires, especially when scaled to field conditions, is less of a problem than completely ignoring the chemistry of much of the emissions. Other less sweeping issues arise from the apparently ad hoc approach to what data are included. For instance, the sum of all isomers is used for “terpenes” even though some studies speciate the terpenes and they have different reactivity and potential to form SOA. At the same time, lab data for the sum of dimethyl and ethyl amine (same mass) are not quoted and the only data reported provided separate results. It’s likely more important to speciate the much more abundant terpenes.

Another critical current issue that is not discussed is measurements of intermediate and semivolatile compounds. These species are important SOA precursors and we need the SOA precursors to get BB-PM and its significant health and climate impacts right. Expert assessment helps because this also gets into the realm where the EF of an SVOC and the EF of organic aerosol can depend strongly on the concentration of the smoke being measured.

Per the other reviews:

I read the comments of Referee #1 and they all seem reasonable.

Ichoku review: I agree with this Referee's important clarification/correction re delineation of top-down and bottom-up, but add a few points. Bottom-up estimates are difficult for many reasons, but top-down is perhaps presented in overly favorable terms and a bit incomplete. Aerosol emissions are not measured globally but estimated on an extensive scale based on column AOD. AOD is reactive and not conserved, and gaps in AOD exist due to clouds, the cloud mask, orbital gaps, extensive time between overpasses, etc., etc. Importantly, attribution of AOD to specific sources is highly uncertain: e.g. plume injection altitudes are not operationally measured, crop waste burning can occur in forest clearings, or cooking fires and crop residue fires occur side by side in Asia where industrial sources, biogenic SOA, and sometimes peat fires also contribute to AOD. Comparing top and bottom is however super helpful. Finally, top-down using CO exists in numerous studies and gets around the "reactive issue" for AOD, but not the other issues although CO sources may be better constrained than AOD sources. The use of CO in inversions is discussed on page 10 when estimating uncertainties in global totals. The potential to use multiple CO sources could be stressed. E.g.

Kopacz, M., Jacob, D. J., Fisher, J. A., Logan, J. A., Zhang, L., Megretskaia, I. A., Yantosca, R. M., Singh, K., Henze, D. K., Burrows, J. P., Buchwitz, M., Khlystova, I., McMillan, W. W., Gille, J. C., Edwards, D. P., Eldering, A., Thouret, V., and Nedelec, P.: Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), Atmos. Chem. Phys., 10, 855-876, 2010.

Further, since the topic of how much biomass burns is included, then another important approach to how much burns is scaling of a-priori bottom-up emissions to match surface and aircraft data and AERONET AOD as in Reddington et al.

Reddington, C. L., Spracklen, D. V., Artaxo, P., Ridley, D. A., Rizzo, L. V., and Arana, A.: Analysis of particulate emissions from tropical biomass burning using a global aerosol model and long-term surface observations, Atmos. Chem. Phys., 16, 11083-11106, https://doi.org/10.5194/acp-16-11083-2016, 2016.

I don't like unspecified "expert judgment." A11 gives several recipes for estimation of unmeasured EF and they recommend trying several. It may be helpful to reference this discussion and clarify which approach(s) were used in A19.

The short comment by Nic Surawski suggests using "burnt carbon" rather than "dry fuel consumed" as the EF basis. The valid underlying issue is that the %C of the fuel may not be the %C of the emissions, which can make the carbon mass balance (CMB) method less rigorous. Neither %C is known in most field studies but in principle char formation causes the %C of the emissions to be lower than the %C of the fuel. On the other hand, Santín et al., (2015) found that

“higher %C” forest fuel components tend to burn with greater completeness, which tends to cause the %C of the emissions to be higher than the %C of fuel. This tends to cancel the impact of char formation on EFs calculated by the CMB.

In charcoal kilns, large pieces of solid charcoal are formed and the large charcoal yield can be measured reasonably accurately. Bertschi et al., (2003b) describe one practical method to adjust the CMB to get EF both per kg wood used and per kg charcoal made.

