

Referee for

Journal: ACP

Title: Fossil fuel combustion and biomass burning sources of global black carbon

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General comments

This manuscript represents an extensive approach to compare black carbon (BC) fractions of biomass burning from observations all across the northern hemisphere to GEOS-Chem simulations. The authors further conduct four sets of experiments to test uncertainties associated with the modelling of BC biofuel emissions, BC hygroscopicity, BC ageing, and BC size-resolved scavenging.

The overall presentation, language, and figures are of high quality. The study is within the scope of ACP and presents novel data. However, the presentation of the numerical data is currently not up to the desired accuracy. Specifically, it is not always clear if presented percentage numbers are absolute or relative, and uncertainty values are mostly missing or not labeled sufficiently.

The used methods and assumptions are mostly clear and valid, yet some conclusions appear vague and a little too uncritical. I suggest to revise the manuscript and focus on accuracy and the specific comments given below.

Specific comments

Title: Use a more declarative title. A suggestion: **Fossil fuel combustion and biomass burning sources of global black carbon from GEOS-Chem for 2007-2013 compared to observations from 2002-2014**

f_{bb} values: Is there a reason that no error values or standard deviations are given? In Table S1 all presented values include some form of error. I am writing "some form" because it is not clear what the error value in the table means statistically. What is the confidence interval?. Also, sometimes it remains unclear if the values refer to atmospheric or deposition values. For example in L11-17¹. Please be more specific. Are the presented numbers for atmosphere or deposition?

Page 1

- L12: Be more specific. What do you mean by "comparable contribution"? Give a number or range.
- L17: Be more specific. What do you mean with "reduction of discrepancies"? Discrepancies between what?
- L18: Be more specific. Which "discrepancies" do you mean?
- L23: Explain what the numbers in the brackets mean. What does this range refer to?
- L24-25: This sentence is not very clear. What does increase? Is it concentration, ratios, size or something else? Does "the former" refer to fossil fuels? Replace "the former" with a more specific description.
- L26-27: Is this finding novel? Please put this, your final conclusion of the abstract, in a bigger context.
- Abstract in general: Please write a sentence that explains your 4 experiments, in brief and put it before "We find" in L17.

¹ L refers to line numbers

Page 2

- L14-16: You write: "Fossil fuel combustion often has an anthropogenic origin". When does it not have an anthropogenic origin? Please change this sentence.
- L23-25: The studies you refer to all looked at local emissions only. Besides that, their assumption is basically the conclusion of your study. Can you reflect more on this in the discussion part of your study?

Page 3

- L5-15: The model description is very very short. The study would benefit from more detail. Explain why you picked this model and why you think that exactly this set-up (i.e. microphysics scheme, particle size resolution, model resolution, and emission inventories) is best suitable to run the four different tests you laid out before.
- L5-6: Please give a reference
- L11: Discuss in your discussion section what it means for your analysis if you compare observations from 2002-2014 to emission inventories from 2007 publications (in reality the emission information is older than that) or 2017 (in case of Asia).
- L12: Discuss here how you account for potential double counting of open fire emissions, included as agricultural burning in emission inventories and as open fires in GFED data.
- L15: briefly explain what updates you applied to the wet deposition scheme of Liu et al., 2001.
- L16-23: This part appears to be results, rather than model description. Please move it.
- L17: Explain why you only simulate 2007-2009 for Fig 1S.
- L19: What do you mean by "preferably" ? Can you quantify it?
- L24-25:
 - Please explain what the criteria was for picking the 41 sites. There is more data available for the observational period you picked (2002-2014) (e.g., (Zotter et al., 2014). You even cite at least one study, who's data you have not included in your analysis, despite falling in that time period (Winiger et al., 2017). Also, is it true that there are only data available from the Northern hemisphere? How confident can you be regarding simulations of the Southern hemisphere, if there is no observational data available?
 - Why are only the atmospheric samples mentioned in Table S1, when you also discuss the (snow) deposition measurements in your study?
 - Please indicate the measurement technique in Table S1, since there can be considerable differences in BC values between different observational methods (i.e. protocols).
 - Please indicate what the error values for f_{bb} mean in Table S1.
 - Please fix the altitude values in Table S1. Some are 0, e.g., The station in Barrow is at 11 meters above sea level.
 - The reference for Szidat et al., 2004 (in Table S1) is missing in the SI references.
- L25-27: "Generally" is not really accurate. It depends largely on the season. There are cities (in Europe and Asia) where considerable (>40%) fbb values are observed in winter (Zotter et al., 2014, Bikkina et al., 2019)

