

Interactive comment on “Black (pyrogenic) carbon in boreal forests: a synthesis of current knowledge and uncertainties” by C. M. Preston and M. W. I. Schmidt

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R = Response. Please also see general Author Comment

Response to Rev. 3 general comments: R-A The manuscript is as long as we felt that it needed to be, and other reviews are of a similar length. Downsizing will be attempted. R-B A lot of effort went into developing the order, although apparently not completely successful. There is a small amount of deliberate recapitulation of key concepts at the beginning of some sections. This was to assist the reader to follow a long paper, and also to assist the reader who might only want to read specific sections. R-C This seems to be a key issue with both reviewers, and is discussed in our general comments. Expansion to the whole world is not so easy as suggested, was never our intention, and

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would be stretching our expertise. We will improve the focus on the boreal, although our discussion goes far beyond highlighting gaps (especially sections 7 and 8). We will try to downsize, but need to supply supporting information appropriate to our interdisciplinary target audience, and to supplement the limited data from boreal regions. **R re Methods:** For comparison of methods, see our general comments about the real world. Also, this is not a specialized comparison of BC methods, and ample references are provided. Also, we commented throughout, on the importance of the choice of method to the results. Comparison of results obtained by different methods will be clarified where necessary. There seems to be misinterpretation of the PAH section (see more detailed comments later). First, there is no suggestion in the MS that the referenced PAH studies or their methods specifically target industrially-produced PAHs. The sources, including forest fires, are inferred from the product distribution. Second, we do not suggest anywhere that this is a method for PyC per se, and this seems to be a misreading. The section on PAHs is included because they constitute a small portion of PyC, but PAH analysis is not a method for BC.

R We put a lot of effort into checking references and proofreading, and “carelessness” is an unfair charge. Of course, a few things were missed, and we appreciate the opportunity to correct these errors. Based on careful study of the comments, the two instances of apparently “relying on one study” are probably Bird et al. (1999) about the the longevity of char produced in tropical savannas, and Knicker et al. (2005a) about 15N NMR of fire-altered SOM. As discussed in more detail later, we stand by these references in the context in which they are cited.

R to recommendation: For the first point; see our general comment. For the second, it appears that the reviewer is reading in biases where there are none.

Specific comments: Line 26: “large gaps in the basic information” are not only limited to the boreal regions but are an issue for all areas in the world that are highly influenced by fire. **R** (p 212, l. 14-15, Abstract) Agreed, but our goal is to highlight knowledge gaps and research needs for boreal regions. At the risk of increased redundancy, “for

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boreal regions” will be added.

Line 29: techniques need to include also physical separation (as mentioned by the authors on p. 7). R (p. 212, l. 15-17, Abstract) This sentence lists the methods used to distinguish charcoal or BC from other soil C. Physical separation can be associated with any of these approaches, but this seems to be too much detail for the abstract.

Line 30: what do the authors mean with “PyC continuum? R (p. 212, l. 18, Abstract) Apostrophes usually suffice to indicate that this is a specialized term that will be defined later, but the sentence will be reworded to “and capture different fractions of the range of thermally altered C structures often described as the PyC “continuum” ” Line 31: here, boreal peatlands are emphasized but line 34 and 35 appear to stress boreal forests - thus, it is not even clear which boreal ecosystem the authors would focus their review on. R (p. 212, l. 19, Abstract) Title will be changed to boreal “regions”, and text clarified in other places as appropriate.

Line 41: the idea of consumption of PyC by subsequent fires is highlighted here but is sadly not really expanded in much detail in the text in terms of what temperature and time would be required to achieve this consumption. R (p. 213, l. 1, Abstract) We would say more in the text (including the use of data from other regions) if this information were available. Indirect evidence is that studies have found lower amounts of BC or charcoal than would be expected from fire frequency and production rates. This specific point will be added to the boreal knowledge gaps (Section 8). It would also be better to say in the abstract that “it has been suggested that the main limitationĚ.”.

