Response to Reviewer 1

I thank the reviewer for his/her constructive comments. My responses are detailed below. (Reviewer comments in italics).

1-21: Fires are obviously a source of CO2, but it would be good to add a statement on whether fires are a net source of CO2 to avoid confusion

This is an extremely complex subject that requires comprehensive earth system models to address and goes far beyond the scope of this paper, which is focusing on gross emissions. To give the reader some indication of the magnitude of fire contributions to atmospheric CO_2, I have included the following sentences: “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).”

2-10: The Johnston et al paper estimated 339,000 annual premature deaths, the number mentioned here is an interquartile range.

The original text says “…accounting for as much as 600,000 premature deaths per year globally”, indicating that this number is an upper limit. It is not the interquartile range, which would be the difference between upper and lower quartile. In response to the reviewer, and to make things fully transparent, I have changed the text to “…accounting for up to 600,000 premature deaths per year globally (75th percentile of model estimates; Johnston et al., 2012)”.

2-22: Please specify the units, C or DM? Also, a range of estimates is not necessarily the same as uncertainty, please see final point below

The text here does not discuss biomass or carbon burned, which could be expressed as C or DM. Rather, it discusses carbon released, which could be expressed as C or CO_2. However, the sentence here is completely unambiguous: “…the estimates in these studies of the annual amounts of **carbon** released still range over a factor of three from 1.5 to 4.7 Pg a^{-1}.” The rules of the SI system explicitly forbid including the use of constructs such as “4.7 Pg C a^{-1}”, and suggest including this information in the text, as I have done here.

I did not state that the estimate range is the same as uncertainty. Rather, in the introductory sentence to this paragraph I state “… large uncertainties persist regarding the amounts emitted…”. This is illustrated in the following text by wide range of current estimates, which I feel clearly supports the existence of large uncertainties, without equating the range of estimates with the range of uncertainty.

2-34: To some degree this differentiation was also done by Akagi et al. (2011), would be good to credit them

Sentence on 2-34 modified to "…the other by Akagi et al. (2011), which included more recent data and additional species and burning types..."

3-19: I applaud using top-down constraint, but it also makes for blurring the distinction between bottom-up and top-down measurements. For example, it is widely accepted that there is about a factor 3 difference in AOD calculations based on bottom-up and top-down methods (e.g., Kaiser et al., 2012), merging both approaches may hide this issue and thus requires a bit more information on whether and when merging these estimates is appropriate. Also, the author talks about 'appropriate correction methods' but this is not further specified as far as I can see.

It was not an easy decision for me whether to include these data from remote sensing or not. In the end, I felt that most users of my compilation would be better served if this information was included, since it provided large-scale coverage and data for some species poorly sampled by other techniques, e.g., COS. Users who do not want these data included for particular reasons can easily eliminate them by using the spreadsheet in the supplement and removing the corresponding lines. I did not include any aerosol data in these estimates, as I do not feel that there is any way to address post-emission changes to reconstruct aerosol emission data from large-scale AOD measurements. The data included are only for species that are either long-lived on the time/space scales in question (e.g., COS and HCN), or for which emissions can be reconstructed with some confidence by the use of chemical modeling, e.g., ethane. The "appropriate methods" are actually already discussed in the preceding sentence "…included a correction for atmospheric transformations, using model calculations involving transport times and reaction rates of the species concerned." I have made this more explicit by changing the sentence to: "…appropriate correction methods (i.e., chemistry-transport model calculations to correct for atmospheric transformations)". Readers interested in the details for each paper would have to go back to the original papers, as going into specifics here would not be of interest to most readers.

One of my main questions is to what degree the approach of this paper ("Ideally, these measurements had been made within minutes after the smoke was released from the fires") differs from that of Akagi ("smoke that has cooled to ambient temperature, but not yet undergone significant photochemical processing"). What does that mean for the number of studies included, what does it mean for the average values, to what degree do ground-based studies (which in general include the smoldering phase) differ from airborne studies which may be biased towards flaming combustion with more pyroconvection, etc? The latter is mentioned in the text (e.g. 6-22) but in the end all measurements are averaged. In general, the modellers which will ultimately use this dataset need to know whether and why these numbers are different from the numbers being used so far. This is a key question but not addressed at all and a table that addresses these differences and potential causes for the most frequently used species would be welcomed

In the end, what both Akagi et al. and I want to represent is “fresh smoke”, i.e., the material that is emitted by the fire and that can serve as starting point for model calculations at scales typically used by a variety of models. In that sense, there is no difference between my approach and Akagi’s. Ideally, one would have an objective criterion, such as Akagi’s “cooled to ambient temperature”, but in reality I had to look at the data in some 400 papers, and had to decide for each species and each set of measurements whether in my judgement they represented “fresh smoke”. This led to the exclusion of numerous studies, of some shorter-lived species from some studies from which somewhat longer-lived species could be included, and the exclusion of some flights or samplings from studies where both fresh and somewhat aged plumes had been investigated. I don’t think there is any reasonable way to discuss these issues for each of 370 papers (now included in the revised version) and 121 species. In the end, I can only ask the user of these data to trust my judgement, gained over working on this subject for almost 40 years, about which data to include. Averaging over as many data (that are judged to be valid) as possible should reduce biases that result from the inclusion of any atypical data.

The issue of airborne vs. ground-based studies has been discussed extensively in the literature, including A&M2001, A2011, Burling et al. (2011), and briefly in this paper. I did not feel that there is anything new that I could contribute here other than highlighting the problems resulting from this issue. In response to the reviewer, I added the reference to Burling et al. (2011) on p. 5 and a sentence to the last paragraph of section 3.2: “A representative measurement of fire-average $\Delta CO/\Delta CO_2$ emission ratios from large forest fires is very difficult if not impossible, as ground-based measurements in such violent fires are not possible and aircraft measurements are prone to undersampling the smoldering emissions, especially the contributions from RSC.”

8-26: This is a somewhat surprising statement to me. Differences in bottom-up and top-down results can originate from uncertainty in many parameters, emission factors being one of them. The standard deviation of CO in boreal and temperate forests is relatively speaking not that much larger than in savannas which to me is not surprising given the large variability in moisture regimes and tree species and density in those forests. I feel it would be more useful to analyze whether there is a difference in ground and airborne studies to say something about RSC.

