Response to Reviewer 3 (Robert Yokelson)

I thank the reviewer (and his colleagues) for his positive and constructive comments. My responses are detailed below. (Reviewer comments in italics).

I will note here that many updated averages calculated now will likely soon be superseded by large-scale recent (WE-CAN, https://www.eol.ucar.edu/field\_projects/we-can) or planned work (FIREX-AQ, https://www.esrl.noaa.gov/csd/projects/firex-aq/)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

I am looking forward to seeing these results. They will be incorporated in the online spreadsheet that I am making available (see last sentence of the conclusions with URL of data archive).

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/).

This information was added in the Introduction.

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.

Since tropical peat fires are likely the largest global source of these emissions, and since there are no field data from extratropical open peat fires, this type of snapshot is the only available option.

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.

In general, the data set contains both BC (optical) and EC (thermochemical) measurements. Admittedly, this is not ideal, but the entire issue of BC and EC measurements is so full of problems (see our Andreae and Gelencser, 2006, paper for an overview) that I did not feel that this assessment was the place to take on this issue. In the case mentioned by Yokelson, I ended up selecting the EC value, since it seemed a more direct measurement that the PAS one. Both are peer-reviewed published results. I am open to suggestions, which value to pick. Also, there was no entry for SSA for peat despite the data in Stockwell et al allowing a reasonable SSA estimate at any wavelength.

SSA is an intensive optical property and as such does not fit into a Table of emission factors,

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).

Again, this is a published value, and I am not in a position to make up my own data by factoring in below-detection data in some arbitrary way. The fact that it is listed here with "N=1" and that the value is higher than most other emission factors should alert both the users to exercise caution and the researcher community to the need for more measurements.

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!

Actually, this is a typo. There should not have been a 2 there – this entry is the sum of measured methylbutenes. Corrected.

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?

I did my best to avoid double listing of data that were published in several different publications to avoid bias in the averages. The updated HCOOH, acetol, and glycolaldehyde data has been entered in the database.

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:

I deliberately refrained from applying weighting factors, because I did not feel that I have enough of a basis to derive objective and quantitative values for such factors. I did not want to bias the results based on some kind of factors based on "best guess" or "expert judgement". This paper has a global focus, and there are many regional differences. Appropriate weighting factors are likely different in different regions, and I doubt that anyone has the information required to derive globally representative weighting factors. Specific comments follow:

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.

I am well aware of this issue. However, what are the alternatives? Putting in some weighting factor based on "expert judgement" (which the reviewer dislikes, see below) on each study? I went on the assumption that as more "modern" studies accumulate, their weight will increase in the average.

As mentioned above, BC and EC are the most problematic category. The optical properties sensed by PAS can range easily over a factor of two for the same amount of soot carbon. The SP2 measures well-formed soot, but may miss some of the BC end of the BrC/BC continuum, which should be included with BC. For climate modeling purposes, do we want to know the actual mass of soot carbon, or rather the optically effective equivalent, BCe? And so on... After having been in the BC field for 40 years now, I just think there is no "best" measurement or best general answer, and no way to plug in some weighting factor to "correct" or eliminate bias. I am including a statement in Section 2.1 to make the reader aware of these issues.

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. All has separate cooking fire categories for this reason and that should be mentioned in the Al9 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).

This issue is discussed briefly in section 2.1. A full review of the widely diverse emissions from biofuel use would require a separate paper, if not several papers. I have selected as the basis of Table 1 those studies that reported measurements in actual households or lab studies that tried to recreate household conditions. I have not included modern clean-burning stoves used in first-world countries. In papers that studied both traditional and modern stoves, I have only extracted the data from traditional stoves. Consequently, my average is intended to be representative of traditional biofuel use. At present, there are huge and rapid shifts in the patterns of domestic fuel use, which make any weighting by activity of limited use. I have added a sentence referring to A11 in Section 2.1. Users with specific interests, who need less aggregated data, can easily obtain them by using the Supplement. I have also added a sentence in Section 3.2: "Valuable detail about the various burning types and further breakdown of some categories, e.g., biofuel use, into relevant subcategories can be found in Akagi et al. (2011)."

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.