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- L9-L13: This seems to be an important point. The difference in the used methods is bigger than the uncertainties you found with your four experiments. Could you extend on this point? Additionally, add information of the used methods to Table S1.
- L10: I would be careful with highlighting single methods. Water extraction is also known to have considerable drawbacks (Azeem et al., 2017). A method for EC extraction (and BC measurements in general, actually) that satisfies all needs is currently nonexistent.
- L15: the factor 2 in Fig.1 (a) is not very clear. Some values (including error) go beyond the factor 2. Could you give these numbers in a table in the Supplementary Information (SI) ?
- L16-22: Please rewrite this. Within the context of these 3 sentences you write 3 times that "the low bias of f_{bb} in [region X] is due to underestimation of biofuel combustion" in one form or another.
- L18: What do you mean by "current emission inventories"? Can you give references? You use an emission inventory with emissions older than 2007. Is that also a current emission inventory?
- L20: While there is little doubt that it is a great study, Qi et al., 2017c is hardly a reference for the sentence that precedes it. It is also not very specific what "large" means. Based on AMAP 2015 (see e.g., their Figure 11.1) European contribution to the BC burden in the Arctic is smaller than that of Russia or East and South Asia (and some other regions, based on the used model). Please consider rewriting this.
- L23: Where can we see the "factor of 2"? This is a general issue in the manuscript. These numbers ("factor of [X]") are presented in many places in the narrative but it is hard to follow. Would it be possible to provide a table in the SI?
- L23: Is there a reason you discuss possible reasons for underestimation of f_{bb} in the Arctic, but decide to discuss overestimates of f_{bb} in North America somewhere else, in Section 4.2.1.?
- L24-25: What makes you so certain that the large variations of f_{bb} values is "due to the coarse horizontal and vertical resolutions."? Why shouldn't this be the case for all your other modelled sites as well? North-America, for example, has quite some orography as well.
- L30: You write that the model captures the spatial and temporal variations of f_{bb} in BC deposition in this region [Himalayan-Tibetan plateau] and that "GEOS-Chem reproduces the average f_{bb} in snow perfectly". How does that fit with the previous comment (L24-25)? Could you discuss this here, instead of section 4.2.1? Your analysis should also be more critical towards the model. What makes you think that the model simulates precipitation well, but less so atmospheric concentrations? Especially since many (if not all) other models struggle with orography (this GEOS-Chem run is at $4^\circ \times 5^\circ$ resolution and results discussed are in the Himalaya's !) and deposition schemes (e.g. Textor et al., 2006, Tegen et al., 2019).
- L31-L2 page 5: This sentence is good, because it gives the %-values that makes it possible to follow what you mean with "factor 2.3".
- f_{bb} values in general: Please give and define uncertainties for your values or justify why you are not doing so.

Page 5

- L7-9: You are using annual fossil fuel and biofuel emissions. Why would you expect anything else than "seasonal variation is relatively flat"? Or did you apply some sort of seasonality? This is not clear from reading the model description. Where (daily

resolved) open fires (from GFED) too weak in seasonality to show a difference? Or do you attribute this to model resolution entirely?

- L11: "probably underestimated" is a very vague formulation. Please be more accurate and specific.
- L12: It is unclear what you mean with "The similar magnitudes". Similar to what? Consequently, your conclusion of summer values (L12-14) "atmospheric fbb is largely determined by local emissions" is incomprehensible and appears speculative.
- L14: please specify "other seasons".
- L14: please specify if you mean model or observational "atmospheric fbb values".
- L9-15: This entire section appears vague and speculative. Please revise it.
- L15-18: Please give references and write that these are observational values.
- L24: "along the Mustang valley and Langtang valley." To me, it is not clear where these valleys are located. Can you show a figure in the SI?
- L24-27: Would you expect any such local effects with the model resolution you are using? Is your model fit for purpose (coarse resolution and no annual resolved emissions) to compare seasonality's in such a terrain? Your conclusion (L29-30) seems not well supported, given the previously mentioned model limitations.
- L31: What do you mean with "no statistically significant differences"? Since you reference the figures here, you could write "no big differences". Better even, give a value for the variations: "no big differences (+/- XX %)".
- L32: which "four sites" do you mean?