The situation changes for landscape fires. The charcoal yields are small and the charcoal is manifested mainly as a fine powder mixed in the exported plume or ash layer or a thin black surface layer on otherwise unburned fuel. Further “burnt C” arguably becomes undefined and unmeasurable in practice since some biomass is “affected by the fire” in ways that do not make char. The canopy can be scorched (turned brown by heat from below), creating emissions, but no char. Distillation of stored terpenes in wood occurs at temperatures below those creating char. Deciding what part of a forest was part of the “carbon burned" is not well defined.

On a practical level, there is a large historical database in the literature on fuel consumption, which was estimated as pre-fire minus post-fire biomass. Examples are included in A11 and there is a recent compilation (van Leeuwen et al., 2014). In contrast, there are few to none data for “burnt C” for major fire types. In general though, the impacts on the EF from the CMB is likely less important than the need for more quality measurements of char yields from landscape fires. This needs to be addressed to improve C-cycling estimates since the char is a carbon sink.

References:

Santín, C., S. H. Doerr, C. M. Preston, and G. González-Rodríguez (2015), Pyrogenic organic matter production from wildfires: A missing sink in the global carbon cycle, Global Change Biol., 21(4), 1621–1633, doi:10.1111/gcb.12800.

van Leeuwen, T. T., van der Werf, G. R., Hoffmann, A. A., Detmers, R. G., Rücker, G., French, N. H. F., Archibald, S., Carvalho Jr., J. A., Cook, G. D., de Groot, W. J., Hély, C., Kasischke, E. S., Kloster, S., McCarty, J. L., Pettinari, M. L., Savadogo, P., Alvarado, E. C., Boschetti, L., Manuri, S., Meyer, C. P., Siegert, F., Trollope, L. A., and Trollope, W. S. W.: Biomass burning fuel consumption rates: a field measurement database, Biogeosciences, 11, 7305-7329, https://doi.org/10.5194/bg-11-7305-2014, 2014.

Line by line comments in P, L format

1, 10: “critically evaluated” is probably better as “considered”?

General on abstract include a sentence on how many species changed by e.g. a factor of two since A11?

1, 2: Some carbon cycle people argue that much of the CO2 from fires should not be counted as emissions if the vegetation grows back.

1, 22: A glance at Table 1 seems to show higher EFN2O than I expected. N2O has been found to account for <1% of fuel N while NH3 is a major fate of fuel N. Are the N2O/NH3 ratios in Table 1 high due to including older studies with artifact N2O in canisters? I think not, but worth checking.

1, 22: Insert "BB is the second largest global source of non-methane organic gases (Yokelson et al., 2008, A11)."

2, 3-4: Fire increases locally available P by raising soil pH. See Jordan, C. F. 1985. Nutrient Cycling in Tropical Forest Ecosystems: Principles and Their Application in Management and Conservation. Chichester: Wiley.

2, 5-6: suggest retiring the term "VOCs" and using non-methane organic gases (NMOG) to recognize important gas-phase emissions with intermediate and lower volatility. Cite the following or equivalent:

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E., Pierce, J. R., and Pandis, S. N.: Rethinking organic aerosols: Semivolatile emissions and photochemical aging, Science, 315, 1259–1262, doi:10.1126/science.1133061, 2007.

May, A. A., Levin, E. J. T., Hennigan, C. J., Riipinen, I., Lee, T., Collett, J. L., Jimenez, J. L., Kreidenweis, S. M., and Robinson, A. L.: Gas-particle partitioning of primary organic aerosol emissions: 3. Biomass burning, J. Geophys. Res.-Atmos., 118, 11327–11338, doi:10.1002/jgrd.50828, 2013.

Hatch, L. E., Yokelson, R. J., Stockwell, C. E., Veres, P. R., Simpson, I. J., Blake, D. R., Orlando, J. J., and Barsanti, K. C.: Multi-instrument comparison and compilation of non-methane organic gas emissions from biomass burning and implications for smoke-derived secondary organic aerosol precursors, Atmos. Chem. Phys., 17, 1471-1489, https://doi.org/10.5194/acp-17-1471-2017, 2017.

Hatch, L. E., Rivas-Ubach, A., Jen, C. N., Lipton, M., Goldstein, A. H., and Barsanti, K. C.: Measurements of I/SVOCs in biomass-burning smoke using solid-phase extraction disks and two-dimensional gas chromatography, Atmos. Chem. Phys., 18, 17801-17817, https://doi.org/10.5194/acp-18-17801-2018, 2018.