I’m not quite sure what the reviewer is getting at here. In response, I have changed “…which may be responsible for…” to “… which may significantly contribute to…”. The role of RSC is highlighted in the new sentence added: “A representative measurement of fire-average $\Delta CO/\Delta CO_2$ emission ratios from large forest fires is very difficult if not impossible, as ground-based measurements in such violent fires are not possible and aircraft measurements are prone to undersampling the smoldering emissions, especially the contributions from RSC.” Regrettably, I am not able to propose a simple solution to this problem.

My other main point of criticism relates to Table 2 and the statement in the conclusions that the uncertainty in biomass burning emissions nowadays is as large as in the 2000s. Table 2 shows various estimates and the large range stems for a substantial part from outliers such as

FLAMBE which predict 10 times higher emissions in tropical forests compared to savannas, totally different from for example GFED4 and GFAS1.2 (derived from GFED3). I understand that it is beyond the scope of this paper to assess which one is right but this deserves some explanation, for example using previously mentioned top-down estimates based on CO. Simply combining the 8750 Tg DM in tropical forests from FLAMBE and the CO emission factor (105 g CO per kg DM) indicates CO emissions from this biome alone of 900 Tg CO per year, something not corroborated by inverse estimates and also at odds with the best estimates of deforestation (e.g., Houghton and Nassikas, 2017, https://doi.org/10.1002/2016GB005546).

My point here is to highlight a problem, not to analyze the validity of the various studies that are listed in Table 2, for which I am not qualified and which would also go far beyond the scope of this paper. Therefore, I had to accept each of these peer-reviewed (by peers much more qualified than me) and published studies at face value. I did not feel that it was my place to call any of these studies an “outlier”. What I am hoping for is that the community sees these large discrepancies by having them juxtaposed in one Table, and makes efforts to address them. Thus also the deliberatively provocative statement that “the uncertainty in biomass burning emissions nowadays is as large as in the 2000s”. I would very much welcome a paper by the remote sensing community, maybe in the form of an intercomparison project, that will prove me wrong! But to address the reviewer’s specific concern about FLAMBE, I have added a statement paraphrasing his/her comment and a reference to the review itself: “The inverse analyses may also be useful to indicate unlikely estimates based on remote-sensing techniques. For example, the burning of 8750 Tg dm in tropical forests estimated by FLAMBE, combined with the corresponding EF_{CO} (105 g kg^{-1}) would produce CO emissions of 900 Tg a^{-1} from this biome alone, well above the range of inverse CO emission estimates for all open burning (see also the comments by Reviewer 1, https://doi.org/10.5194/acp-2019-303-RC1).”

Response to Reviewer 2 (Charles Ichoku)

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

Page 3, Line 15: I believe it is more conventional to refer to the FTIR technique as "spectroscopy" rather than "spectrometry".

There is a lot of discussion about this in the literature. I use the definition of the "IUPAC Gold Book", the authoritative reference for chemists: "Spectroscopy: The study of physical systems by the electromagnetic radiation with which they interact or that they produce. Spectrometry is the measurement of such radiations as a means of obtaining information about the systems and their components." (http://goldbook.iupac.org/html/S/S05848.html).

Page 6, Line 9: Change "depending of" to "depending on".

Corrected.

Page 9, Lines 5-10: My main concern here is the use of Fire Radiative Power (FRP) as the sole basis to distinguish "top-down" satellite BB emissions methods from "bottom- up". All satellite BB emissions methods described in the article utilize satellite observations (fire-pixel counts, burned area, or FRP) as a way of estimating the biomass burning activity. The use of one or another parameter (FRP or not) does not make a method "top-down" or "bottom-up". Since the driving variable for estimating BB emissions are the factors that convert the activity to emissions (e.g. emission factors, as eloquently discussed in the article), it is the spatio-temporal distribution/configuration of the original input emissions, which went into deriving these EFs that determine whether a method is "top-down" or "bottom-up". If those input emissions were observed at a few locations and limited time periods, and then scaled up globally, the method is "bottom- up". But, if the input emissions were observed globally and regularly, and then scaled down to their sources, it is "top-down", as in the use of satellite-derived aerosol optical depths (AOD) of smoke-dominated aerosols to constrain the "emission coefficients" used to derive the emissions. Bearing this in mind, among the satellite methods described in this article, only QFED and FEER used globally-observed AOD to derive the coefficients that were then used to derive their final BB emission products, and thus may be categorized as "top-down". The others (including GFAS, which is scaled to GFED emissions) used locally-observed BB-emitted constituents to derive emission factors that were then generalized for their global BB emission products, and thus may be categorized as "bottom-up".

Since the distinction between top-down and bottom-up satellite products is not really important to the discussion in this paper, I am limiting the use of "top-down" to inverse studies, where an emission is estimated from a concentration field. "Bottom-up" is now used only as a general term, where emissions are estimated based on a product of activity and emission factor estimates.

The satellite emission products are now referred to based on the quantity sensed (FRP, burned-area, etc.).

Page 9, Line 17: I am concerned about the use of FAO (2015) as the primary reference for a quantitative value, as I am not sure whether FAO (2015) was peer-reviewed. I believe it would be better to find and cite the original (peer-reviewed) source of the 53 Tg/yr estimate reported in FAO (2015).

The FAO studies are not based on a particular paper, but on reports from the individual UN member states to FAO regarding the amounts of agricultural and forestry activity in each country. As such, the FAO reports are only as good as the quality of reporting from each country, which of course varies considerably. Nevertheless, the FAO reports are generally considered to be an authoritative source on agricultural activity. FAO states: “Prior to publication, these reports are subject to quality control through a standardized peer-review mechanism that allows relevant stakeholders to provide valuable feedback on initial drafts of the reports.”

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. 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).*

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 species-specific 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." 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 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 fire-line 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 on-going 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, l6 (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, l6 (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 average values from the 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.: 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.

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 ΔCO 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: 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.

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: A11 also reported these differences so useful to change to "As noted in A11, 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.

Emission of trace gases and aerosols from biomass burning – An updated assessment

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Abstract. Since the publication of the compilation of biomass burning emission factors by Andreae and Merlet (2001), a large number of studies has greatly expanded the amount of available data on emissions from various types of biomass burning. Using essentially the same methodology as Andreae and Merlet (2001), this paper presents an updated compilation of emission factors. The data from over 370 published studies were critically evaluated and integrated into a consistent format. Several new categories of biomass burning were added, and the number of species for which emission data are presented was increased from 93 to 121. Where field data are still insufficient, estimates based on appropriate extrapolation techniques are proposed. For key species, the updated emission factors are compared with previously published values. Based on these emission factors and published global activity estimates, I have derived estimates of pyrogenic emissions for important species emitted by the various types of biomass burning.