Line 53: the Hicke et al. reference does not say that fire is a “driver of ecosystem processes and the C cycle” but that fire affects the C cycle. This is a big difference! R (p. 213, l.12) Is it? Hicke et al. (2003) actually describe fire as a “major disturbance”, not just that it “affects the C cycle”. “Driver” is commonly used in a similar sense in many papers; however, the offending word will be changed to “disturbance”.

Line 76: explain the whole “continuum” aspect better: does that imply that there are no

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distinct groups, yet still a considerable effort is made to distinguish one from the other by different terminologies? R (p. 214, l. 8) This is indeed the problem with BC analysis! An analytical chemist's nightmare. This section will be rewritten to improve clarity.

Line 78: why “graphite”? graphite is highly metamorphosed organic C and to my knowledge, there is no firm evidence that indeed it had originally formed by pyrogenic processes. R (p. 214, l. 10) Graphite is considered as part of the BC continuum, and the most severe oxidation methods leave only graphitic black carbon (GBC). Re graphite, line 78 says “highly resistant graphite formed by geological processes”, which, in agreement with Rev. #3, does not imply a pyrogenic origin, which seems to have been inferred. Certainly, graphite does not originate only from fire-altered biomass - although its formation could be considered a pressure-enhanced pyrolysis. Soot contains highly graphitized structures, and there is some difficulty in absolutely separating true geochemically-formed graphite from some highly graphitized structures produced by combustion, or “proto-graphite” (Haberstroh et al., 2006). The revision will minimize or eliminate the description of PyC forms (l. 76-79) in Section 1, and perhaps even the introduction of the term “continuum”, and save it for Section 2. This should also reduce repetition.

Line 86-89: this paragraph is highly misleading as it implies that this review will be focusing on formation, characteristics, stocks and losses of PyC in boreal regions only. R (p. 214, l. 17-20). These lines appear to have been somewhat misinterpreted (the reviewer has inferred “only”. The first lines of the sentence are about PyC in general, followed by the phrase “the role of PyC in boreal forests [forests to be changed to regions] (C cycle, effects on soil processes)”. As discussed in our general Response, the Introduction and Abstract will be rewritten to clarify why a boreal focus does not require exclusion of all information from other regions; on the contrary, the larger context is a necessity.

Line 131-133: to my knowledge, most analytical techniques that aim to analyse for PyC do not aim to differentiate inorganic C - usually, acid treatment prior to analyses is used

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to remove this fraction but it is usually not specifically quantified or characterized. R (p. 216, l. 3-5?) What is the problem? It is a very fine point to claim that pretreatment to remove inorganic C is not actually part of the BC analysis; on the contrary, most analysts would consider pretreatments an integral part of the process. Also, some researchers do quantify the inorganic C component. Section will be reworded, but information on inorganic C retained.

Line 158-163: I find this section highly confusing (particularly when being referred to before as PyC as a “continuum”) R (p. 216, p. 27) This will be clarified.

Line 185: authors stress “boreal wildfires” but then quote studies from slash-and-burn in Virginia, burning in Amazonia, burning in a temperate deciduous forest, and from crownfires in Yellowstone National Park! R (p. 218, l. 24-25) The justification for being more inclusive is presented in our general comments, and this point will be made more clearly and in more places in the revision. Also, tables 1 and 2 will be split into subsections for boreal and other sites. That these rather small tables represent worldwide information, much based on visually-determined charcoal, not chemically-defined BC, with so little for boreal sites, is really rather alarming.

Line 214: not clear what are high and low values - needs explanation. R (p. 219, l. 1) There is nothing in the text about low values, and “high” refers to the higher values in the previous couple of lines, from Clark and Royall (1994). Text will be clarified.

Line 224: so what are the consequences from this statement??? R (p. 219, l. 7-9) It is an example of how different methods give quite different estimates of PyC. There would be consequences e.g., if you tried to use the data for modeling.