As reflected in the standard deviations, PM emissions vary widely, and results are also highly dependent on measurement approaches (lab vs field, aircraft vs ground, optical vs gravimetric, etc.). I'm not sure whether we can at this point generalize that one type of fire always has larger emissions than another, based on the limited data available. Does the work of Liu et al represent all wildfires and prescribed fires worldwide? And again, the data are easily separated out using the Supplement Table.

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).

Maybe, but I don't know how much residue is burned in piles or loose across five continents. Again, this paper has a global focus, and thus cannot address fine-grained issues such as this.

*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.

There are always costs and benefits in splitting and lumping. Modelers who want to use this data would probably prefer a split between C3 and C4 plants over one between grasslands and savannas. Then, the difference between grassland and savanna is often not very clear. In a miombo woodland, or a southern or West African savanna, a large part of the fuel is grass. How to draw the line? Then, splitting reduces the number of data in each category, increasing the likelihood that "atypical" values bias the mean.

# *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).

Again, what is the appropriate weighting factor? 157 to 1? Or 10 to 1? I worked on the concept that a study represents a particular biome, and that in some cases there is only one measurement in that biome and in other cases there are many. For some studies that refer to several biomes, I have included these biomes separately.

*f*) Some attention is paid to how representative the sampling is, but not a lot.OK.

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.

I added a sentence in the conclusions: "While the approach here was focused on global averages, future work should also emphasize regional and seasonal differences in order to better support more highly geographically resolved modeling."

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.

I have now added a direct reference to the A11 website with an URL.

Some of the equations and other detail from A&M2001 have not been included, since I wanted to avoid duplication. The methods section is focused on what's different in the approach of the current paper from that in our previous one.

Comparing the EFs in this paper to previously published data is a valuable suggestion. However, I don't feel that it would be appropriate to make a comparison between the results in this paper and data provided informally by me or made available by others on a website. I have therefore decided to compare selected values from A19 to the EFs in A11. The results are in Fig. 2, and discussed in the text in Section 3.2.

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.

I was motivated to write this paper by requests for updated emissions data by several modelers, all of which were focused on global studies. In inversion studies, location and timing is provided by the model, whereas this paper is focused on emission factors and uses global emissions only as illustration of the magnitude of emissions and to highlight global scale uncertainties. Some of the motivation to include Table 3 was also that I am seeing this paper as an update of A&M2001 rather than as a completely new approach, which I am encouraging other authors to undertake.

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.

This paper is not meant to be a general review of the progress of BB emission studies. The main objective was to provide an update to the A&M2001 data set in published and referenceable form. All the papers have been included that I am aware of and which provide data from which emission factors can be calculated. Because of the restriction to field measurements, some of the really exciting lab studies are not included in the data on which the EFs for the different fire categories are based. I am always happy to include pertinent information if brought to my attention.

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.

The compounds were selected based on the availability of enough field data to derive meaningful estimates and the importance of the species for climate and/or chemistry or their use as burning tracers. To address the very serious issue of underestimation of total volatile organics emissions, I have added some discussion in Sections 2.1 and 3.2, and included the NMOG emissions estimates from the online updates to A11 in Tables 1 and 3.

Unfortunately, there are not enough consistent field data on specified terpenes to enable speciesspecific data in Table 1. Regarding dimethyl and ethyl amine, I have avoided including mixtures of isobaric species from PTRMS measurements that could not be resolved to specific compounds.

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.

This issue is now discussed in the conclusions.

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." All 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 Al9.

The methods used for estimating unmeasured EFs are discussed in Section 2.4 and the specific method used for each compound was given in the last column of Table 1. I now see that somehow this column was lost when the pdf was transferred to the published version.

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.

See my response to the comment by Surawski.

# Line by line comments in P, L format

# 1, 10: "critically evaluated" is probably better as "considered"?

I did evaluate them critically for validity and appropriateness for inclusion.

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

I added a sentence: "For key species, the updated emission factors are compared with previously published values."

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.

This issue has now been addressed in the Introduction: "While a significant fraction of the emitted  $CO_2$  is taken up again by vegetation regrowth, much of it remains in the atmosphere for years and potentially even up to centuries, e.g., in the case of tropical deforestation fires or peat soil burning (van der Werf et al., 2017). Model simulations suggest that in the absence of fires, atmospheric  $CO_2$  concentrations would be about 40 ppm lower, indicating the importance of fires for the atmospheric carbon budget (Ward et al., 2012)."