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1 Introduction

Biomass burning, in the form of open vegetation fires and indoor biofuel use, is one of the largest sources of many trace gases and aerosols to the global atmosphere. For some important atmospheric pollutants, like black carbon (BC) and primary organic aerosol (POA), biomass burning is the dominant global source; based on the estimates of Bond et al. (2013), it accounts for 59% of BC emissions and 85% of POA emissions worldwide. Open vegetation fires alone represent about one-third to one-half of global carbon monoxide (CO) and 20% of nitrogen oxide (NO_x) emissions (Olivier et al., 2005; Wiedinmyer et al., 2011). Fires are also a major source of greenhouse gases, including carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) (Ciais et al., 2013; Tian et al., 2016; Le Quéré et al., 2018). 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). Biomass burning is the second largest global source of non-methane organic gases (Yokelson et al., 2008; Akagi et al., 2011). Numerous other studies have reached similar conclusions about the importance of biomass burning for atmospheric composition (e.g., Crutzen and Andreae, 1990; Andreae and Rosenfeld, 2008; Andreae et al., 2009; Kaiser et al., 2012; van der Werf et al., 2017).

The resulting perturbations of the atmospheric burdens of trace gases and aerosols have important consequences for climate, biogeochemical cycles, and human health. Aerosols from biomass burning affect the regional and global radiation balance and impact cloud properties and precipitation (Andreae et al., 2004; Andreae and Rosenfeld, 2008; Rosenfeld et al., 2008; Ward et al., 2012; Tosca et al., 2013; Rosenfeld et al., 2014; Jiang et al., 2016; Braga et al., 2017; Cecchini et al., 2017; Hamilton et al., 2018; Thornhill et al., 2018). By shifting the proportions of direct and indirect solar radiation, they also influence primary productivity and thereby forest growth and agricultural production (Artaxo et al., 2009; Rap et al., 2015; Malavelle et al., 2019; McKendry et al., 2019). Fires mobilize nutrients, such as nitrogen, phosphorus, and potassium, which can deplete local ecosystem nutrient reservoirs on one hand and provide nutrients to other ecosystems by atmospheric transport on the other (Andreae, 1991; Andreae et al., 1998; Mahowald et al., 2008; Chen et al., 2010b). The volatile organic compounds (VOCs) and NO_x in biomass smoke undergo smog photochemistry in the atmosphere, leading to the production of ozone, secondary organic aerosols, and other pollutants, which impact plant productivity (Crutzen and Andreae, 1990; Andreae, 1991; Robinson et al., 2007; Jaffe and Wigder, 2012; May et al., 2013; Pacifico et al., 2015; Hatch et al., 2017; Yue and Unger, 2018). These gaseous pollutants, and even more so the particulate matter from biomass burning, pose grave risks to human health (Naeher et al., 2007; Akagi et al., 2014; Dennekamp et al., 2015; Knorr et al., 2017; Apte et al., 2018). Recent estimates of global excess mortality from outdoor air pollution range from 4.2 to 8.9 million annually (Cohen et al., 2017; Lelieveld and Pöschl, 2017; Shiraiwa et al., 2017; Burnett et al., 2018; Lelieveld et al., 2019), with smoke from open vegetation burning accounting for up to 600,000 premature deaths per year globally (75th percentile of model estimates; Johnston et al., 2012). In addition to outdoors exposure, pollution from indoor solid fuel use, much of it biofuel burning, has been estimated to cause 2.8 million premature deaths annually (Kodros et al., 2018).

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In view of the immense impact of biomass burning emissions on climate, ecosystem function, and human well-being, it is disconcerting that large uncertainties persist regarding the amounts emitted and their spatial and temporal distribution. For bottom-up emissions estimates, two basic types of information are required: the amount of the various types of biomass burned as a function of time and space and the emission factors for the various emitted species, i.e., the amount of a given species emitted per unit mass of biomass burned. Considerable effort has gone into quantifying the magnitude of open biomass burning by remote sensing approaches (Mouillot et al., 2006; Reid et al., 2009; Mieville et al., 2010; Wiedinmyer et al., 2011; Kaiser et al., 2012; Ichoku and Ellison, 2014; Darmenov and da Silva, 2015; Chuvieco et al., 2016; van der Werf et al., 2017), but the estimates in these studies of the annual amounts of carbon released still range over a factor of three from 1.5 to 4.7 Pg a^{-1}. A model intercomparison based on state-of-the-art dynamic global vegetation models (DVGMs) yielded an even wider range of 1.0 to 4.9 Pg a^{-1} (Li et al., 2019a).

Efforts to narrow the uncertainties in the emission factors for the large number of species emitted from the diverse types of burning are ongoing in the form of numerous field campaigns and laboratory studies. The results of these studies are,

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however, widely dispersed among hundreds of papers in a large number of journals, each dealing with a particular campaign or experiment. Over the last two decades, there have been two efforts to synthesize these data on a global scale, one by Andreae and Merlet (2001; below referred to as A&M2001) and the other by Akagi et al. (2011). The latter included more recent data and additional species and burning types, and is available in updated form at http://bai.acom.ucar.edu/Data/fire/. As part of the The Fire INventory from NCAR (FINN) model, Wiedinmyer et al. (2011) selected data from these two sources into a “best estimate” set of emission factors. In the present study, I am presenting an updated set of emission factors, which includes the results of studies published since the writing of the two previous compilations. It provides emission estimates for 28 more chemical species, for which a sufficient amount of field data has become available since A&M2001, as well as an extended set of burning types. The extratropical forest category is differentiated into boreal and temperate forest burning, domestic biofuel use is separated into non-dung and dung burning, and peat fires and domestic waste burning are added as new categories. Based on these emission data and recent activity estimates, I present a compilation of global emission amounts and make some recommendations regarding priorities for future investigations.

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2 Methods

2.1 Data selection

This paper applies the same methodological approach as A&M2001, and therefore the methods section will only provide a brief overview of the definitions and calculation methods used, and highlight those points where the present approach differs from the previous one. For all other details, the reader is referred to A&M2001. The original data, which form the basis of the emission factor averages presented in Table 1, can be found in an Excel spreadsheet in the Supplement.