Line 225-227: discuss conversion rates of biomass obtained by resistance to thermal oxidation but before authors say that methods may vary by a factor of two or more, so how can these data reliably used??? R (p. 219, l. 10-12) Our point precisely - the title includes “uncertainties” and one of our main conclusions is that a lot more research and standardization needs to be done. This is why large-scale BC production

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estimates come with such wide error margins, and and these estimates are used with understanding of their uncertainties, following standard practices of statistics. Reliability increases, or rather uncertainty decreases, as estimates are refined. They are a starting point. Lines 225-227 are the introduction to the paragraph, not the conclusion, but these lines will be rewritten as part of the general revision. Here and elsewhere, we will refer to new results from the BC ring trial (Hammes et al., 2006b).

Line 252: “less detailed”? still 4 pages! R (p. 220, line 7) Yes, but still less detailed than the sections on the solid forms of PyC. Four (small-format) pages are a small part of this review, and only summarize key findings of the many excellent papers on BC emissions and atmospheric aerosols. However, “less detailed” will be removed to avoid reader stress.

Line 287: discussion of BC emission in Asia. R (p. 222, l. 14-15) Yes, Asia, and preceded by a global estimate (l. 12-14). All part of the requirement to place boreal BC in a global context.

Line 305: what are the “total C trends”? R (p. 222, l. 5) This will be reworded for clarification as “trends for total ecosystem C in Canadian forests reported by (Kurz and Apps “1999).

line 368: this range (63-321ug kg-1) could mean anything and it is not giving any useful information that Wilke and Amelung found similar values. R (p. 224, l. 12-13) We find this comment astonishing, and totally disagree with both points. It is most important to have information on baseline soil PAH values (including those impacted by aerial deposition), and also important to look at more than one study (as the reviewer has pointed out). A new paper will be added on PAHs in the Moscow region (Wilke et al. 2005).

Line 381: seems to imply that PAH is particularly good for industrially-derived combustion products R (p. 224, l. 26-27) We do not how see this is implied. The studies described in this section analysed PAHs and then used the ratios of PAH abundances

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to derive information on their sources, including fossil fuel combustion and forest fires.

Line 391-392: this is an important point (formation of melanoidins) and should be discussed as a potential source of error in determining PyC. R (p. 225, l. 6-7) We will provide more information on this topic, including reference to Glaser et al. (1998), Hammes et al. (2006b), Brodowski et al. (2005b).

Line 126: this value (126 ppm) does not seem to be consistent throughout the literature and other studies quote 131 ppm - authors should aim to be inclusive. R (p. 226, l. 404? - Incorrect line number by referee) We are not trying to exclude any one, or be politically incorrect. We say “a broad aromatic signal with maximum around 126 ppm”, which is not the same as saying exactly at 126 ppm. This will be reworded to “with maximum around 125-131 ppm”.

Line 426-429: however, authors should also note that lots of problems exist with ¹⁵N-NMR, particularly with respect to observability and therefore the results need to be discussed critically! R (p. 226, l. 14-17) This is not an NMR paper, and in particular, it is not about ¹⁵N NMR. The many attempts to improve quantitation in solid-state ¹⁵N NMR are still not inconsistent with this general statement. We will change “proportion” to “increase” to enhance clarity, but this is not an appropriate venue for such a discussion.

Line 443: this is clearly not boreal! R (p. 227, l. 1, section 5.2) Agreed, see general argument earlier.

Line 488: just mentioning studies in boreal regions is not sufficient to make this review central to boreal studies R (p. 228, l. 14) Of course not; the broad information base on PyC characteristics, analysis, stocks, production and losses, including the limited data for boreal regions, are used as the foundation for sections 7 and 8.

Line 493: repetitive - as been discussed before in text. R (p. 228, l. 20) Yes, one line reiterates some previous information (start of p. 227?), but now it is in a different

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context.

