

Interactive comment on “Investigations of primary and secondary particulate matter of different wood combustion appliances with a high-resolution time-of-flight aerosol mass spectrometer” by M. F. Heringa et al.

Anonymous Referee #1

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The present work describes experiments that were done to investigate the photochemical aging of biomass burning emissions from several wood burning devices under different operation conditions. The work is new and meaningful to the atmospheric science community, particularly in regard to the evolution of biomass burning emissions. The manuscript is well written and well organized, and the data are of high quality. After the following comments are addressed, I recommend that it be published in ACP.

Specific Comments 1. Abstract, pg. 8082, ln. 27-28: “...possibly indicating a higher ratio of acid to non-acid oxygenated compounds in wood burning OA compared to other

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OOA.” Given the harsh ionization source employed by the AMS, I don’t believe such a conclusion can be drawn about the nature of the oxygenated functional groups. Of course, it is appropriate to discuss O:C and H:C, but not functional group contributions.

2. Pg. 8086, ln. 27: upper size range of the AMS is given as 1.5 μm , however, the particle transmission efficiency in the AMS decreases for particles $> \sim 0.7 \mu\text{m}$. If this size range of 35nm to 1.5 μm is stated, it should accompanied by some discussion of the transmission efficiency in the 0.7-1.5 μm range.

3. Figure 2 shows results from experiment 15 – which in Table 1 lists a POA concentration of 31 $\mu\text{g}/\text{m}^3$. However, in Figure 2, the POA concentration after mixing appears to be $\sim 8 \mu\text{g}/\text{m}^3$. What is the cause of this difference?

4. Pg. 8089, ln. 23: Since the wall loss correction method is the same as that used by Grieshop et al. (2009), it is appropriate to cite that reference here.

5. In the determination of emission factors and MCE values, CO_2 and CO measurements were used. Based on the factor of 220 dilution, it seems as though the CO_2 and CO enhancements in the chamber were quite low, as compared to the instrument spans – what is the uncertainty in emission factors due to this issue?

6. Also on the topic of emission factors, Table 2 gives emission factors \pm some value, which is presumably a standard deviation from the repeated tests, and not an actual uncertainty. For these emission factors, an uncertainty should also be given due to the uncertainty in estimating the amount of fuel burned and uncertainty in the measurements (see above comment).

7. Pg. 8094, ln. 26-28: some of the pure SOA experiments also seem to show a decrease in O:C with aging?

8. Grieshop et al. 2009a and Grieshop et al. 2009b should both be cited

9. This current work relates to some other studies that also investigate aging of biomass burning emissions. Perhaps some discussion comparing the present results

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to these other studies would be interesting: Capes et al. (2008); Cubison et al. (2011); and Hennigan et al. (2011).

10. One of the unique features of this study is the observed increase in m/z 60 from SOA production. The uniqueness of this result should be highlighted more than it is, and the discussion expanded (i.e., why is it that m/z 60 increases in the log wood burner experiments, but never in the pellet burner experiments?). Also, is there any evidence from ambient measurements that m/z 60 production occurs in the atmosphere? My impression from the extensive body of AMS studies is that m/z 60 is not produced from secondary processes, but is thought to characterize primary BB emissions. Some additional discussion on this topic would be appropriate.

11. Is there any correlation (or can one be derived) between the increase in O/C and the amount of SOA formed?

12. Could variability in the amount of SOA formed be accounted for by differences in OH levels? It would be interesting to view Figure 3 (and possibly Figures 6 and 7) on an OH exposure axis in addition to time. Since emissions from the appliances used should be highest in winter (corresponding to lowest OH levels), OM enhancement ratios vs. OH are important for the atmospheric implications of this work.

13. Figure 9; pg. 8096 – is it necessary to call the CO_2^+ ion ' pCO_2^+ '?

14. Conclusions: "This implies that the gas phase emissions from log wood burners play an import role in the total contribution of OM from residential wood burning appliances to ambient OM and should be considered to be included in future legislations" – Dilution ratios were >200 , could this SOA not be from evaporated POA (i.e., according to the mechanism proposed by Robinson et al. (2007))?

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