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.

The old artefactual data were not included.

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

Done.

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.

This is correct and important, but is not directly related to atmospheric emissions, the topic of this paper.

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:

Since VOCs is used much more widely, I am retaining it here when referring to the species set used in A&M2001. I am introducing NMOG for the more comprehensive species set measured in more recent work.

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.

Added here and/or further down in the text.

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.

Done.

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

Done

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

Done.

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

Done.

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

OK.

2, 26: I would add "numerous" before "field" as there are probably too many recent and ongoing studies to provide complete references.

Done.

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".

I looked at the reference list in the supplement and reached ten different sources before I had gone past the letter B in the authors list.

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

Done.

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

I prefer retaining the full reference.

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/).

Done.

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

Done

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

As mentioned above, criteria were availability of sufficient field data and perceived importance for climate, chemistry, and/or tracers.

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.

The fact that A11 included additional fire categories and species is mentioned already in my addition on p3, 16 (new), and does not need to be reiterated here. I prefer not to specifically refer to future activities.

#### 2, 33: After "burning types" insert "following A11"

The fact that A11 included additional fire categories is mentioned already in my addition on p3, 16 (new), and does not need to be reiterated here.

3, 9: since this paragraph paraphrases A11 should add "following A11" before "I only ..."

Not quite sure what the reviewer means here. Actually, the approach here is different from A11, since they generally used a blend of field and lab data and I used lab data only as a "last resort", as stated here.

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

Solar was added. With regard to "spectrometry" I am following the IUPAC Gold Book definitions. See my comments to the Ichoku review.

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.

Done.

# 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.

I replaced "usually" with "often" and changed "typically" to "may be". The use of field-adjusted lab data has also been added. I don't think this is the place to go into an extended discussion about the merit of lab vs field data. This discussion can already be found in the literature, especially in papers from the Yokelson team.

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

Extreme, but not altogether untypical of lots of the lab studies on biofuel burning, unfortunately. I pointed out in the text that this is an extreme example.

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.

Again, I really did not want to go into an extended discussion on this. This topic would require a separate review paper. I am just talking about data selection here.

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

I put mixing ratio in parenthesis behind concentration. In principle, this should not make a difference, since the properties are ratioed.

4, 27: change "easy" to "straightforward"

Done.

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.

Changed to "can be". The main point here was to point out that it is very difficult in the field.

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

Added: ", and elemental carbon [EC] or black carbon [BC]"

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

Ok, but many authors just use 45%. That's why the sentence starts with "Often". No way to go into the individual assumptions used in each and every study.

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 C2H4/CH4 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 EFCH4 is 5, then EFC2H4 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 EFX? However, if EFY is not known, then it should be made clear this is not the same as a measurement of EFC2H4, 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."

Equation numbers should not be necessary, since the equations are not being referred to further. If the Journal style requires it, they will be added. EF(X/Y) was a typo, it should simply be  $EF_X$ . The text was amended to point out that this is an estimate. The heading "Conversion of Units" was removed.

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

If you assume the EFX is unknown but is proportional to EFCO 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 ERCO should be EFCO, 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 ERX/ERCO anyway? Is it just ER(X/CO) used above? If so be consistent – especially since ERCO uses what as a reference species?

Sorry, another typo. It should have been  $EF_X/EF_{CO}$  and  $EF_{CO}$  etc. I hope that makes sense now. I use simply the proportionality of the emission of species X to that of CO to scale smoldering-dominated emissions.

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.

Unfortunately, the column in Table 1 that specified the estimation technique was lost in the process of creating the ACPD version. AV was actually only used for  $N_2O$ ,  $SO_2$ , DMS, and HCl. I started out using this category for flaming-dominant species, but in the end applied it only to some hetero-element-containing species, where the N or S content of the fuel are likely more important than fire type. I amended the text.

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

I added "Specifically, for missing values of total particulate carbon emissions, the sum of OC and EC emissions was used, and for aerosol potassium emissions in boreal forest fires I used the temperate forest value."

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".

Done.

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".

Done

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)"

Done.

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

Done.

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.

Done.

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).

Of course. But what I am leading up to here is the specific problem of measuring the contribution of RSC to fire-integrated EFs, given the RSC emissions into a shallow boundary layer where both pyrogenic and biogenic  $CO_2$  are present at high levels. Ground-based sampling gives EFs from specific RSC point sources, but the problem is integrating that into fire-integrated averages. I tried to rewrite this passage to make the point clearer.

