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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Characterization of gas-phase organics using proton transfer reaction time-of-flight mass spectrometry: fresh and aged residential wood combustion emissions" *by* Emily A. Bruns et al.

Anonymous Referee #2

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This paper presents measurements of gas-phase species emitted from combustion of beech in a 'modern' residential heating stove in both fresh and aged states. Five replicate laboratory experiments were conducted in which emissions from a steady flaming combustion were sampled into a laboratory smog chamber, from which there were sampled for both a primary characterization period and during oxidation by OH formed via HONO photolysis. The use of replicate experiments is useful, as it assesses the extent to which emissions vary even within narrowly controlled circumstances – as it turns out, quite a bit. Interestingly, many of the emissions of concern (CO, OA, BC) are quite consistent across tests, as is the SOA produced during aging. In contrast,

the composition and emission factors of NMOG varied substantially, with two tests having markedly different NMOG emissions. The evolution of NMOG is described in which many identified species show the expected decay with OH oxidation, while other species (acids, other O-containing species) showed enhancement.

This paper is clearly written and the measurements and analysis appear to be of high quality. This topic is of great interest to the readership of ACP as it provides important insights into the composition and evolution of an important class of biomass burning emissions. Below I have highlighted several points that would like to see addressed in revisions. The main focus of my comments is on the difference between the two sets of experiments – I would like to see a bit more discussion of the conditions that lead to these differences and how the two 'anomalous' experiments (#2, #3) differ from the others. This seems to be a key point, and while the differences are discussed, there's little investigation of what might have influenced this difference. For example, the fuel consumed was nearly half during these experiments than the others, why? One general comment is that I was really required to read the other Bruns et al. 2016 paper in order to understand and interpret these results. While I understand that the authors split these aspects of the reporting of the project to avoid a cumbersome manuscript, I would like to see this one 'stand alone'. So, at the least I would suggest that a table of basic test parameters (like Table 1 in Bruns et al 2016) be included, perhaps in the SI. Another general comment is that there is a bit of an over-emphasis on differences between results observed here (one fuel, one combustion condition) and observations more generally. In most cases, these comparisons are appropriately caveated, but in some cases the generalizations are a bit sweeping (e.g. Line 283) – I ask the authors to give this a once over to ensure that these results, while certainly providing key insights and data, are not over-extrapolated. Finally, I second many of the concerns/questions of the first referee.

Once these general and specific points have been addressed, I recommend the manuscript for publication in ACP.

Specific points

I am a bit confused by Table 2 – this indicates acetonitrile as the only N-containing species, but EF of acetonitrile is ∼10 times lower than that for N-containing species? Is the rest of this mass contributed by un-identified compounds? It also doesn't seem as if N-containing species contribute 20-30 mg/kg to NMOG mass on Figure 1, but it would be hard to see there.

N-containing species were higher for Expts. 2-3, but aerosol-phase nitrate was substantially lower (even accounting for lower fuel consumption) (Bruns et al. 2016). Were there any other notable differences? E.g. NOx levels? Have you examined secondary nitrate formation during aging?

L255 – Figure 2 is just mentioned here, and this could bear a bit more discussion and the differences between experiments discussed a bit further. One thing that stands out about these two experiments (2, 3) is that the CO2 loading in the chamber was substantially (almost half) lower than the other three experiments (despite the fact that the MCEs and many other quantities are essentially the same. If the injection time and dilution conditions were the same, this suggests that perhaps the combustion rate was lower (which would probably be indicated by lower flue temperature). Are there any other contextual or operational details that were different?

L283 – This is too broad/definitive of a statement to make based on the narrow set of conditions tested here.

L317-319 – It would be useful/instructive to attempt a mass balance on the NMOG and SOA loadings to estimate how much of the measured NMOG may be ending up in the condensed phase in your experiments.

L358 – Were terpenes actually quantified? I don't expect much from birch wood, but if you measured them (or found them BDL) this should be noted.

L362 – 'Good agreement' is a bit vague, there is not-great agreement in panels c) and

f). Could this indicate possibly misattribution of these compounds? For this figure, it would be helpful to show smoothed data (and probably a log y-axis) to make this a bit more readable.

L365-370 - Somewhat confusing lead-in to discussion of Figure 3, as discussion emphasizes differences between experiments and this figure shows averages across all experiments - may make sense to just discuss this result then discuss inter-experiment differences (using Fig. 2)

References

Bruns, E. A., El Haddad, I., Slowik, J. G., Kilic, D., Klein, F., Baltensperger, U., and Prévôt, A. S. H. (2016). "Identification of significant precursor gases of secondary organic aerosols from residential wood combustion." Scientific Reports, 6, 27881.

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