Jen, C. N., Hatch, L. E., Selimovic, V., Yokelson, R. J., Weber, R., Fernandez, A. E., Kreisberg, N. M., Barsanti, K. C., and Goldstein, A. H.: Speciated and total emission factors of particulate organics from burning western US wildland fuels and their dependence on combustion

efficiency, Atmos. Chem. Phys., 19, 1013-1026, https://doi.org/10.5194/acp-19-1013-2019, 2019.

2, 7: Cite review of O3 formation in BB plumes; Jaffe, D. A., and Wigder, N. L., 2012. Ozone production from wildfires: A critical review. Atmospheric Environment 51, 1–10, doi:10.1016/j.atmosenv.2011.11.063.

2, 7: change “other pollutants” to “secondary PM” or some equivalent term

2, 8: delete “emitted” – these last two changes provide at least minimal recognition that much of the BB-PM impacts are from secondary PM.

2, 9: Akagi et al., (2014) present likely the most comprehensive assessment of toxic gases in fire-line smoke (add to reference string).

2, 16: “disconcerting” perhaps, but given the difficulty of measuring how much BB occurs, not at all surprising.

2, 26: I would add “numerous” before “field” as there are probably too many recent and on-going studies to provide complete references.

2, 27: Most of the EF results can be found in just 2-3 journals. I’d rephrase “The results of these studies are, however, widely dispersed among hundreds of papers in a large number of journals” to “The results of these studies are dispersed among hundreds of papers”.

2, 28: add “on a global scale” after “data” since most papers do synthesis/comparison at some scale.

2, 29: define Akagi et al., 2011 as “A11” to facilitate further citation.

2, 30: After “emission factors.” insert ~ “I have provided informal updates to A&M2001 and A11 maintains an update website (http://bai.acom.ucar.edu/Data/fire/).

2, 32: Insert “first appeared” after “previous compilations” to make it clear updates have already been readily available.

2, 32: Why 28 out of hundreds of new species?

2, 32: Are any species in A19, but not the original A11? Text should be inserted to clarify that new species were in the updated tables and papers posted on the A11 update site and in informal updates to A&M2001 distributed by the author. Also would be ideal to insert a mention here of recent or planned work that will modify these values, i.e. campaigns I cited above.

2, 33: After “burning types” insert “following A11”

3, 9: since this paragraph paraphrases A11 should add “following A11” before “I only …”

3, 15: Add “solar” before “Fourier” and “spectrometry” should be “spectroscopy”

3, 21: Good place to add that some lab data is adjusted to reflect field conditions using “overlap species”, ERs, or MCE as discussed in Yokelson et al., 2013. I think that data appears to have been used.

3, 28: I would change “usually” to sometimes”.

3, 31: change “typically” to “may be”

The logic is that some lab studies were carried out in the Missoula Fire Lab using fuels that were locally-collected by forest fuel experts or fuels “Fed-Exed by forest fuel experts. The fires were burned at a scale with flame lengths etc close to real world conditions. Fuel moistures in the FIREX-2016 lab experiment were quite high for example. Canopy fuels sent from the SE US had fuel moistures on the order of 136% on a dry weight basis. Also some lab studies report data adjusted by the MCE, ERs, or field/lab ratio for overlap species (Selimovic et al., 2018; Stockwell et al., 2015; Yokelson et al., 2013; etc). *Most importantly, for a vast number of species, there is only lab data.* To some extent this is clarified on P4, lines 2-4, but these points are important to make consistently in a revised paragraph here.

4, 1: This MCE is of course unrealistic, but not even close to typical of most lab fires.

4, 5-9: This discussion is well done, but some references could be included for the reader interested in more details. The Bond group, for instance, has a number of papers that find lab attempts to replicate field cooking fall short. Stockwell et al 2016a show how MCE dropped off significantly from the lab to field and describe correction factors for the lab data. The risk of lumping all this data together should be clear as noted above.

4, 10-26: This section is good. Should the equation be numbered? Probably apparent that “mixing ratios” can be used interchangeably with “concentrations”?