Line 497-499: but only in productive ecosystems! R (p. 228, l. 25-26) This addendum is exaggerated. Depends on your definition of “productive” and “few”. Regardless of the site productivity, there is an input from fire-killed biomass, unless everything is consumed, which is not usually the case. Actually, Hicke et al. (2003) and other papers (and our field observations) show quick recovery of boreal forest productivity in many cases, especially as many boreal ecosystems are adapted to fire. Also, litter is not just leaves from trees, it include inputs from shrubs, herbs, mosses and roots. We can say something about the char being diluted as new organic matter is built up, without specifying the timescale.

Line 521: less sensitive Bloch decay? I don't understand, before it was argued that Bloch decay needs to be employed to correct for the underestimation of aryl C, thus appears to be more sensitive as it “sees” the actual C and not the H associated with the C (as in CP). R (p. 229, l. 21-22) This is playing with words and making an overly precise interpretation. Presumably reviewer means “not the C associated with the H?” which is not strictly correct anyway as aryl carbons may be underdetected by CP, but they are not invisible, except in extremes of remote protonation (rather pure soot or graphite). Reviewer is correct that BD is more sensitive in terms of quantitative detection of BC, but then CP is more sensitive in terms of how fast S/N increases with acquisition time, especially for samples that are not exclusively highly condensed BC. In fact the previous line 520 already noted that these structures were underrepresented by CP. “Less sensitive” will be deleted as unnecessary and potentially inflammatory.

Line 528-529: repetitive R (p. 229, l. 29 - p. 230, l 1) Repetitive of what? Unless I misunderstand the comment, these lines briefly explain the changes due to burning as seen by thermogravimetry, which is discussed nowhere else in our paper.

Line 546: two studies only? This is hard to believe and I'm certainly aware of more than just two studies on that subject! R (p. 230, l. 16-17) If there are more than two useful

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references in this specific context, we would like to know about them. This paragraph clearly refers only to quantitative studies of forest organic horizons. Other studies noted in the following paragraphs and in Table 2 deal with organic PLUS mineral horizons, or mineral horizons only.

Line 572: why do authors say that charcoal proportion would have been higher if small particles if smaller particles were isolated, but the study by Skjemstad et al. quoted there, did exactly that! R (p. 231, l. 13-14) The reference (Hopmans et al. 2005) was left out of the text, although it is in Table 1 and in section 4.1.1 (p. 218, l. 14), to which the reader is also pointed. Hopmans et al. (2005) discussed extensively the limitations of sampling charcoal >2mm, and pointed out that charcoal yields in their study would have been higher if smaller particles had been isolated, similar to results in many other studies. The reviewer apparently concluded that this referred to work by Skjemstad, previously discussed in section 5.2.2. While we erred in leaving out this reference, it should not be assumed that all Australian studies are by Skjemstad. The text will be corrected.

Line 617: needs reference to back up “presumed high production of BC”. R (p. 233, l. 1) This will be reworded e.g., to “with visually-dramatic charring of biomass and forest floor” or “associated production of PyC”.

Line 620-626: wouldn't it be useful to find more studies that use 14C dating here? R (p. 233, l. 4-9) Certainly, and we would appreciate information on such studies, after scouring the literature for them. However, most 14C-dated charcoal is from sediments or peat bogs, where it is well-preserved by lack of oxygen. Second, most of these studies look at relative abundance of charcoal in order to reconstruct vegetation and fire history, and except for Clark and Royall (1994) do not translate their data into mass of charcoal per unit area or other quantitative units useful for our purposes. Some very useful 14C data from a forest soil study (Gavin 2003) are discussed later (l. 830-834/p. 240).

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Line 631-63: this process/hypothesis needs to be explained more to convince (at what temperature, over what time span, are there experimental data?) R (p. 233, l. 11-12) These lines report a suggestion/hypothesis already made in two published studies as a possible explanation of their results. We welcome any studies that could provide further information, as this appears to be an important and underdocumented aspect of the BC cycle. We could say “they suggested that some charcoal might be consumed by subsequent fires”. This topic will be emphasized in the research requirements.

