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Atmospheric Chemistry and Physics
Discussions

# *Interactive comment on* "Evolution of the chemical fingerprint of biomass burning organic aerosol during aging" *by* Amelie Bertrand et al.

**Anonymous Referee #1**

Received and published: 2 February 2018

## General Comments

This study investigated the chemical composition of biomass burning organic aerosol using online analysis techniques, principally Thermal Desorption Aerosol Gas Chromatography coupled to an Aerosol Mass Spectrometer (TAG-AMS). Solid wood fuels were burned in stoves and emissions were characterized. The smoke was exposed to hydroxyl radicals (OH), which increased organic aerosol (OA) mass and also reacted with components of the primary smoke aerosol. A variety of compounds were characterized and quantified, and the manuscript documents the quantities thoroughly, if not in such a dense manner as to make the paper challenging to read. Two main scientific comments arose during this review:

(1) The unidentified fraction of OA should be treated more carefully in the presentation

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of results. It is critical for the community to understand what was, and what was not, measured. In some cases, the treatment of unidentified fraction of OA in the text obscures the meaning of the results, which has been outlined in at least one comment below.

(2) The authors should be careful in their treatment of Stoves A and B versus Stove C. The authors describe their resolution of different burning conditions, however different fuels were used. At another point in the manuscript, a difference between Stove A/B and Stove C is attributed to the difference in fuel. Since both burning conditions and fuel type were different between the ‘smoldering’ and ‘flaming’ experiments, the authors should be vigilant and conservative in their interpretation.

With these concerns described and detailed below, it is the judgement of this reviewer that the manuscript is publishable in Atmospheric Chemistry and Physics with relatively minor revisions.

Specific Comments

Page 3, Lines 18-30: This introductory discussion would be improved by re-organizing the topics. It is suggested that after introducing levoglucosan, the possibility that it can be oxidatively degraded in particles should be discussed, followed by a discussion of the secondary OA mass production from biomass burning emissions. The study investigates the topics of (1) emission factors for POA, (2) oxidative degradation of tracers, and (3) changes in OA composition due to secondary oxidation. The introduction should also follow this logical progression.

Page 3, Line 21-23: Is this statement meant specifically for biomass burning emissions? Please clarify.

Figure 2: It appears that TAG-AMS sampling is represented by both light green and light blue. If this is the case, please edit the legend to clarify.

Page 4, Section 2.1: How was the total OA quantified? If by HR-ToF-AMS, then this should be clearly specified and qualified by any sampling biases.

Page 6, Lines 17-21: This can be removed from the methods section as it is actually a statement of the results (and will, indeed, be discussed at great length in the results section).

Equation 1 (and supporting text): What value of k_wall/p was used?

Page 7, Lines 28-29: Please clarify what is meant by “we identify between 26-85% of the POA mass concentration”. Ensure that the manuscript clearly identifies how total OA was quantified. It is assumed that the ‘identified OA’ is the mass concentration quantified by TAG-AMS.

Figure 3: Showing this figure with an ‘unidentified’ section of the bar would be more appropriate, since different amounts of OA were identifiable in each experiment.

Page 8, Lines 2-3: The text states “The MCE values for the experiments conducted with this stove [the pellet stove?] are 0.97,” but Table 1 shows “MCE > 0.99” for pellet stove experiments. Which of these values is correct?

Page 8, Lines 14-17: Please clarify (with evidence and explanation) that the nature of the artifacts that should have been associated with the prior studies based on methodology are actually consistent with the differences between the prior studies and the present study, as is vaguely suggested in the submitted manuscript.

Page 8, Line 27: Methoxyphenols are not the ‘predominant’ class of species in the primary BBOA – Figure 3 clearly shows that anhydrosugars comprise the predominant fraction of the identifiable OA.

Page 9, Lines 5-8: This sentence embodies an important criticism of the data interpretation within this study. The authors describe differences between the softwood and hardwood samples. At the same time, hardwoods and softwood fuels were burned under different conditions. The authors seem to interpret differences between the log wood stoves and the pellet stove as differences in fuel at this point, while at other points, the differences are attributed to burning conditions. If the interpretation given here is thought to be accurate, then an explanation for why burning conditions don't influence the observed difference must be described.

Section 3.2.1: The authors state that the primary compounds initially represent 48% of the POA, but after oxidation (4-6 hours) they represent < 8%... of what? Of POA? Of total OA? Can this just be described by the addition of SOA mass to the particles? Figure 6 shows that the relative contribution of levoglucosan is somewhat stable around 40% of identified OA throughout the experiment. Please clarify what is being quantified and to what fraction of the aerosol the percentage is referring. It seems from this text that the mass concentration of levoglucosan, for example, decreased substantially during exposure to OH, but Figure 6 suggests that it is relatively stable against aging based on how the data are shown. Then again, Figure 7 shows the ER = 0.6, so the actual concentration is decreasing during aging. Figure 8 shows that levoglucosan is decreasing as a fraction of total OA. Basically, Figure 6 seems inconsistent with Figs 7 and 8. Is the change in the identifiable fraction playing a role? It is very likely that the data are somehow telling a consistent story, but it is not presented clearly. [Note that this comment explicitly references levoglucosan, but the comment also applies to other Primary Compounds.]

Page 12, Line 33 – Page 13, Line 2: What is the relevance of the ratios of nitrocatechols to levoglucosan, since it is known that the latter is consumed during OH exposure?

Page 13, Line 3: Higher levels of 4-NC are discussed with reference to prior observations, however far larger discrepancies between the prior observations and the present study exist for MNC/LG. The 4-NC/LG ratio range at least overlaps the prior studies, but the MNC/LG ratios are shifted by about a factor of 10, yet are not discussed.

Page 13, Line 7: Does “the mass spectra (EI, 70 eV)” refer to those obtained from a database? Please clarify. If from a database, please cite the reference.

Page 13, Line 23-24: The impact of burning conditions is less clear since the fuel was different for Stove C. Please clarify this issue in all instances within the manuscript, including Page 9, Lines 5-8 as discussed in a comment above.

Page 14, Lines 1-3: How might measurements be improved to detect and quantify a greater fraction of the POA and SOA mass? Can this be inferred from the limitations of TAG-AMS? [A discussion of this topic may not be entirely appropriate for the conclusion section, but may be a worthwhile topic to include elsewhere in the manuscript.]

Technical corrections –

Page 3, Line 4: correct the in-line citation

Page 9, Line 3: correct the in-line citation

Page 9, Line 21: consider beginning a new paragraph for fatty acids

Page 13, Line 8: “Figure 10” should be “Figure 9”

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