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Atmospheric Chemistry and Physics Discussions

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 #2

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The submitted manuscript investigates primary and secondary organic aerosol derived from wood burning, with a special focus on SOA production and evolution. Experiments have been conducted with atmospherically relevant concentrations and several wood burners and combustion conditions (ie. flaming, starting, smoldering) have been tested. Considering the growing body of evidence of the great influence of this specific aerosol source (especially in urban area during winter) the work presented here is very interesting and provides significant data for our understanding of biomass burning aerosol particles and their evolution in the atmosphere. I recommend the publication

of this work in ACP after the following comments are addressed.

General comment

My major concern is related to wall losses corrections (WLC) used here. Indeed biases in WLC can greatly affect the conclusions of this study and especially the relative increase of m/z60 during the photo-oxidation phase (see specific comment). WLC are determined using BC concentrations and based on the assumptions that the aerosol is internally mixed (One can regret the absence of VHDTMA measurements). Thus the WL are assumed to be the same for BC and OA and it sounds like a suitable assumption. However in a recent study carried out in a combustion chamber (data not published yet) we observed a systematic higher deposition rate for BC (measured by MAAP5012) than for OA (measured by AMS). These experiments had also been performed with atmospherically relevant concentrations of biomass burning aerosol particles but in dark conditions. Such behavior is still unexplained in our case. If this behavior is also observed here the conclusions regarding m/z 60 could be drastically different. Please can you verify this specific point with your data obtained before the photoxidation phase. Considering fig 15, this verification appears possible in order to validate your WLC procedure.

Also on a related topic, BC time series presented in fig15 is a bit suspicious. I assume that the spacing between each series of points correspond to the filter spot changes. Here I do not comment the global increase of "BC" after the lights were switched on, but the slight increase of BC in almost each series of points. Can you comment this behavior? Does any intercomparison between MAAP and Aetholometer has been performed in this study?

It would have been valuable for the paper to present and discuss some pToF results. What are the size distributions of the aerosol and their evolution during the photo-oxidation phase? Is the aerosol mass distribution close to 1μm ? As particle transmission efficiency in the AMS strongly decreases for particles larger than 800-1000 nm

and as BC measurements are associated with TSP, the presence of particles larger than 1 μm can affect the WLC used here.

Specific comments.

1- p 8082, line 16. “Average emission factors of BC+POA+SOA, calculated from CO2 emission,. . .” From a conceptual point of view, it is difficult to assess a SOA emissions factor as SOA formation is not a linear process, depending on many external parameters. Especially in urban areas where NOx concentrations should be more higher than those encountered here. In this respect no NOx concentrations are reported in the paper although measurements have been performed (p8088, line 15 and fig 1). Is it possible to dissociate the SOA emission factor from the other two (BC and POA) and had one or two sentences to avoid misunderstanding with this SOA emission factor.

2- p8084 line 20 : “In addition, results from pure SOA experiments, where only the gas phase emissions are injected and processed, are presented”. According to this sentence I would have expect more information related to these very interesting pure SOA experiments. Results of these experiments are not highlighted in the text. Is there any significant differences between this pure SOA and the classical OOA factor? Do you observe a modification of the mass spectra during aging? Does the absence of POA affect substantially the SOA formation?

3- P8086 line 16 : Is there any influence of the use a gas torch ?

4- P8086 line 27 : AMS results are generally assume to correspond to the PM1 fraction. This sentence suggests that the results presented here correspond to the PM1.5 fraction. Transmission efficiency for particles larger than $\sim$1μm should be discussed in the text.

5- P8087 line 19-22 : As CO2 has been measured during the experiments, does the fragmentation table has been modified in order to consider the real concentration of CO2?

6- P8087 line 25 : The inlet of the TEOM was heated at 50°C?

7- P8093 line 12-13. Do you have an explanation of the increase of the CHN family? Is this increase significant?

8- P8093 line 25. As almost every organic compound levoglucosan is not inert. The reactivity of levoglucosan is still a hot topic, and need to be discussed in more than a sentence.

9- P8093-8094. m/z 60 increases during photo-oxidation processes is most probably the point of this paper and needs to be discussed in more details. Such results have not been observed in the study performed by Hennigan et al (2010) (cited line 22). As discussed in the general comments I consider that WLC can play an important role on this unique behavior. In addition to what I propose in the general comments do you observe the same behavior for PAHs or m/z 137 (for example) ? In a more general point of view, regarding fig 6, only one experiment exhibits a high increase of m/z60. If we do not consider this particular experiment, the average relative increase of m/z60, considering all the other experiments, is more or less equal to 1 or 1.2. The important variation of this relative increase between experiments is also puzzling. Results should therefore be discussed more carefully.

10- P8095 line 10 : “indicating that the condensing OM has a similar O/C ratio during the entire experiment”. Do you suggest that the increase of O/C ratio for the other experiments is due to the aging of POA and not the aging of SOA? Since O/C ratio of SOA is constant all along the experiments, we should observe a decrease of O/C ratio for experiments in which POA exhibit higher O/C ratio than SOA.

11- P8096, line 9 : aging instead of ageing

12- P8096 line 14 : “which represent the OA functionalities better than the UMR data” As O/C and H/C ratios have been calculated here, Van Kervelen diagram could be a better representation in order to discuss the functionality of OA and its evolution during

aging.

13- Table 1 : Can you add in this table BC and NOx concentrations?

14- Figure 3, 6 and 7 : These 3 figures are not clear (too many curves). I suggest to split them in two figures : one for log wood burner and one for pellet burner. Also the use of point + fitted curves instead of lines between point could increase the readability. At last a link between each curve and experiments listed in table 1 could be very useful for the reader.

15- Figure 9 : Can you reverse the color scale (time after lights on)?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 8081, 2011.