Line 642: study from 1966 on incubation of artificial graphite? If certainly biological oxidation of charcoal is occurring than wouldn't there be lots more recent studies? R (p. 233, l. 25-26) Clearly, this sentence only refers to graphite, for which to our knowledge there is only one published incubation study. The recent charcoal incubation studies are presented starting about 20 lines later.

661: what is “hardly any”? R (p. 234, l. 1). The incubation study by Shindo (1991) was unreplicated and the results reported qualitatively. The CO₂ evolution from soil amended with different size fractions of charred plant material was described as “almost the same” as from control soil, indicating that “these fractions were hardly decomposed” during the incubation. The text will be amended to make it clear that the results are qualitative.

Line 691: Chcarcoal production occurs within organic horizons? How so? R (p. 235, l. 14) Well yes, they do burn, along with fallen woody debris, joined by trees that fall over during or shortly after the fire. Some char is produced on trees that remain standing. Note that the sentence says “mainly” and “organic horizons and detritus”, which includes CWD, and even snags, depending on your definition. Both CWD and thick organic horizons are characteristic of many remote, unmanaged boreal forests, for which fire is a major disturbance. Will modify or leave out.

Line 708: wouldn't that migration rate heavily influenced by the soil type (clay content, water holding capacity), vegetation and climate? R (p. 235, l. 3) Of course that

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would be expected; these are the results for that particular study, presented as a direct quotation. We can mention the need for more information on this topic.

Line 727: I don't see the necessity for this section. R (p. 236, section 6.4) We do, as discussed earlier. This section will be rewritten to make its relevance abundantly clear. It may also be useful to change this and other section titles.

Line 742: why black shale??? Black shale is not a material that is derived by pyrogenic processes! Looks like authors are comparing apples with oranges here. R (p. 237, l. 7) See general comments at beginning. Our text nowhere says or even remotely implies that black shale is derived by pyrogenic processes; we compare substances on the basis that they have carbon structures resistant to decomposition. Anyway, maybe they are a bit like apples and oranges - I mean, if one is considering decomposition, both are fruit which would be expected to decompose similarly in soil. This is an overused metaphor, and we are not aware of any general prohibition of comparing apples and oranges; e.g., with regard to nutritional value, price, taste, etc. It is not as though we were comparing PyC with granite, ice, hamsters, etc..

Line 746: if lifetime of char is 8000 to 80000 years, then why does it occur in Devonian sediments now? R (p. 237, l. 11) The same question applies to coal and oil. As noted at the beginning of this response, except for the very small fraction of photosynthate that is preserved through geological time by lack of oxygen (less than 0.5% of C fixed), everything is decomposed eventually, by chemical or microbial processes. As elegantly put by Hedges and Oades (1997): "the only long-term shelter from mineralization is within anoxic marine sediments which accumulate one mole of organic carbon for every 500-1000 fixed by photosynthetic organisms". Text will be modified to include this, and to make it clear that we are talking about decomposition with oxygen exposure.

Line 778: exactly! Please use this in your critical evaluation of the studies that you discuss R (p. 238, l. 13) We feel that we have been doing just that, and the issue is raised at several points. In an imperfect world, one does not have all suitable data,

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and in we are always careful to specify the method used, especially in 7.1, the general estimate of how much BC is produced in boreal regions.

Line 782-785: repetitive R (p. 238, l. 17-20) We chose to repeat this information here, to remind the reader at this point in a long paper. However, the sentence will be shortened.

Line 821: Repetitive R (p. 240, l. 2) This presumably refers to the half-line “field data on the fate of PyC hardly exist”, which hardly seems “repetitive” as the first few words in a whole section. This introductory sentence will be rewritten to clarify why we should look to other recalcitrant forms of organic matter in attempting to predict lifetimes for the more highly condensed forms of PyC.