4, 27: change “easy” to “straightforward”

4, 31: I would change “is readily” to “can sometimes be”. The fuel moisture evaporation contributes to mass loss in the lab and fuel moisture is variable within components and between components, which have different combustion factors. We use the carbon mass balance method in the lab, which has the advantage in the lab of minimal distortion of excess CO2 via mixing.

5, 1: We include EC or BC in sum of carbon.

5, 2: More accurately fuel %C tend to be 40-45 for crops and grasses, 50 for wood/foliage, 55+ for peat.

5, 7-12: The equation; number it and check it! What is $EF_{(X/Y)}$?

The equation as presented makes no sense. I think it's trying to say something like:

Say the ER mol/mol of C_2H_4/CH_4 was measured as 0.1, but the data needed to compute EF was not collected in the study. If we know from other work that a reasonable guess at $EFCH_4$ is 5, then EFC_2H_4 can be estimated as 0.1*(28/16)*5 or 0.875. If this is the intent of the text here, $EF_{(X/Y)}$, which is undefined should be EF_X? However, if EF_Y is not known, then it should be made clear this is not the same as a measurement of EFC_2H_4, but just an estimation. Thus this discussion, after any needed corrections, from line 7 on, belongs in the discussion of estimates, not under "conversion of units."

5, 17-23: This whole discussion is confusing and may have errors.

If you assume the EF_X is unknown but is proportional to EF_{CO} then that seems to just be suggesting using a corrected version of the equation above with CO as the reference species "Y". If so, then on line 19 ER_{CO} should be EF_{CO}, the mass conversion ratio needs to be included, and it makes sense to use the $ER_{(X/CO)}$ from the most similar fuel type available rather than a global average. I.e. crops are grasses so if $ER_{(X/CO)}$ is not known for crops, but is for grasses, use that instead of factoring in the X/CO ratio for e.g. peat and garbage burning.

On lines 19 and 20: what is ER_X/ER_{CO} anyway? Is it just $ER_{(X/CO)}$ used above? If so be consistent – especially since ER_{CO} uses what as a reference species??

Next, for flaming compounds using the consumption weighted average of all categories makes less sense than using the most similar biomass type category as noted just above. Otherwise, the implication is that fire type doesn't matter; inconsistent with the rationale for creating fire type categories in the first place.

Finally, on line 23, what is a subjective best estimate? Some procedure was followed that should be spelled out.

The next four comments are related because smoldering is a combination of distillation, pyrolysis, and glowing combustion; and both glowing combustion and flaming combustion can induce distillation and pyrolysis.

5, 28: change "combustion" to "flaming or glowing".

6, 4-5: I would change "Once most volatile matter is consumed during flaming combustion, the remaining char undergoes gas-solid reactions between oxygen and carbon at the fuel surface, called the smoldering phase" to "In addition to volatile matter being consumed by flaming combustion, char undergoes gas-solid reactions between oxygen and other gases and solid carbon at the fuel surface, called gasification or "glowing" combustion".

Then on 6, 7: change "pyrolysis, flaming, and smoldering combustion" to "flaming and smoldering combustion (vernacular for a changing mix of distillation, pyrolysis, and glowing)"

Also on line 7: since fires can have more than one plume say “the fire plumes at any place and time contain”

6, 15: “peatland” should be “peat” since peatland will have surface fuels that are consumed partially by flaming. Stockwell et al 2016b gives a better overview of how peatland fires play out than Bertschi et al., 2003a and Guillermo Rein’s group has published detailed papers on peat combustion dynamics.

6, 18: change “a nocturnal” to “the” and change “serious problems” to “limitations”. It is entirely possible for RSC to occur during the daytime and to measure RSC EF using ground-based sampling (e.g. Bertschi et al., 2003a; Christian et al., 2007, Akagi et al., 2013).

6, 19: I would change “will completely miss” to “have trouble measuring”

6, 20: add “or fire blow-ups” after “daytime convection”

6, 20-21: Change “get lost” to “may be distorted by mixing”

6, 21-26: It is not any harder to measure CO/CO2 near the source for RSC than it is for any other source, but it should be done from the ground (see references above). The main problem is the RSC component of fuel consumption is difficult to measure to get a weighted fire average for overall emissions. Thus this paragraph should end with “Ground-based studies of RSC can obtain EFs of trace species, but these are difficult to relate to the corresponding amount of fuel burned.” Delete the rest of the paragraph as it is misleading. Refer the reader to Bertschi et al., (2003a) for scenarios of how RSC impacts EF.

