

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.

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We would like to thank the referee for the constructive comments and suggestions made to improve the manuscript. Our responses to each comment are given below.

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

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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.

Response: We changed the title to “Diurnal variations of ambient residential particulate wood burning emissions and their contribution to the concentration of Polycyclic Aromatic Hydrocarbons (PAHs)”

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.

Response: The discussion on the diurnal pattern was reconsidered and rewritten. All weeks and weekends were compared individually regarding their meteorological conditions and aerosol concentration. Unpaired t-test analyses were also performed for each compound. In respect to the diurnal profiles of HOA, BBOA, OOA and PAH, the

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following discussion was added: "The diurnal profiles of the main organic components (OOA, BBOA and HOA) are shown in Fig. 6. As previously described for the main aerosol components, the diurnal profiles were divided in 3 weekdays and 3 weekends (Fig. 1) and similar conclusions could be drawn by comparing the different weeks and weekends patterns. The diurnal profile of week 2 differed from the profiles of weeks 3 and 4. While no clear diurnal variations of each OA factor was observed during week 2, even if it corresponded to the highest concentrations, a clear variation of BBOA, HOA and OOA could be observed during weeks 3 and 4. During these two weeks, tracers of fresh/primary organic aerosol (POA) emissions (i.e. BBOA and HOA) showed two maxima: a first around 8:00 and a second around 19:00. Their concentrations were slightly lower during day which may be attributed to a possible diminution of the emission as well as to a dilution effect by an increase of the mixed layer height. The evening peaks of BBOA and HOA were higher than the morning peaks, as previously observed by Lanz et al. (2010) and Sandradewi et al. (2008). Although this suggests larger emissions in the evening, the concentrations could also be enhanced by local meteorological conditions like a lower mixed layer height limiting the dilution process during the night. Interestingly, the OOA factor presented a profile similar to that of POA during week 4 which was less pronounced during week 3 and therefore confirms residential heating as a possible source of OOA. During weekends 1 and 2, an increase of BBOA and HOA concentrations during day time could be observed, reaching a maximum in mid-afternoon and then slowly decreasing to a minimum during the night with a small peak at 22:00. During these two weekends, the OOA concentrations followed a tendency similar to that of BBOA and HOA. Not surprisingly, these concentrations decreased during the day of weekend 3, as previously described for the total organics. As for total OA, comparisons of the hourly BBOA and HOA concentrations between weeks 3 and 4 as well as between weekends 1 and 2 were performed using unpaired t-test analyses. The differences of concentrations were not statistically significant. Although different tendencies could be observed between weekdays and weekends, these differences were small in term of concentration. This may result from the limiting sampling

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time as well as the fact that emissions from the local artisanal activity during weekdays were basically replaced by residential emissions during weekends, as suggested by the time switch observed between weekdays and weekends. The fact that HOA and BBOA had similar profiles confirms that HOA is correlated with residential heating rather than car exhaust. This is in agreement with the low car traffic mentioned above. Therefore, our results suggest that residential heating (BBOA and HOA) represented at least 37 % of the total OA at Seiffen during our measurement period.

[. . .]As done previously for the different NR-PM1 and organic components, diurnal variations of PAH concentrations in the different weeks and weekends were compared (Fig. 6). The difference between weeks is less pronounced than previously reported for HOA and BBOA. A morning and evening peaks were observed during all 3 weeks, being less pronounced during week 2. During night, the PAH concentration decreased near to zero except for week 2. In agreement with the larger influence of the long range transport of pollutants during this period, this suggests a higher background concentration. The PAH concentrations were highest during weekend 1. As in week 2, this certainly correlated with the influence of more polluted air masses. Similarly to the week comparison, the different PAH diurnal profiles of all weekends showed some similarities: the PAH concentrations started to increase later than during weekdays, reached a maximum before noon and decreased in the afternoon. Like for BBOA and HOA, the PAH concentrations also reached a peak at around 22:00, confirming the link between PAH and residential heating. In opposition to the statistical analyses performed before with BBOA and HOA, the difference of PAH concentrations at 07:00, 10:00, 12:00, 13:00 and 16:00 were statistically significant between weekends 1-2 and weeks 3-4. Taking together, the BBOA, HOA and PAH diurnal profiles do not suggest pronounced differences in terms of emissions between weekdays and weekends. However, the significant differences found between the PAH concentrations during weekdays vs. weekends suggest differences in the burning conditions. As wood combustion emissions strongly depend on the burning conditions (Johansson et al., 2004), this difference in PAH concentration and not in HOA and BBOA might be interpreted as resulting from the

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difference in aerosol composition emitted from residential heating systems and bigger heating systems used by the artisanal factories. Nevertheless, further measurements are necessary to confirm this hypothesis.”