With few exceptions, and consistent with the approach used in A&M2001, I only used results from field measurements in young fire plumes for the compilation of the emission factor data in Table 1. Ideally, these measurements had been made within minutes after the smoke was released from the fires to avoid significant chemical changes during atmospheric aging, especially in the case of reactive trace gases. This is only possible, however, when sampling at the ground or from aircraft very close to the fire. In many other cases, aircraft were sampling at some distance from the fires, often without actually knowing the exact location of the fire. In such cases, I have rejected the data for the more reactive trace gases. A special case is presented by emission data calculated from remote sensing by either satellite measurements or ground-based solar Fourier Transformation Infrared (FTIR) spectrometry. Here, the authors have often included a correction for atmospheric transformations, using model calculations involving transport times and reaction rates of the species concerned. Because of their large spatial and temporal coverage, such measurements are quite valuable, and I have therefore included some of them in this assessment, as long as they were either dealing with long-lived species or used appropriate correction methods (i.e., chemistry-transport model calculations to correct for atmospheric transformations) (Rinsland et al., 2007; Mebust et al., 2011; Tereszchuk et al., 2011; Tereszchuk et al., 2013; Schreier et al., 2015; Viatte et al., 2015; Lutsch et al., 2016; Adams et al., 2019). They can be compared with in-situ measurement results by referring to the original data in the Supplement spreadsheet.

Another special case are the emission factors for gaseous elemental mercury (Hg^0). Here, only relatively few actual field measurements are available for most of the combustion types listed in Table 1. Therefore, I have followed the approach of Friedli et al. (2009) and included Hg^0 emission factors from studies that are based on the Hg content of the fuels and the assumption of total volatilization of Hg from the fuel during combustion, which appears well justified for this volatile element.

Generally, the results from laboratory combustion studies have not been included in the emission factors for the different fire types in Table 1, but they are given for comparison in a separate column in Table 1. The reason for this decision is that such experiments often do not reproduce realistic burning conditions in the field. For example, it has been shown that the emissions of many trace gases are strongly dependent on fuel moisture, temperature, and other fire environment parameters (e.g., Chen et al., 2010a; Robertson et al., 2014; Liu et al., 2017; Thompson et al., 2019). The fuels in lab experiments, however, may be well aged and dried, and thus have a much lower moisture content than fuels in the field. This can be seen in the values of the modified combustion efficiency [MCE; the ratio of $\Delta CO_2/(\Delta CO_2+\Delta CO)$] in many lab studies, which are much higher than those typical in field burns, an extreme example being the study by Sirithian et al. (2018), who reported a mean MCE of 0.9996 in a lab study on biofuel burning. Therefore, lab results are only used in some special cases, where little or no field data are available and where the lab data appear representative based on their MCE, e.g., Christian et al. (2003), or had been adjusted to reflect field conditions using “overlap species”, ERs, or MCE as discussed in Yokelson et al. (2013b). Some lab data are also used as estimates in Table 1 and are shown in italics.

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The studies on emissions from biofuel burning for cooking or heating represent a borderline case, as they are often conducted in a laboratory environment, but with an effort to simulate actual fuel use conditions and stove setups used in the households. Here, I have favored studies performed in actual households, but also included results from lab studies that appeared to realistically emulate field conditions. Results from modern residential biofuel combustion units, such as automated pellet burners or modern low-emission stoves, etc., have not been included. A more detailed analysis of emissions from different types of domestic biofuel use can be found in Akagi et al. (2011), albeit without the benefit of the numerous papers that have been published on these emissions in the last decade. A special review on this issue would be desirable, but is beyond the scope of this paper.

In contrast to gaseous compounds, which are chemically well defined, aerosols are complex mixtures of organic and inorganic species and comprise particles across a wide range of sizes. This affects in particular the measurements of organic aerosol, black/elemental carbon, and size fractionated aerosol mass. Organic aerosol is usually measured either by a variety of thermochemical or mass spectrometric methods, both of which may have positive and negative artefacts, for which different authors have applied different corrections. Since some techniques report the result as organic aerosol mass and others as organic carbon mass concentrations, a conversion had to be applied. To convert between organic carbon and organic matter (OM), a default OM/OC mass ratio of 1.6 is used in the absence of specific information. This value is based on the data from fresh biomass smoke aerosol in the literature (Turpin and Lim, 2001; Aiken et al., 2008; Yokelson et al., 2009; Takahama et al., 2011; Kostenidou et al., 2013; Brito et al., 2014; Collier et al., 2016; Fang et al., 2017; Tkacik et al., 2017; Ahern et al., 2019; Lim et al., 2019). Where only O/C ratios were given, they were converted to OM/OC ratios using the relationship given in Aiken et al. (2008).

Black carbon (BC) and elemental carbon (EC) are an even more problematic category. Various definitions for these species have been used (Andreae and Gelencsér, 2006), but most commonly BC refers to carbon with specific optical properties (light absorption) and is measured by optical techniques, whereas EC is defined by its chemical properties and determined by a variety of thermochemical methods. Not all authors, however, adhere to these definitions, and soot, EC, and BC are often used interchangeably. Unfortunately, while some techniques have been shown to have less bias than others (Li et al., 2019b), there is no general answer as to which technique is best, and which property, optical or chemical, is more representative. In view of the lack of a better alternative, both BC and EC data have been merged in the "BC" category here.

The size distribution of biomass smoke aerosols is a continuum ranging from tens of nanometers to millimeters (Reid et al., 2005), with most of the mass present in a mode at a few hundred nanometers. Mass concentration measurements are typically reported as PM_1, $PM_{2.5}$, PM_{10}, or TPM, referring to the size ranges below 1, 2.5, and 10 µm, and total mass, respectively. For convenience, data reported as PM_1 and $PM_{2.5}$ have been grouped together in the $PM_{2.5}$ category, which in view of the typical BB aerosol size distribution is not expected to result in significant bias. The same applies to the PM_{10} and TPM data, which were grouped together in the TPM category.

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).

Another problematic "species" is the total concentration of non-methane organic gases (NMOG), also referred to as volatile organic compounds (VOCs). The diverse methods used for these compounds measure different sets of NMOG, which in some instances may quite incomplete. In general, the more recent studies from the last 5-7 years are much more comprehensive and show that the early studies were severely underestimating the amounts of NMOG emitted. Regrettably, these techniques have been so far used mostly in lab studies, and could therefore not be considered for the combustion category emission estimates. To highlight this issue, I have added the NMOG emission factors from the online updates to Akagi et al. (2011) in Tables 1 and 3.