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- L4-5: What kind of solid fuel would be mostly used, i.e. what are the limited effects on fbb in the atmosphere? According to Mouteva et al., 2017, fraction modern (BC) doesn't change much throughout the year.
- L5-7: I don't think this sentence ("which is proved to have limited effects on fbb (Mouteva et al., 2017)") makes much sense. fbb does not vary much throughout the year. Why would one expect different values during strong winter inversions?
- L7-9: Which region are you referring to in this sentence? "Modeled fbb in the atmosphere is much higher than the fbb values of local emissions, suggesting a large regional effect on fbb in this region." I assume it is Salt Lake City, but it is not clear. And where do the numbers come from in "The model overestimates fbb in all seasons by a factor of 2–4."
- L9-10: How does "This mismatch of model representation and observations partly explain[s] the large positive bias of fbb."? 'The model overestimates fbb' is not exactly an explanation of why we see this mismatch.
- L11-12: What kind of regional effect do you mean? What if emissions are wrong? This is an oversimplified analysis.
- L13-14: Which part in the "Comparison of fbb in local emissions and in the atmosphere" suggests that "fbb at MCOH is largely affected by long-range transport..."? Figure 2(g) does not even show if there are local emissions or not.
- L14-15: Please rewrite to reflect that both MOCH and SINH have relatively high fbb (~50%), very similar to each other.
- L16-30: Please state more clearly when you speak of observation or model.
- L23-26: It is unclear where the numbers here come from. Also, the highest fbb in the Northern hemisphere is found in African countries, according to your Fig. 3.
- L27-30: Two things: 1. It appears that Antarctica has strong seasonal fbb variation in atmosphere and deposition as well. However, it is a bit hard to quantify this from the

figure alone. 2. the "large contribution from open fire emissions" can not be seen in Figs S6 and S7, which show fbb alone. We would need to see BC concentrations before we can conclude that high biomass and biofuel emissions are responsible for the seasonality at the poles.

- L29-30: Two things: 1. To write that continental/regional (modelled) data is consistent with observations, when you have mostly one site per region (Fig 2) is a bit of a stretch. Please reflect this limitation more accurately or include more data, e.g., with a table in the SI. 2. Technically you do have one site for Europe, but there (Abisko) is a clear fbb seasonality.

Page 7

- L3-8: I suggest moving this section to the introduction.
- L8: Could you give additional information why you chose to double biofuel emissions from domestic heating north of 45°N? Why doubling? why domestic heating? And why N of 45°N? There is, for example, evidence that open biomass burning might also be underestimated in the Arctic (Konovalov et al., 2018).
- L11-12: Could you reformulate this? It is not clear to me what you mean by "the model discrepancies... are reduced from -XY% to -YZ%"
- L20: What are the base run conditions for % of hydrophilic fresh biomass burning BC?
- L21-24: It is not clear what the percentages refer to exactly. If you say that the effect of this experiment lowers atmospheric fbb by "up to 11%", do you mean that the absolute fbb value decreases from e.g., 51% to 40% (-11%) or from 51% to ~46% (-11%)? In the second sentence you write that the largest reductions are "-7%". How does 'largest reduction of -7%' stand in relation to 'lowering of up to 11%'? Please revise these sentences and check the rest of the manuscript as well, where such comparisons take place. Again, a table would eliminate such uncertainties.
- L26: Can you quantify "large precipitation"?
- L27-28: It is unclear what you are trying to say with this sentence.
- L29: Which region are you referring to?