6, 19: I would change "will completely miss" to "have trouble measuring" 6, 20: add "or fire blow-ups" after "daytime convection"

Done.

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

Done.

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.

Done.

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:

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. I guess I had not make my point clear enough for the reviewer. What I wanted to say is that, while MCE is a wonderful tool for specific parametrizations within a confined group of fires, it is not a generally useful "one size fits all" way of deriving unknown EFs or parametrizing EFs in global models. Yes, this is obvious to the reviewer, but not necessarily to all potential users. I also thought it would be worthwhile to test this possibility by using this fairly comprehensive data set. In contrast to what the reviewer states, I am not throwing out data, but using all data in the form of their means. If there were a significant general relationship between EFs and MCEs, this relationship should show up even clearer in the means than the individual values. I am making some changes as suggested by the reviewer to make this more clear, but I leave any deeper analysis of EF/MCE relationships to future authors.

With the above discussion as background I suggest the following revisions at a minimum. 7, 3: change "unfortunately" to "however"

Done.

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

Done.

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.

I changed the sentence to: "As an illustration, I show in Fig. 1a and 1b plots of the EFs of ethene  $(C_2H_4)$  and ethane  $(C_2H_6)$  vs MCE, based on the <u>average values from the</u> studies in the supplemental spreadsheet...". I don't see a reason to change the rest, since it is simply a description of what is in the figures.

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"

Done.

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."

Again, this is simply a description of the data and figure. I don't see what's wrong with that.

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."

Done.

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

Done.

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.: Highand 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.

A sentence regarding the Sekimoto study was added.

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.

OK.

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. Reference added.

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.

References added. I prefer not to go deeper into this specific issue.

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."

Done.

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,"

Changed to: "The averages in this column can only be seen as general indication of the magnitude of emission factors found in the lab studies, since all types of fuels and burning methods are included in the averages. However, the original data and references are provided in the supplement for readers interested in the details."

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

Thanks!

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.

Reference added.

# 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.

Text added: "The rapid coagulation of particles very near the source makes it difficult to choose the most appropriate plume age for such a measurement (Hobbs et al., 2003; Sakamoto et al., 2016; Hodshire et al., 2019). However, a survey of available meas-urements suggests that the ratio of excess particle number concentration to  $\Delta$ CO stabilizes at the scale of typical aircraft measurements in plumes as a consequence of the sharp decrease of the coagulation rate with increasing dilution (Janhäll et al., 2010). More field studies on the development of aerosol number concentrations and size distributions as a function of plume age under different conditions (fire size, wind speed, flux density, etc.) are warranted."

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 m2/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: Fuelbased 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.

Text and references added: "Providing EFs for this species is problematic because of the very complex and variable mixture of compounds that make up BrC as well as its potential for rapid change in abundance and optical properties during plume evolution (Forrister et al., 2015). To some extent, data on the optical properties of BB aerosols can substitute for direct measurements

of BrC (Stockwell et al., 2016a; Stockwell et al., 2016b; Goetz et al., 2018; Selimovic et al., 2018)."

Information about ions and metals was added in Section 2.1: "Emission data for ionic species and trace metals are not included in this data set. They are tabulated in Akagi et al. (2011), and additional information can be found in a number of papers (e.g., Goetz et al., 2018; Jayarathne et al., 2018a; Jayarathne et al., 2018b)."

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.

I changed the beginning of this paragraph to: "Regarding the role of vegetation fires in the global carbon cycle, the most problematic uncertainty pertains..." Some of the text in this paragraph has also been changed in response to other comments. At no point does the text state that the problem is limited to tropical forests.

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.

I eliminated the whole top-down/bottom-up terminology. See my response to Ichoku.

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.

Such a summary has been added (see above)

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

Changed to "our previous assessment".

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.

Done.

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.

Thanks!

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

Done.

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.

I prefer my text. The conclusions are the place where the author should present his take-away from the previous discussion. I admit to having a bias towards global perspectives and carbon cycle issues.

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

This has now been added in the conclusions.

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.

I admit to having a bias towards global perspectives and carbon cycle issues. I am also looking forward to the results of the campaigns mentioned by the reviewer.