Line 827: the study by Bird et al is about the only study I know that quotes such short MRTs for BC in tropical savannas - authors need to be more inclusive and should also refer to other studies that report much older MRTs; otherwise, it appears like charcoal turns over rapidly in tropical savanna but has a more than 10fold greater residence time in other areas of the world (refere also lines 838). R (p. 240, l. 7 and l.19) As far as we could find out, Bird et al. (1999) still remains almost the only study to estimate turnover times of charcoal in any setting - and designed with that purpose. As we discuss rather extensively (and perhaps repetitively), there is hardly any real information on turnover times of charcoal or BC in boreal or other settings. In order to come up with some estimates of turnover times of BC and char in boreal regions we had to draw on some circumstantial evidence, including studies of other recalcitrant geochemicals and sediments (section 6.4). If there are other relevant studies that we missed in our extensive search and discussions with other scientists, we would appreciate being informed about them. We discuss quite extensively (starting l. 838) why the turnover times found by Bird et al. (1999) could be quite short, due to both environmental factors and finer size of grass char. Indeed, it does not seem unreasonable that there indeed could be a tenfold (and greater) difference in residence times of charcoal or other PyC forms, depending on physical size, degree of thermal transformation, and environmental fac-

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tors. It should be noted that very old fragments preserved in peat represent survivors preserved under mostly anoxic conditions, and we do not have the remains of what decomposed previously, and especially what decomposed with oxygen exposure. It is certainly dangerous to develop turnover times based on a few radiocarbon-dated fragments retrieved from low-oxygen environments. That is why we were so pleased to find the paper by Gavin et al. (2003) on charcoal in forest soil profiles in coastal British Columbia. It had both radiocarbon dates and other information enabling us to estimate a turnover time (which we discussed with Gavin). It may well be that only a small fraction of most PyC formed in forest fires has the very highly condensed structures with high resistance to decomposition, and it would be wrong to automatically assume long residence times, and thence infer long-term C sink strengths.

Line 847: explain what you mean by “loss” here? R (p. 240, l. 28) Replace by “turnover times”. However one defines it though, we were rather surprised to find no studies.

Line 849: wouldn't the influence of charcoal on soil function be important to know for all regions and not just boreal forest soils? Implies that the effect of charcoal on soil function is well understood for other areas. R (p. 241, l. 1) Of course, and we have covered this to some extent in previous sections on the general effect of fire on SOM, and studies showing a positive influence of charcoal amendment on soil fertility. However, as the reviewer has emphatically pointed out, our focus is on boreal regions and we do not intend to make a global study. But the second sentence goes beyond a logical interpretation. Does writing about topic A imply that everything about topics B to Z is known? Also, reviewer is inconsistent. First, the MS is heavily criticized for including non-boreal data and information, and now it is equally criticized for not discussing the whole world and focusing exclusively on the boreal. Sections 7 and 8 are definitely the most boreal-centric parts of our review.

Line 861-865: needs references!!! R (p. 241, l. 12-16) Sorry, they got lost. Should have been Richter et al. (2000) and Preston et al. (2006), plus a new reference (Kasischke and Johnstone, 2005).

Line 933: what do you mean by “char height”? R (p. 243, l. 26) This is height of char on tree stem. Text will be modified for clarity.

Line 893: can you really compare “activated char” with naturally-produced char? R (p. 242, l. 18-20) Not really, which is exactly the point we make, and especially emphasized by Hille and den Ouden (2005.). Some studies have used commercial activated charcoal (AC), but they do not have the same properties as natural char from fires. Text will be modified to emphasize the greater surface area of AC. Will add the new references Brown et al. (2006) on production and characterization of synthetic wood chars with properties similar to natural wildfire charcoal, and Naydenov et al. (2006), a boreal study unfortunately again using AC. It is surprising that so many studies use AC, not even in comparison with natural charcoal, when the latter is abundantly available after fire.

Line 904: shouldn't you also quote the work by Lehmann et al. here? R (p. 242, l. 27) Lehmann et al. (2005) was cited at l. 422 in relation to detection of aromatic clusters, and at l. 638 for oxidation. We did not feel that it was necessary to cite it at this point.

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