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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diurnal variations of residential particulate wood burning emissions and their contribution to the concentration of Polycyclic Aromatic Hydrocarbons (PAHs)" *by* L. Poulain et al.

Anonymous Referee #2

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General comments. The manuscript analyzed the impact of residential wood combustion on the submicron aerosol load of a small village (Seiffen) in Germany. To this end, a set of on-line (AMS) and off-line measurements (filter sampling and laboratory analysis) was conducted. The positive matrix factorization (PMF) method was used to estimate the contribution of the emission sources to the aerosol organic concentration and several tracers were included to help identifying the different sources. The contribution of the wood combustion to the highly-resolved PAH concentrations was also discussed. Due to the renewed interest in residential wood combustion as alternative

of fossil fuel and nuclear power consumption and its high impact on air quality, the topic of the manuscript by Poulain et al. is relevant. The manuscript is recommended for publication in the Atmospheric Physics and Chemistry Journal after a major review. The authors should address all comments and modify the manuscript accordingly. The manuscript should be also improved from the grammatical point of view.

(A) Major comments

1. Title review. You measured ambient concentrations of particulate matter (immission) in an area impacted by residential wood combustion emissions, rather than measuring residential wood combustion emissions. Thus, the title should be reviewed to correctly describe the content of the manuscript.

2. Weekday vs. weekend diurnal cycles (AMS measurements and modelled factors). In order to test the significance of aerosol concentrations being different during weekends than on weekdays, a statistical test should be performed for each hour of the day. Once you know for which hours the aerosol concentrations are statistically significantly different, you can discuss your results and relate them to urban anthropogenic emissions expected for weekdays and weekends at that particular site.

3. PMF parameters. Before reporting your PMF results, you should address different topics related to the selection of the PMF parameters (number of factors, rotational ambiguity, error model, PMF mode, outliers treatment) and perform tests on PMF runs (global minimum, goodness of the model fit, and model uncertainties). The selection of the parameters is critical for the correct interpretation of the results and was not mention at all in the manuscript. There are many papers presenting PMF results which addressed the above mentioned topics. I recommend strongly you use as a guide the papers by Lanz et al. (2007) and Ulbrich et al. (2009).

4. Interpretation of PMF results. This section is unclear and incomplete in relation to the discussion of the PMF factor and the identification of the possible emission sources of the organic material. The methodology used for the emission source identification

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should be explained in more detail as well as the selection of the tracers for each factor. The objective of a receptor model (e.g. PMF) is to identify the emissions sources after statistically analyzing the measurements conducted at the receptor. Once you identified the emission sources, the next step would be to determine the contribution of each source to the aerosol load (in this case, to the organics concentrations). Thus, you should clearly state the contribution of each emission source to the organics concentration at Seiffen. If you are not able to separate the contribution of the residential wood combustion emission because it is contributing to two different factors, you should inform the reader on this problem.

5. High PAH concentrations and linear correlation with BBOA (sections 3.2.2 and 3.2.3). Figure 6 presents the time series of PAH concentrations derived from AMS measurements. During the sampling period, several short-term very high concentrations (500 ng m-3 vs. a mean value of 10 ng m-3) were observed. The authors attributed these high peaks to strong local sources of PAH such as a plume of a chimney exhaust of neighbouring houses. Figure 7 is a scattergram of PAH concentrations derived from AMS measurements vs. modelled BBOA concentrations. Samples in red were labelled as no correlating to wood combustion emissions. You should check whether these "red" samples have a linear correlation with BOAA but with another slope coefficient of determination, before discarding the non-correlation. These "red" samples correspond actually to the high values observed in Figure 6 that were attributed to strong local sources of PAH. In the case the origin of these high PAH peaks is not connected to biomass combustion, they might be linked to the use of liquid fuel for residential heating. Then, you should review your statements connecting very high PAH concentrations and wood combustion emissions.

6. Linear correlation. In different occasions along the manuscript, you presented linear correlations between AMS and off-line measurements. You should review the validity of your results due to the low number of samples (4-5 filters) used in the correlation.

(B) Minor comments

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Section 1, Introduction. You could comment on the biofuel consumption trend and share (wood logs, pellets, etc) in Germany and, particularly, in the Saxony region where your study took place.

Section 1, page 11581, lines 24-25: Add a comment on the European Union targets for biofuels consumption to reduce greenhouse gases emissions and energy dependency (UE, 2007).

Section 1, page 11582, lines 12-14: "Aerosol particles . . .". Not all particles emitted by residential wood combustion are considered to be carcinogenic agents. Thus, correct your statement.

Section 1, page 11582, lines 22-24: "During the winter time, . . .". A reference to previous studies conducted at Seiffen is needed to affirm that residential wood combustion is an important and significant source of aerosol particles in Seiffen.

Section 2.1, page 1583, lines 9-11: "The AMS instrument was . . .". What do you mean by writing that the AMS instrument was installed at a "safe" distance from the main street?

Section 2.1, page 1583, lines 7-9: Add population living in Seiffen.

Section 2.1, page 1583: Add some words on the fuel share for space heating in residential areas in Seiffen.

Section 2.1, page 1583, lines 15-16: Where was the meteorological site installed?

Section 2.2.1, page 1583, lines 21-22: Replace "January 10" by "10 January". Add details on the cut-off size and sampling frequency of the AMS instrument. Include details on the CO and NOx measurements conducted at the site and used in Figure 3.

Section 2.2.2, page 11584, lines 18-21: Add filter diameter, flow rate of the sampler and sampling period of the off-line measurements.

Section 2.2.2, page 11584, line 26: Define EC and OC.