Page 8

- L25: What do the values in brackets mean?
- L29-30: Explain why you chose these diameters, since they diverge from the numbers you referred to in L20-21.
- L30: "Size resolved coagulation, condensation, nucleation and cloud processing" are implemented in TOMAS? Please specify or give a reference.
- L31-L1 page 9: That is an interesting find. Can you elaborate more on the reasons that lead to this effect (i.e. the larger decrease of fossil BC)?

Page 9

- L20: "within 30%" is a bit unclear. Please be more specific.
- L28: increase "by 18–23%" is a bit unclear. Please be more specific. Essentially this is the same issue with my comment on L21-24, page 7 (see above).

Page 10

- L8-12: This last paragraph is really disconnected from the rest of the text. Your research doesn't look into mixing states (internal vs external) and lensing. Consider changing it to reflect the scope better.

Figure 1

- Please use more descriptive axes, e.g., "modelled atmospheric BC fraction from biomass burning [%]"
- what standard deviation is shown in the figures?
- Could you show error bars in (b) ?
- consider different colors in (a) like in your previous publications (e.g. Qi et al., 2017)
- consider using different shapes in (b)

Figure 2

- Why do some sites not have emission bars?
- Please use more descriptive axes
- There is observational data for Barrow summers available now, see Winiger et al., 2019.
- Sometimes the standard deviation bars are not visible. Are they missing? Please specify
- Is the model grid for each site $4^\circ \times 5^\circ$? Please specify

Figure 3

- Justify why large parts of Indonesia are missing in your regional analysis.

Figure 4

- Please use more descriptive axes
- include error bars if possible

Figure S2

- Why is the data for 2007-2009, unlike the rest of your study?

Colorscale

- Just a general comment. Consider a gradual color scale for your maps in future work, that doesn't contain rainbow colors. This is friendlier for color blinds and has the co-benefit that b/w prints are better understandable.

Technical corrections

Please consider the following suggestions:

Page 1

- L12: remove: Specifically
- L17: replace "northern than" with "north of"
- L17-19: Redundancy. Remove "in winter" or "in cold season"
- L30: Quantify "large" or remove it.

Page 2

- L2: I assume "BC distribution" refers to atmospheric surface concentrations? Do clarify.
- L5: Remove "In addition"
- L7: Remove "Moreover"
- L8-9: Remove the last sentence as it is redundant.

- L10: replace "separating" with "distinguishing".
- L16: insert "come" between "can" and "from"
- L22: There appears to be a word missing after "Alpine"
- L23: change "compare" to "compared"
- L24-25: change to: "The assumption behind these studies is, that the major..."
- L31: replace "following" with "consecutive"
- L31: remove "so far"
- L32: insert "in terms" between "sources" and "of global BC"

Page 3

- L8: change "3 mn" to "3 nm"
- L9: What does MERRA2 stand for?
- L20: change " NC_Norhest" to " NC_Northeast". Northeast is also misspelled twice in Fig S2 (legend inside right figure).

Page 4

- L3-4: Please give a reference
- L4: change "end member" to "end members"
- L4-5: Please give a reference
- L8-9: Please give a reference
- L9: change "thermal-optical method" to " thermal-optical methods"
- L15: change "GEOS-Chem simulated fbb in the atmosphere agree with observations within a factor of 2" to "GEOS-Chem simulated atmospheric fbb agrees with observations within a factor of 2"
- L24: consider using "horizontal lines" instead of "error bars". The lines show a variation and not actual errors or deviations.

Page 5

- L11: change " to site Abisko " to " to the Abisko site "
- L12: give a reference for the Barrow values.
- L23: "are shown" where?

Page 6

- L3: "area sources" sounds a bit odd. Maybe "non-mobile" is better in this context. Or explain what you mean by it, like they do in Mouteva et al., 2017. (page 9851).
- L3: specify "solid" as "wood and coal".
- L10: Please start a new paragraph for Tokyo data.
- L16: consider changing to " 4.2.2 Spatial variation of modelled fbb"
- L22: change S3 to S4

Page 7

- L9: In the first instance that "Exp." appears, please write what you mean by it.
- L10: change " Specifically " to " As a result "

Page 8

- L25: specify κ
- L26-27: This sentence is redundant. Consider removing it

- change led to led
- Healy et al., 2015 appears to be missing as a reference.

References

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