2.2 Definitions

In the literature, emission information is generally found as either emission ratios (ER) or emission factors (EF). Strictly speaking, most data presented as "emission ratios" are actually enhancement ratios (EnR), often also referred to as normalized excess mixing ratios (NEMR; Akagi et al., 2011). They are defined as the ratio of the excess concentration (mixing ratio) of the species of interest in the plume, (ΔX), to the excess concentration of a reference species, e.g., carbon monoxide (ΔCO),

$$EnR_{X/CO} = \frac{\Delta X}{\Delta CO} = \frac{(X)_{plume} - (X)_{backgr}}{(CO)_{plume} - (CO)_{backgr}}$$

where Δ stands for the difference between the concentrations in the plume and in the background atmosphere. Because of its abundance in fire emissions and its relatively low ambient background concentration, CO is most commonly used as reference species, but other gases, such as carbon dioxide (CO_2), methane (CH_4), or acetonitrile have also been used. The use of CO_2 can introduce large errors because it also has strong surface sources and sinks, which can lead to erroneous estimates of the background concentration, as discussed in detail in Yokelson et al. (2013a). A statistical method using multiple fire tracers (Mixed Effects Regression Emission Technique, MERET), which can resolve the problems associated with variable CO_2 background concentrations, has recently been developed (Chatfield et al., 2019).

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An enhancement ratio can be interpreted as an emission ratio when it is assured that the concentrations of both species X and the reference species have not been affected by chemical production or loss since the emission, and that both concentrations have changed proportionally during dilution of the plume with background air. In the case of very long-lived substances, e.g., acetonitrile, EnRs can be very close to ERs even after days, while for reactive compounds, e.g., nitric oxide (NO), significant changes can occur in minutes. At the other extreme, for very rapidly reacting species, it becomes difficult to define an appropriate time after emission at which an EnR can be treated as an effective ER. A good example is the emission of primary organic aerosol mass, where the apparent EnR decreases substantially (by about a factor of two) over the first few minutes to hours as a result of the evaporation of semivolatile compounds during plume dilution (May et al., 2013; Konovalov et al., 2019). Whether the ER at the moment of emission or the EnR after cooling and dilution to typical ambient conditions is the more meaningful value will depend on the intended application. In general, field measurements are likely to represent somewhat more aged conditions (tens of minutes to a few hours), whereas lab measurements often represent very fresh emissions. For further discussion of the advantages and disadvantages of the different reference gases, the effects of flaming vs. smoldering combustion, and ground-based vs. airborne sampling, see A&M2001, Burling et al. (2011), and Akagi et al. (2011).

While the measurement of ERs is relatively straightforward in the field, because it requires only the measurement of the atmospheric concentrations of target and reference species, it is generally desirable to obtain the amount of a species emitted per unit mass of fuel burnt, i.e., the emission factor, EF. For biomass burning, this is usually expressed as the mass of target species X released per mass of dry fuel burnt, in units of g kg^{-1}. This, however, requires knowledge of the mass of fuel burned, which can be measured in the lab, but difficult to obtain in the field. As an alternative, the mass balance method can be used, where the mass of fuel burned is approximated by the sum of carbon contained in the emitted carbon species (CO_2, CO, CH_4, volatile organic compounds [VOC], organic aerosol carbon [OC], and elemental carbon [EC] or black carbon [BC]), divided by the carbon fraction in the fuel. Often, the carbon mass is approximated by the sum of CO_2 and CO, and a default fuel carbon content of 45% is assumed.

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To provide a uniform representation of the various types of data found in the literature in the form most useful to modelers, all emission data was converted to emission factors, in units of g kg^{-1} dry fuel burnt. Where emission factors relative to other fuel mass indicators were given, e.g., the mass of carbon burned or released, I applied appropriate conversion factors, such as the known or assumed carbon content of the fuel. Very frequently in the literature, only EnRs or ERs in units of mol/mol are provided. These can in principle be easily converted to EFs by the following equation:

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$$EF_X = ER_{(X/Y)} \frac{MW_X}{MW_Y} EF_Y$$

where EF_X is the emission factor for species X, $ER_{(X/Y)}$ is the emission ratio of species X relative to the reference species Y, MW_X and MW_Y are the molecular weights of the species X and the reference species Y, and EF_Y is the known or assumed emission factor of the reference species (often CO or CH_4). Since the value of EF_Y is often not known for a specific study, the mean EF_Y for the appropriate type of fire (forest, savanna, etc.) was applied to derive an estimate of EF_X.

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2.3 Estimates where no data are available

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For some combinations of fire type and emitted species, no suitable field data is available to provide a basis for estimating EFs. Where possible, I have used appropriate methods to derive estimates based on other information, shown in italic font in Table 1. For each species, the estimation method is given in column EM. For species predominantly emitted during smoldering combustion, e.g., most VOCs, I have based the estimate on the assumption that their emission factors for the various fire categories are proportional to those of CO for the same categories. The estimate was then obtained by calculating the mean of the ratios EF_X/EF_{CO} for the fire categories with available data and multiplying this mean ratio by the EF_{CO} of the fire category for which an estimate was needed (labeled CO in column EM). Where no suitable ratios ER_X/ER_{CO} were available from field studies, the lab ratio was used instead (labeled LV). For some species containing heteroelements (N_2O, SO_2, DMS, and HCl), the mean of the ERs from fire categories with available data, weighted by the amounts of biomass globally burned in those categories, was used (labeled AV). Subjective "best estimates" are labeled BE. 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.

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3 Results and discussion

3.1 Combustion process and pyrogenic emissions

Our fundamental understanding of the biomass combustion process remains unchanged since the 1990s, as reviewed in A&M2001 and other papers (Lobert and Warnatz, 1993; Yokelson et al., 1996; Yokelson et al., 1997; Akagi et al., 2011), and will thus be summarized here only briefly. As the flaming or glowing front of a fire moves towards the uncombusted fuel, the fuel is heated by radiative and sensible heat transfer, leading first to evaporation of water and other volatiles, then to pyrolytic decomposition and the release of volatile and semivolatile (tar) decomposition products (Collard and Blin, 2014). When this released mixture ignites, flame chemistry sets in, which breaks down the more complex pyrolysis compounds to small molecules and radicals, but also produces new larger molecules by radical chemistry, such as alkynes, polycyclic aromatic hydrocarbons (PAH), soot, and organohalides. 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, in which a large fraction of the fuel carbon is released as CO. In a typical vegetation fire, all these

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Depending on the vegetation type and burning conditions, the relative amounts of fuel consumed by flaming and smoldering combustion can vary considerably. Dry grassland fires, for example, are dominated by flaming combustion and a rapid passage of the fire front, with little residual smoldering. Forest fires, on the other hand, especially those in fuels with relatively high fuel moisture and large diameters, have a long phase of residual smoldering combustion (RSC), during which larger-diameter fuels are consumed over time spans of up to several days (Ward and Hardy, 1991; Ward et al., 1992; Yokelson et al., 1997; Bertschi et al., 2003; Hao and Babbitt, 2007; Burling et al., 2011; Akagi et al., 2013; Geron and Hays, 2013; Urbanski, 2014; Reisen et al., 2018). The smoldering mode of combustion can become dominant in peat fires, which often proceed without a flaming phase and below ground (Bertschi et al., 2003; Stockwell et al., 2016b).