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Section 2.2.2, page 11584, lines 26-27: Comment on the use of blank filters and/or other methods to account for positive and negative sampling artefacts affecting the EC/OC measurements.

Section 3.1, page 11585, line 8: Replace “parameters” by “variables”.

Section 3.1, page 11585, line 18: Does the AMS instrument really cover the entire PM1 range? Depending on your answer, modify the sentence accordingly.

Section 3.1, page 11585, lines 23-25: Besides classifying the temperature inversion as light, report the temperature gradient. Section 3.1, page 11586, lines 2-4. More explanations and comments on Table 1 are needed.

Did you use the same time period to perform the mean calculations?

Did you match AMS measurements with the measurements from daily filters collected every 4 days?

Comment on the different cut-off sizes of the AMS and filter sampling and their effect on the concentrations results.

Comment on the organics obtained by the filter sampling (organic carbon) and the organics by the AMS (OM).

Section 3.1, page 11586, lines 5-23: Define working days and non-working days. If working days match the Monday-Friday period, then switch to weekday (Monday-Friday) and weekend (Saturday-Sunday) notation to avoid confusion.

Section 3.1, page 11586, lines 17-19: “During the week, . . .” Do you mean that the diurnal pattern observed in the measurements during weekdays and weekends is linked to anthropogenic activities? Reword this sentence to clarify it.

Section 3.1, page 11586, line 21: Replace “appended” by “happened”.

Section 3.1, page 11586, lines 19-23: Reword this sentence since it is not clear.

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Section 3.2.1, page 11586-11587: Add details on the AMS data treatment prior to apply the PMF method, such as number of samples for weekdays and weekends, size of the matrix, exclusions of any m/z ratios.

Section 3.2.1, page 11587, lines 21-23: Why the comparison of daily EC concentrations with HOA factor time series was not conclusive? Did you perform a linear correlation between them? Add details to your statement.

Section 3.2.1, page 11587, lines 26-28: It seems the sentence is not complete.

Section 3.2.1, page 11588, lines 1-4: You should perform a linear correlation between HOA and BBOA times series to statistically check the similarities between these modelled series.

Section 3.2.1, page 11588, line 6: Define “OAA-1”.

Section 3.2.1, page 11588, line 20: Define “OAA-2”.

Section 3.2.1, page 11589, line 5: It is not clear your reference to “the authors”. Do you mean “Poulain et al.” or “Iinuma et al. (2010)?

Section 3.2.1, page 11589, lines 25-29: The increase of concentrations in the evening can suggest, but not indicate a stronger emission. During winter time and especially at night, the lower layers of the troposphere can be very stable and prevent the atmospheric pollutants from vertical dispersion. Hence, you cannot rule out the possibility of observing higher concentrations due to the lack of dispersion under similar levels of emissions. I suggest you reword your sentence.

Section 3.2.1, page 11590, lines 13-16: Your statement is not correct: 1) Krecl et al. (2008) found that aerosol concentrations were statistically significantly higher on weekends than on weekdays between noon and midnight with peak values in the evening, whereas your study reveals another pattern for the aerosol concentrations. 2) The difference between weekday and weekend concentrations cannot be generalized as shown by the measurements conducted in Seiffen (your study) and in Lycksele (Krecl

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et al., 2008).

Section 3.2.2, page 11590, lines 24-25: “Wood combustion . . .”. Add references.

Section 4, page 11595, lines 10-15: I agree with the authors on the advantages of using a combination of on-line and off-line measurements in future field campaigns. You should also highlight the importance of conducting longer field campaigns (a 3-week campaign is not long enough to be representative of all meteorological situations and emissions conditions) and to carry out daily filter sampling everyday to increase the number of samples when applying statistical analysis.

Section 4, page 11595, lines 21-23: This sentence is incomplete and should be revised. Residential wood combustion emissions are highly variable and depend on factors such as appliance type, burn rate, type and amount of wood, and wood moisture content (Johansson et al., 2004). The impact of residential wood combustion on air quality depends on the atmospheric dispersion conditions as previously mentioned (see comment Section 3.2.1, page 11589, lines 25-29).

Table 1. Define “OM” and the values after the symbol “±”.

Figures.All figures present small font sizes in legends and axis labels. Thus, provide new figures enlarging the font size.

Figure 1. This figure should be taller to have more space to plot the meteorological variables.

Figures 1, 3-7. What is the temporal resolution of the data plotted in these figure?

References.

EU: Combating climate change: The EU leads the way, EU report, 2007. http://reports.eea.europa.eu/eea_report_2006_7/en, access: June 2011.

Johansson, L. S., Leckner, B., Gustavsson, L., Cooper, D., Tullin, C., and Potter, A.: Emission characteristics of modern and oldtype residential boilers ïňĄred with wood

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logs and wood pellets, Atmos. Environ., 38, 4183–4195, 2004.

Krecl, P., Ström, J., and Johansson, C.: Diurnal variation of atmospheric aerosol during the wood combustion season in Northern Sweden, Atmos. Environ., 42, 4113–4125,doi:10.1016/j.atmosenv.2008.01.026, 2008.

Lanz, V. A., Prevot, A. S. H., Alfarra, M. R., Weimer, S., Mohr, C., DeCarlo, P. F., Gianini, M. F. D., Hueglin, C., Schneider, J., Favez, O., D'Anna, B., George, C., and Baltensperger, U.: Characterization of aerosol chemical composition with aerosol mass spectrometry in Central Europe: an overview, Atmos. Chem. Phys., 10, 10453–10471, doi:10.5194/acp-10-10453-2010, 2010.

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, Atmos. Chem. Phys., 9, 2891–2918, doi:10.5194/acp-9-2891-2009, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 11579, 2011.