6, 32 – 7, 14: This discussion needs to be rewritten from a perspective with more realistic hopes for what MCE can accomplish. Figure 1 throws out almost all available useful data by using one point per study and needs to be deleted or replaced with something useful.

Some general comments followed by specific recommendations: MCE, CO/CO2, BC/CO, and BC/OA are all useful to illustrate how the relative amount of flaming and smoldering can cause BB EFs to vary; especially within a single fuel-/fire-type or study. MCE is most common and as MCE decreases the total products of incomplete combustion increase. The author cites numerous papers with examples of good correlation of EF, even for specific species, vs MCE and this helps make sense of the observed variability and might drive a model at a useful scale (TBD). Low MCE dependence can be “OK” too and can sometimes increase confidence that the average value is close to correct for a range of burning conditions (Table 4, Liu et al., 2016). CO is the indicator of smoldering, but smoldering is a dynamic mix of complex processes and a simple parameter based on two gases should not be expected to predict all the outcomes of thousands of relevant chemical reactions across the planet for all emitted species. On broad scales other factors like fuel type (as noted), fuel N (Burling et al., 2010), geometry (Bertschi et al., 2003a), weather, etc impact emissions and correlations decrease as more conditions are considered.

Every model has a scope and every model has limitations. There might be a user-specific scale/scope where the coverage and correlation of an EF vs MCE model are both adequate to improve emissions estimates. Figure 1 skips over that question, throws out the data, and just demonstrates the obvious conclusion that EF vs MCE is not universal. The proper next step in evaluating EF vs MCE is to compare slopes based on all the data in the original studies aggregated at some intermediate level. To illustrate what I mean I insert a table where that process is started:

	Fire Type	savanna		Trop for		Conifer		Eucalypt	
	Study	Yok-03		Yok-07		Burl-11		Reis-18	
Species		slope	r^2	slope	r^2	slope	r^2	slope	r^2
CH4		-48	0.87	-47	0.52	-96	0.94	-96	0.93
CH3OH		-21	0.8	-15	0.48	-40	0.98	nm	nm

A glance at the table suggests some potential for a “fire-type-specific” EF vs MCE model with the level of correlation and aggregation perhaps depending on species also. I have not pursued this due to lack of time and because MCE is not available operationally as fire model input anyway. For now MCE remains most useful as a way to partially deconstruct variability in reported EF data.

With the above discussion as background I suggest the following revisions at a minimum.

7, 3: change “unfortunately” to “however”

7, 4: change “general parameterization of EFs” to “global parameterization of all EFs”

7 4-7: delete “As an illustration, I show in Fig. 1a and 1b plots of the EFs of ethene (C2H4) and ethane (C2H4) vs MCE, based on the studies in the supplemental spreadsheet. In both cases, the results scatter widely, and especially the data from the lab studies, biofuel burning, peat fires, and RSC-dominated fires introduce a large amount of scatter.”

Fig. 1 is one point per study rather than comparing slopes using multiple points per study, which might tell a different story and preserves whatever information there is.

7, 7-8: change “The poor correlation between EFs and MCE has been noted previously” to “The limitations of EFs versus MCE have been noted previously”

7, 8-11: delete “In the case of ethene, the correlation using all data points is not significant (R2 = 0.07). However, when only the data from open vegetation fires are included (and after removing three outliers), the correlation improves to an R2 of 0.27. For ethane, the correlation coefficient is R2 = 0.38 for all data, but does not improve substantially by removing the peat fire data.”

7, 11-12: change “These results suggest the potential of using MCE as a meaningful, but rough predictor of EFs for at least some species.” To “The level of aggregation at which MCE is useful

as a meaningful, but rough predictor of EFs for at least some species has not yet been determined."

7, 13: change "supplement" to "original studies"

7, 14: insert "A new approach to modeling NMOGs from pyrolysis using PMF has potential (Sekimoto et al., 2018); especially if the factors can be related to operationally available input."