Since the rate of heat release during RSC is relatively low and much of it occurs during nighttime, the resulting emissions tend to accumulate close to ground in the boundary layer. The nighttime emissions are confined in a nocturnal boundary layer, often less than 100 m thick, where the fire-emitted CO_2 becomes mixed with CO_2 from biological respiration. This presents serious problems for measuring accurate and representative fire-integrated emission factors for fires where RSC emissions are important (Bertschi et al., 2003). Ground-based studies during the RSC phase can obtain EFs of trace species, but these are difficult to relate to the corresponding amount of fuel burned. Aircraft studies have trouble measuring the RSC component of these emissions, as they are not lofted in the form of discrete plumes to aircraft altitudes, but only mixed upward during daytime convection (or fire blow-ups) where they get distorted by mixing in the ambient atmosphere (Guyon et al., 2005). The mixing of biogenic and pyrogenic CO_2 in fire plumes that entrain such boundary layer air into a deeper mixed layer present serious problems for deriving fire-integrated ERs and EFs from aircraft measurements (Yokelson et al., 2013a)

Because the flaming phase is characterized by CO_2 being the dominant combustion product by far, while the smoldering phase yields relatively large amounts of CO, the MCE has been established over the last two decades as the key metric representing the relative role of flaming vs. smoldering combustion in vegetation fires, spanning a range of 0.77 in peat fires to 0.98 in some grassland fires (see Supplement). Mean MCE values for the different combustion categories are presented in Table 1.

Since the MCE was introduced by Ward and Radke (1993), numerous papers have used this metric and have shown significant negative correlations for many trace gases between emission factors and MCE, especially for the various VOCs that are emitted predominantly during smoldering combustion (e.g., Korontzi et al., 2003; Yokelson et al., 2003; Yokelson et al., 2008; Soares Neto et al., 2009; Urbanski et al., 2009; Burling et al., 2011; Urbanski, 2013; Yokelson et al., 2013b; Liu et al., 2014; Urbanski, 2014; Collier et al., 2016; Coffey et al., 2017; Fortner et al., 2018; Hodgson et al., 2018; Reisen et al., 2018; Jen et al., 2019). However, the correlation slopes between EFs and MCE vary considerably between studies in different fuels and burning environments, so that a global parameterization of all EFs based on observed or modeled MCE remains problematic. 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

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on the average values from 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. The limitations in correlation between EFs and MCE have been noted previously (Yokelson et al., 1997; Bertschi et al., 2003; Burling et al., 2011; Urbanski, 2014). In the case of ethene, the correlation using all data points is not significant ($R^2 = 0.07$). However, when only the data from open vegetation fires are included (and after removing three outliers), the correlation improves to an R^2 of 0.27. For ethane, the correlation coefficient is $R^2 = 0.38$ for all data, but does not improve substantially by removing the peat fire data. These results suggest that the level of aggregation at which MCE is useful as a meaningful, but rough predictor of EFs for at least some species is yet to be determined. This approach is not pursued further here, but the data in the original studies listed in the supplement can be used by investigators to derive such relationships for specific compounds and combustion types of interest. An interesting and novel approach to generalizing VOC emissions is provided by Sekimoto et al. (2018), who showed that most of the variability in VOC emissions measured in a lab study using a wide variety of fuels was explained by just two factors, related to low and high temperature pyrolysis.

Using MCE as a predictor variable may be an alternative to providing separate EFs for smoldering and flaming combustion, which has been frequently requested by the modeling community, but for which there is still not enough data to provide robust estimates, as we had already remarked previously in A&M2001. However, once vegetation fire models are able to provide estimates of the contribution of flaming and smoldering combustion from a given fire, the resulting MCE could be predicted. This could then form the basis of a more fire-specific prediction of trace gas and aerosol emissions based on MCE correlations. An alternative approach was proposed by Hoffa et al. (1999) and further developed by Korontzi et al. (2003), who showed a correlation between vegetation greenness and MCE, which allowed the prediction of seasonally-dependent emissions from African savanna fires (Ito and Penner, 2004; Korontzi et al., 2004; Korontzi, 2005). In view of the limitations seen with regard to more general parametrizations, it appears that for now one should keep using the category-average EFs, but be aware they can vary considerably from region to region and from fire to fire.

3.2 Emission factors for chemical species from the various combustion categories

In Table 1, I present the updated estimates of emission factors for the combustion categories, savanna/grassland, tropical forest, temperate forest, boreal forest, peat fires, open agricultural waste burning (in the fields), biofuels (excluding dung), dung cakes, charcoal making, charcoal burning, and garbage burning. As more data have become available, it is now possible to split the extratropical forest category into temperate and boreal forest. The transition between these two types is not always clear, but in general, I have followed the authors' choice of category; where this was not possible I have taken a latitude of 60 ºN as boundary.

The large number of studies on residential biomass burning, which have been published in the last two decades, has made it possible to separate dung cakes from the other biofuels, such as fuel wood and agricultural residues. As mentioned above, I only included studies that used fireplaces and traditional or simple "improved" stoves, as they are used in developing countries, and not modern appliances, such as automated pellet stoves.

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Deleted: For information purposes, I also include a column for the results of laboratory studies. The averages in this column can only be seen as general indications, since all types of fuels and burning methods are included, but the original data and references are provided in the supplement for readers interested in the details. As more data have become available, it is now possible to split the extratropical forest category into temperate and boreal forest. The transition between these two types is not always clear, but in general, I have followed the authors' choice of category; where this was not possible I have taken a latitude of 60 ºN as boundary.

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The publication of a few papers that provided emissions data for open garbage burning, still quite prevalent in many countries and a serious source of pollution especially in urban areas (Wiedinmyer et al., 2014), has made it possible to provide EFs for this category.

Obviously, the categories used here are still quite highly aggregated, but they correspond closely to the fire types used in many global modeling studies, such as those involved in the Fire Modeling Intercomparison Project (FireMIP) (Li et al., 2019a) and in model- or satellite-based emission inventories (Wiedinmyer et al., 2011; Kaiser et al., 2012; Ichoku and Ellison, 2014; Darmenov and da Silva, 2015; van der Werf et al., 2017). Should a reader require less highly aggregated data, they can use the Supplement to split the data into subcategories or even use the supplemental references to get back to the original literature. 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).

