

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

Anonymous Referee #2

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Review.

Evolution of the chemical fingerprint of biomass burning organic aerosol during aging

Overall.

This paper uses TAG-AMS to quantify compounds in organic aerosol from two log stoves and one pellet stove after emission and after equivalent atmospheric aging in a chamber. Three categories of compounds are described, and the emissions factors and emission profiles of biomass burning tracers are measured. The authors quantify how the OA changes after atmospheric aging. This paper is generally well-written but would benefit from a read through for typos (see examples in editorial section). Additionally, more discussion on implications of this study could be included in the

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conclusions. Some suggestions for improvement are below, but overall the paper is interesting and worthy of publication.

General.

The variability in the total OA and the resolved OA for each stove type is interesting and should be further explained. It is unclear that the three or four samples from each should be simply averaged as was done in Figure 3. That resulted in one composition for each stove type without anything showing the potential variability in the composition for each stove. The discussion in the text regarding this composition should clarify if it is in regard to the total POA or the resolved fraction of POA. This should be stated clearly and be used consistently. Specific comments are below.

In the section on atmospheric aging, the authors should be more explicit about what measurements went into the results of the gaining experiment. It seems as though all of the OA was aged, but there is no explicit mention of which stove was used to generate the POA that was then aged. The initial composition of the POA could influence the SOA composition, so there should be more discussion of this, unless the resolved fraction is too small to draw conclusions.

Specific.

P. 3, L. 6-9: It would be better to incorporate these references into the paragraph and the following sentences. More detail is included later about the biomass burning, but not the other sources.

P. 3, L. 17: This is redundant with the previous mention of Levoglucosan at L. 11. Remove or rephrase.

P. 4 – Is particle bounce a factor in the TAG-AMS? Was that taken into account in the calculations of EFs? How does the TAG-AMS treat organics on black carbon? Can the black carbon be detected?

P. 4, L. 30: Comment on what is causing such a large range in OA concentration.

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Does this have anything to do with the amount of time it was burning? How was this controlled for? Does the emission factor of each stove change with the amount of time the fuel has been burning? How did the emissions change over the 30 minutes they were stabilizing in the chamber? More details should be added here.

Table 1: What is the difference between “in the chamber before lights-on” and “values retrieved just before lights on”? What is the timing difference between the [BC] and the [NOX]? Why are the concentrations of BC, etc. not consistent? What other conditions were changed?

Figure 1: The words are all very small. It might be possible to shift so that the figure takes up less space (i.e. is less diagonal).

P. 5, L. 16: “discrimination of all fragments below m/z 10” – Explain or rephrase.

P. 6, L. 10: Wall losses decreased the concentration of aerosol, and this was accounted for qualitatively by increasing the sampling length as time progressed. Mention here that the quantitative estimates for wall losses are included in Section 2.4. These questions came up here before reaching Section 2.4: Are there any calculations or quantitative estimates of the wall losses? How much time was added to each sampling period, and how was that calculated?

Figure 2: The legend is somewhat confusing. Suggest removing “TAG-AMS Sampling” and changing the caption to be “Figure 2: Example of the TAG-AMS (light shading) and ...”

P. 6, L. 11: Was this an automated matching process? What confirmed a match? Comment on how were the initial standards and NIST spectra were selected for comparison.

P. 6, L. 30: What is eBC? Write out or explain the first time it is used. On the next page, it is listed as black carbon. In equation 2, it is written as just “BC”. Make sure these are consistent, and if they are different, explain what that difference is.

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Equation 1: Remove the “.” – not standard formatting for equations.

P. 6 – Is there any preferential wall loss based on composition? The only aerosol tested is eBC. Is that representative of all of the aerosol?

P. 7, L. 7: How reasonable is that assumption? Was it tested?

P. 7, L. 19: Clarify or rephrase the discussion of the symbols in Equation 2. “Here refers to the concentrations” – but really, it refers to the change in the mass of each.

Figure 3: Reorganize the legend so that they are left justified. (If this is based on the groups discussed in the text, then include headings for each type). This figure would also be more useful if it could also demonstrate the fraction of total OA that it represents. It would also be nice to have some error bars or some representation of uncertainty. The fact that for Stove B, 26 to 85% of the OA was resolved at any time could also mean that there is large variability in the relative fractions. It appears that the main point of the graph is that Stove A and Stove B have similar compositions. But it would be good to see some representation of the differences in their total OA concentrations and the amount of OA resolved for each. In addition to or instead of these average bar plots, I suggest including a figure with a bar for each sample, grouped by stove type, with the y-axis representing the fraction of total OA resolved, instead.

P. 7, L. 28: Explain where the 26-85% of the number for the 26-85% of the mass comes from. In Figure 3, are the ranges due to averages taken for each sample? That is a lot of variability. That variability is in itself interesting and should be discussed.

P. 8, L. 4: This statement is not consistent with the figure. There is huge variability in the fractions of OA identified in all three stoves. Stove B has the lowest low range but also highest high range, so it is an overgeneralization to say that Stoves A and B are less resolved. Or, more data and discussion needs to be added to support that.

Figure 4: Remove the gap between a/b and c/d and the x-axis labels, since they are the

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same as those at the bottom for e/f. Why are R values on c/d and not on the others? Remove them or add the others to be consistent.

P. 8 – Are these fractions of total POA or resolved POA? There is a huge range in total POA as well as resolved POA, and there appears to be a range in these sugars as well. If the standard deviations are included in the comparison, there is no statistical difference in the percentages of anhydrosugars from the three stoves.

P. 9, L. 27: It is unclear how Figure 5 illustrates that.

Figure 5: State the difference between a and b in the caption. This figure could also be moved to the supplement. It is only referenced once.

Table 2: Remove “Bin” labels, or explain them. It seems like enough for each column to have integrated OH exposure. Is this for one sample of one of the stoves or an average? It is unclear what experimental set up this data came from.

P. 10 – Clarify the units of OH exposure. This may take a new reader some time to interpret. The “integrated OH exposure” seems to be just the OH concentration (molecules/cm³) multiplied by the time exposed (hours).

Figure 6: Same legend comment as Figure 3. What POA was used in this figure? What stove is this from?

P 10 – Why does the fraction of identified OA decrease with OH exposure? This almost implies an inverse correlation between OA mass and fraction of identified OA. It seems like the OA produced at the very end is then all unidentifiable.

Figure 7: This figure is interesting, and the split between the red and blue is nice. But having the primary compounds with upside down bars make them seem negative. Additionally, the ER ratio increases from left to right, but the bars decrease in size, which is counterintuitive. The caption should state what the error bars are calculated from (and the text could comment on why they vary significantly).

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P. 11, L. 19: Interesting discussion! Figure 9: In the text, this is listed as Figure 10. What is the explanation for the difference between the two measurement types? It would be good to show the slope here, in addition to the R squared value.

P. 14 – Add a couple more sentences about the implications of these results and why it is important that they were measured.

Editorial: P. 2, L. 3: Add “of” after “evolution” P. 2, L. 6: Remove redundant “emission”
P. 3, L. 21: Change “during” to “due to” P. 8, L. 19: Figure 6? Should this be Figure 4?
P. 10, L. 23: Add “with” after “along” P. 13, L. 8: Change Figure 10 to Figure 9

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-1196>, 2018.

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