Sekimoto, K., Koss, A. R., Gilman, J. B., Selimovic, V., Coggon, M. M., Zarzana, K. J., Yuan, B., Lerner, B. M., Brown, S. S., Warneke, C., Yokelson, R. J., Roberts, J. M., and de Gouw, J.: High- and low-temperature pyrolysis profiles describe volatile organic compound emissions from western US wildfire fuels, Atmos. Chem. Phys., 18, 9263-9281, https://doi.org/10.5194/acp-18-9263-2018, 2018.

7, 15-20: This may be worth trying, but model estimates of fuel consumption by flaming and smoldering would be difficult to validate in the field since access during the fire is problematic. Also the MCE of flaming or smoldering can vary broadening predicted MCEs.

7, 20: The first paper probing the relationship between greenness and MCE was Hoffa et al., 1999. 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.

Korontzi et al., 2003 updated the MCE/Greenness relationship based on new MCE measurements and then combined measured MCE, MCE vs greenness, and EF vs MCE (from other work in the late dry season) to estimate early dry season OVOC EFs.

7, 22: In addition to Korontzi et al., 2005, greenness (PGREEN) was used to predict combustion completeness in Korontzi et al., 2004 and PGREEN was used to predict MCE by Ito and Penner, 2004 (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2003JD004423).

Korontzi et al., Modeling and sensitivity analysis of fire emissions in southern Africa during SAFARI 2000, Remote Sensing of Environment 92, 255–275, 2004.

This approach has potential, but so far has been used for savannas only and works best for species that correlate strongly with MCE. The results have not been tested with field measurements to my knowledge. The discussion might be revised slightly.

7, 22: Maybe wrap up this section with something like "For now we should use the average EFs, but be aware they can vary considerably fire to fire."

7, 27: Not sure what this means "The averages in this column can only be seen as general indications, since all types of fuels and burning methods are included,"

Pages 7-8 in general: A19 has adopted some of improvements of A11, which is good.

8, 4: after “category” it could be useful to cite this resource of garbage burning activity and EF: Wiedinmyer, C., Yokelson, R. J., and Gullett, B. K.: Global emissions of trace gases, particulate matter, and hazardous air pollutants from open burning of domestic waste, Environ. Sci. Technol., 48, 9523-9530, doi:10.1021/es502250z, 2014.

8, 21-22: An EF for particle number concentration is problematic and potentially meaningless or misleading due to rapid coagulation near sources! Warning label needed.

8, 23: EFs for “brown carbon” (BrC) as g/kg are problematic because there are likely hundreds of contributing trace components with different absorption cross-sections that are also evidently reactive. But there is BrC emissions data in the form of Ångström absorption exponents (AAE) and BrC absorption EFs (as m^2/kg following the Bond and Moosmüller groups) in the UV for fresh emissions from carefully simulated lab fires and numerous field fires for different BB types (Stockwell et al., 2016a, b; Goetz et al., 2018; etc). Total absorption EFs in the UV are also given for users who may prefer them.

To clarify misleading text: the discussions in Selimovic et al., (2018 and 2019) show AAE near 3.7 (field Forrister et al., 2015) and 3.3 (lab Selimovic et al., 2018) for fresh smoke, but decaying with age as shown in Forrister et al and with BrC accounting for ~50% of absorption at 401 nm in *“moderately aged”* smoke (Selimovic et al., 2018). Most of these papers are in the A19 tables, but BrC data, which is important as the author says, is not tabulated in general.

Forrister, H., Liu, J., Scheuer, E., Dibb, J., Ziemba, L., Thornhill, K. L., Anderson, B., Diskin, G., Perring, A. E., Schwarz, J. P., Campuzano-Jost, P., Day, D. A., Palm, B. B., Jimenez, J. L., Nenes, A., and Weber, R. J.: Evolution of brown carbon in wildfire plumes. Geophys. Res. Lett., 42, 4623–4630, https://doi.org/10.1002/2015GL063897, 2015.