For information purposes, I also include a column summarizing the results of laboratory studies. 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 statistics presented here. However, the original data and references are provided in the Supplement for readers interested in the details.

As in A&M2001 and in Akagi et al. (2011), the amount of information for any given combination of species and fire category varies greatly - for some combinations we have no measurements at all and for others there are as many as 50 values. Accordingly, the uncertainty of the estimates is also highly variable. In Table 1, I am using the same convention as in A&M2001 to represent the uncertainty: When three or more values (based on independent references) are available for a given table cell, the results are given as means and standard deviations (x ± s). In the case of two available measurements, they are given as a range, and where only a single measurement is available, it is given without an uncertainty estimate. For single measurements, it can usually be assumed that the uncertainty is no less than a factor of three.

In spite of the fact that this paper is based on data from over 370 publications, rather than the 130 papers that formed the basis for A&M2001, Table 1 shows that there are still many species for which there are little or no field data available. For example, there are still no field measurements of the emission factors for the alkyl amines, which have recently become implicated in aerosol nucleation and new particle formation (Smith et al., 2010; Almeida et al., 2013; Kürten et al., 2014). In view of the importance of the number concentrations of aerosol particles, especially cloud condensation nuclei, for climate change, it is unfortunate that there have only been a few additional measurements of their emission factors in the last two decades. 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 measurements suggests that the ratio of excess particle number concentration (ΔCN or ΔCCN) to ΔCO 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.

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Another climate-relevant component, for which we have no field emission data at this time, is brown carbon (BrC) (Andreae and Gelencsér, 2006), which has been shown to account for about half of the aerosol light absorption by biomass smoke at 401 nm (Selimovic et al., 2019) and 25-45 % at 550 nm (Tian et al., 2019). 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).

Regarding the role of vegetation fires in the global carbon cycle, the most problematic uncertainty pertains to the emission factors of CO_2 and CO from forest fires, which is surprising in view of the many available estimates. This uncertainty stems from the inadequate knowledge of the contribution from RSC, which has already been referred to above, and which may significantly contribute to large mismatches between bottom-up predictions of CO emissions and remote-sensing measurements from satellite (Pechony et al., 2013; Deeter et al., 2016). A representative measurement of fire-average $\Delta CO/\Delta CO_2$ emission ratios from large forest fires is very difficult if not impossible, as ground-based measurements in such violent fires are not possible and aircraft measurements are prone to undersampling the smoldering emissions, especially the contributions from RSC. The uncertainty regarding the $\Delta CO/\Delta CO_2$ emission ratio also seriously hampers our ability to separate the influence of the emissions from deforestation burning from those of biological carbon fluxes in regional carbon budgets (Andreae et al., 2012). For example, the uncertainty of the $\Delta CO/\Delta CO_2$ ratios of tropical forest burning is large enough that it can even change the sign of the net carbon flux between the Amazon forest and the atmosphere (Gatti et al., 2014). A novel multi-tracer statistical technique (MERET; Chatfield et al., 2019) may be able to provide improved estimates of the CO ERs and EFs from such fires.

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Figure 2 presents a comparison between selected EFs from this study with those published in Akagi et al. (2011) in the form of ratios between the EFs from these studies. For this comparison, I have selected species that are of major climatic or chemical significance or are important BB tracers, and for which there are enough data to allow a meaningful comparison. Data are presented for the combustion types with the largest total global emissions, i.e., savanna/grassland, tropical, temperate, and boreal fires, and biofuel use. In the case of biofuel use, the comparison is made with Akagi et al.'s "open cooking" category, because its MCE shows good agreement with that for the "biofuel use" category in this paper. Figure 2 shows close agreement for the main carbon species CO_2 and CO as well as for MCE, suggesting that both species capture comparable combustion conditions. For most other species, the EF ratios fall within a factor of two, with no obvious systematic shift for either the individual species or for the combustion types. A slight exception are the EFs for savanna/grassland, which tend to be somewhat higher in the present study. In one case (isoprene) this is the result of higher values from an individual study, i.e., the lab-adjusted-to-field EFs from Stockwell et al. (2015), but generally the differences appear to be the result of including a larger set of studies from this category in the present study. The lower EFs for glycolaldehyde in this study are the result of corrections made by the Yokelson group to their data based on improved spectral data (see https://www.atmos-chem-phys-discuss.net/12/C11864/2013/acpd-12-C11864-2013.pdf), which have been incorporated here and in the online updates to Akagi

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et al. (2011), but for consistency the values from the original paper were used for Fig. 2. The largest and systematic difference is seen for the NMOG category, where the values from Akagi et al. (2011) are as much as a factor of 10 higher. This is largely due to differences in the analytical techniques used in the original studies. Most of the older studies, especially in field campaigns, were measuring only a very limited subset of NMOG (e.g., non-methane hydrocarbons), whereas Akagi et al. in the original paper and in the subsequent updates used techniques that measured practically all NMOG, including unidentified species. To address this issue, I am also including the corresponding values from the online updates to Akagi et al. (2011) in Table 1.

3.3 Emissions from global biomass burning

In 2001, we estimated the total amount of biomass burned by all combustion types to be 8.6 Pg dry matter annually with an uncertainty of ±50% (A&M2001). This estimate was based on bottom-up inventories and had not yet benefitted from remote-sensing detection and quantification of fires. At present, there are several operational fire detection and emission estimation products based on remote sensing. Three of them (for example) use an approach based on burnt area and hotspot detection: Fire INventory from NCAR (FINN; Wiedinmyer et al., 2011), Fire Locating and Modeling of Burning Emissions (FLAMBE; Reid et al., 2009), and Global Fire Emissions Database (GFED; van der Werf et al., 2017). The other three products are based on fire radiative power (FRP): Quick Fire Emission Dataset (QFED; Darmenov and da Silva, 2015), Global Fire Assimilation System (GFAS; Kaiser et al., 2012), and Fire Energetics and Emissions Research (FEER; Ichoku and Ellison, 2014). The amounts of biomass burned annually in open fires estimated by these systems still spans a wide range, from 4.3 Pg (GFAS) to 11.6 Pg (FLAMBE) (for the FRP-based products, which do not use biomass burnt in their calculations, the biomass estimate was based on the stated emission of carbon compounds and an assumed carbon fraction of 45 % in the biomass).

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For domestic biofuel use, there are three recent global estimates: 2.1 Pg a^{-1} (Fernandes et al., 2007), 2.5 Pg a^{-1} [S. J. Smith, personal communication, 2019, based on the Community Emissions Data System (CEDS) model (Hoesly et al., 2018)] and 2.3 Pg a^{-1} [Z. Klimont, personal communication, 2019, based on the methodology in Klimont et al. (2017)]. These recent estimates are all somewhat lower than those of A&M2001 (2.9 Tg a^{-1}) and Yevich and Logan (2003) (3.1 Tg a^{-1}). For charcoal burning, I am also using the estimate of 53 Tg a^{-1} given for 2014 by FAO (2015), and for charcoal making I am assuming a 25% yield of charcoal relative to dry wood (Yevich and Logan, 2003).

Combining these estimates of open and domestic burning yields a mean estimate of 8.8 Pg (with a range of 6.4 to 14.1 Pg) dry biomass burned annually. Interestingly, this is almost identical to the values given in A&M2001: 8.6 Pg a^{-1}, with an estimated range of 4.3 to 12.9 Pg a^{-1}. Table 2 summarizes these emission estimates. For the various categories of open burning, the satellite-derived emission estimates vary greatly, in some cases by an order of magnitude. Differences in the definitions of the burning categories between the different retrieval algorithms, differing ability to detect small fires, and the fundamental difference between the burnt-area and FRP-based techniques may all play a role here.

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In Table 3, I use the average of the available estimates from the different inventories shown in Table 2 as activity estimates for the combustion categories to derive emission values for major species emitted from biomass burning. For comparison, the last column in Table 3 shows the global total emissions estimated in A&M2001. The totals of the major emitted carbon species and many minor species remain fairly close to those in our previous assessment. Given the large number of measurements for the emission factors for the major species, CO_2, CO, and CH_4, the standard error of the mean is much smaller than the standard deviation, and thus the relative uncertainties of the mean for these emission factors are quite small, 1-3% for CO_2, 4-9% for CO, and 6-18% for CH_4 from the major burning categories savanna, forests, and biofuel. Consequently, the global emission uncertainties for these species are completely dominated by the large uncertainties in the activity estimates.

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The best independent "reality check" for these emissions may still come from the inverse modeling of the CO budget. This species is the most appropriate for such a comparison, because its emission factors are well constrained, biomass burning is a large fraction of all global sources, and there is a large body of measurements both from ground stations and remote sensing. Estimates of CO emissions from the various inversion models range from 190 to 560 Tg a^{-1} from biofuel burning and 360 to 610 Tg a^{-1} from open burning for the years around 2000 (Park et al., 2015, and references therein). The model of Park et al. (2015), which uses a joint inversion of CO concentrations and oxygen isotopic composition and therefore is likely to be the most reliable in separating the different source types, predicts CO emissions of 380 to 610 Tg a^{-1} from open burning, 400 to 520 Tg a^{-1} from biofuel use, and 780 to 1130 Tg a^{-1} for all biomass burning. Using the EFs from Table 1 and the activity estimates from Table 2, we obtain a range of 390 to 1210 Tg a^{-1} for the CO emissions from open burning, in reasonable agreement with the inverse results. The range of biofuel CO emissions estimated from Tables 1 and 2 is only 181-196 Tg a^{-1}, accounting for less than one-half of the inverse estimate. This suggests either that the amount of biofuel use is significantly underestimated in present bottom-up budgets, or that the inversions attribute some of the open burning inaccurately to biofuel use. This could likely be the case for agricultural burning, which uses similar fuels and takes place in similar regions as biofuel use. The inverse analyses may also be useful to indicate unlikely estimates based on remote-sensing techniques. For example, the burning of 8750 Tg dm in tropical forests estimated by FLAMBE, combined with the corresponding EF_{CO} (105 g kg^{-1}) would produce CO emissions of 900 Tg a^{-1} from this biome alone, well above the range of inverse CO emission estimates for all open burning (see also the comments by Reviewer 1, https://doi.org/10.5194/acp-2019-303-RC1).

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Major differences between the present emission estimates and A&M2001 are seen for the oxygenated volatile organic compounds and for HCN (as already noted in Akagi et al., 2011), which all are significantly greater in the present assessment than in A&M2001. This is due to the large number of new and more accurate emission factor measurements for these compounds, which have been made possible by improvements in analytical techniques since the 1990s.

4 Conclusions

We are left with the somewhat frustrating conclusion that, in spite of the great progress in emission factor measurements and detection and quantification of fires, the overall uncertainty of biomass burning emissions has not decreased significantly for most substances since our analysis of almost twenty years ago. Evidently, there is a great need for improved accuracy in the activity estimates, for open burning and especially for biofuel use. For open burning, coordinated regional CO studies in regions and at times of high biomass burning activity, including both FRP and burnt-area based remote sensing approaches as well as inversions, may be a way to resolve discrepancies and improve accuracy. This would be of great benefit for testing and improving fire emission models, which also give quite divergent results and have difficulties in capturing interannual variations and temporal trends (Li et al., 2019a). The modelled estimates of carbon emitted from open burning in the nine models participating in the FireMIP project spans from 1.0 to 4.9 Pg a^{-1} (Li et al., 2019a).

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With regard to emission factors, Table 1 can serve as a guide to prioritizing future research activities. Photochemically active species and toxic compounds for which there are only a few measurements from important fire types deserve more intense study. An example is the emission of PAHs, where we have only one study from boreal fires and none at all from tropical forest fires. Given the toxicity of these compounds and the increasing exposure of populations in these regions to biomass smoke as a result of climate change and population growth, this seems an important knowledge gap. Another example are the emissions of semivolatile and intermediate-volatile compounds (I/SVOCs), which are important in the context of organic aerosol from biomass burning, but for which at this time only laboratory measurements are available (Hatch et al., 2018). I have already referred to the lack of field measurements of alkyl amine emissions, which may be of importance for new particle formation. In view of the grave health risk associated with aerosol particles (see, e.g., Lelieveld et al., 2019, and references therein) and the growing exposure to wildfire smoke in areas like the western U.S.A., the accuracy and fire condition dependence of PM emissions need to be improved. Emphasis should be on field measurements under a variety of representative conditions, to represent the influence of parameters like fuel moisture and fire weather. 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.

A spreadsheet containing Table 1, the data on which the averages in Table 1 are based, and the corresponding references is available at http://dx.doi.org/10.17617/3.26 . This spreadsheet will be updated periodically.

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