Goetz, J. D., Giordano, M. R., Stockwell, C. E., Christian, T. J., Maharjan, R., Adhikari, S., Bhave, P. V., Praveen, P. S., Panday, A. K., Jayarathne, T., Stone, E. A., Yokelson, R. J., and DeCarlo, P. F.: Speciated online PM1 from South Asian combustion sources – Part 1: Fuel-based emission factors and size distributions, Atmos. Chem. Phys., 18, 14653-14679, https://doi.org/10.5194/acp-18-14653-2018, 2018.

The Goetz paper above and Jayarathne papers cited include data for ions and metals in PM. Major ions and metals are tabulated in A11, but not A19, a point worth making in A19.

8, 26-33: I would delete this paragraph or at least revise it extensively. In part because the “most” serious problem is subjective depending on the workers area. For instance, top-down estimates of BB are probably most concerned with the issues such as observational constraints I outlined in my general comment on top-down estimates above. Workers looking at SOA may care more about EFs for SVOC, etc. In general this represents the authors troubles measuring RSC from an aircraft and other issues could lead to the underestimates of regional CO emissions mentioned. Also, it’s misleading because RSC does not affect only tropical forest fires. RSC

accounts for a significant part of the emissions for all forest fires, pasture fires, and wooded savanna, and virtually all the emissions from peat fires for example. However, the situation is far from hopeless. Bertschi et al., (2003a) outlined a range of impacts when RSC accounts for 10% to 50% of the total fuel consumption in a fire. At the upper end with 50% of fuel consumption by RSC the CO2 and CO EF changed by about -7% and +13% respectively. The larger impacts of RSC are for other gases like NH3 and CH4. Further, in A11 the tropical forest EF were adjusted based on an assumed RSC component of just 5% per available evidence at the time.

9, 5: This discussion doesn't include all fire inventories so change "Three of them use a bottom up approach" to "Four of them (for example) use a bottom up approach"

9, 7: change "The other three products are top-down, based on fire radiative power (FRP):" to "Two other products are top-down:" since GFAS is bottom-up, FRP is still just based on hotspots, and (for example) Ron Cohen's group (Mebust et al) also has a top-down approach.

9, 9: Agree with Charles Ichoku, GFAS is bottom-up. In this section on how much biomass is burned it could help to foreshadow the later discussion of CO inversions, list sources of uncertainty, and the other issues I noted in my general comments above.

9, 25: Are global numbers for reactive gases still important? More important than Table 3 might be to include a summary of what is new in this compilation as discussed in my general comments.

9, 28 "the previous assessment" should be "A&M2001" since there are so many global estimates.

9, 30-32: The fire to fire variability and even real day to day variability for a single fire can be much higher than the standard deviation of the literature mean. This can be important in many modeling applications (Yates et al., 2016). Change to "global emissions uncertainties" on line 32.

10, 1-14: This discussion is useful and adds confidence to global totals. There is a large body of work in this area and I have not attempted a comprehensive critique, but like the idea of using multiple CO products as noted above.

10, 15: A11 also reported these differences so useful to change to "As noted in A11, major …"

10, 20-22: I would rephrase this to say that there has been good progress in OVOC and HCN emissions as just noted and in reducing the percentage of un-identified compounds, sampling under-sampled sources, measuring I/SVOC, and sampling post-emission evolution, but quantifying global activity levels remains difficult. This is to be expected due to clouds, orbital gaps, small fires, unknown injection altitudes and diurnal cycles, etc. More measurements can add info but not reduce natural variability. Measuring EF and quantifying biomass burned present a different set of challenges. Most model inputs cannot be measured operationally. Thus, the author's proposed CO inversions are just one idea.

10, 29: Table 1 doesn’t include the major new research front in I/SVOC when it comes to setting future priorities.

11, 6: The conclusions remain focused on the problem of estimating global totals, which is just one part of BB research. It may not be the most important part, but is probably the hardest. Bottom-up or top-down models are super-sensitive to plume injection altitude, terrain flattening, diurnal cycles, complex transport, and chemical/physical evolution; often at subgrid scales. These things cannot be measured operationally. Actual recent/upcoming work such as WE-CAN and FIREX-AQ focus instead on advanced instrumentation and combining an unprecedented scope of airborne and ground-based measurements with new satellite products. This will eventually also be helpful to estimating global totals.

In summary, a quick check identifies some significant improvements needed in the paper. This is all I have time for now, but I may comment again before the discussion closes.