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 mentioned 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).

Response: The authors agree that more details on the PMF analysis are needed. In order to provide a detailed description of the PMF analysis, the section “PMF analysis” was renamed in “factor analysis” and split in two subsections. The following details on the PMF analysis were added: “The analysis of the organic aerosol components was conducted with the unit mass resolution (UMR) organic mass spectra (from m/z 12 to m/z 100) and was performed using the Positive Matrices Factorization method (PMF) developed by Paatero and Tapper (1994) and according to the PMF evaluation Tool (PET), a custom software for solution comparison and analysis, developed by Ulbrich et al. (2009). Prior to the analysis, the organic mass spectra matrix and its corresponding error matrix were determined and the error matrix was corrected for minimum error and low signal-to-noise ratio (see Ulbrich et al., 2009). The contribution of the CO₂ ion to fragments m/z 16, 17, 18 and 44 were downweighted following the procedure described by Ulbrich et al. (2009). Then, PMF analysis was run using 1 to 5 factors. The sensitivity to matrix rotation was explored by varying the f_{Peak} from -1 to +1 with steps of 0.1. The uncertainty of the solution (Seed) was investigated using 100 random starting points. Finally, the quantitative uncertainty of the identified factors was estimated by 100 bootstrapping runs. The 2 factors solution separates Oxygenated

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Organic Aerosols (OOA) from a mixture of primary organic aerosol (POA) with high contributions of masses m/z 55, 57 and 60 ($Q/Q_{\text{exp}} = 5.16$). The 3 factors solution splits the POA into Hydrocarbon-like Organic Aerosol (HOA) and Biomass-Burning Organic Aerosol (BBOA) ($Q/Q_{\text{exp}} = 4.35$). The 4 factors solution additionally splits the OOA ($Q/Q_{\text{exp}} = 3.59$) and the 5 factors solution results from further splitting of the OOA ($Q/Q_{\text{exp}} = 2.94$). The 3 factors solution was retained after having investigated the f_{Peak} and Seed influences on the resulting time series and mass spectra as well as the differences between the solutions in the mass spectra and time series. The final solution was obtained for a f_{Peak} value of 0.0 which was used for Seed and bootstrap investigations. The Q/Q_{exp} and the factors obtained for different f_{Peak} values were nearly identical. The retained solution was stable over the different starting points tested with relatively low uncertainties (Fig. SI 2). The factor identification was performed by comparing the time series and mass spectra of each factor with external tracers (nitrate, sulfate, potassium, chloride, C₂H₄O₂⁺, . . .), available gas phase measurements (NO_x and CO), off-line filter results, and reference source mass spectra available on the AMS MS database (<http://cires.colorado.edu/jimenez-group/AMSsd/index.html>) (Ulbrich et al., 2009). Finally, the 3 factors solution was preferred to the 4 factors solution principally due to the uncertainties regarding the identification of the second OOA factor in the 4 factors solution”.

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 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

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wood combustion emission because it is contributing to two different factors, you should inform the reader on this problem.

Response: We agree with the comment made by the referee. We admit that the identification of the SV-OOA was not justified. Therefore, we decided to consider only the 3 factors solution corresponding to BBOA, HOA and OOA.

The description of the different factor identification was rewritten as follows: "The corresponding BBOA factor was identified by comparing its factor mass spectrum with that one published by Lanz et al. (2007) (Fig. SI 3). The identified BBOA factor is also characterized by a large contribution of the organic fragments m/z 60 and m/z 73 which are considered as tracers of biomass burning aerosols (e.g. Alfara et al., 2007; Schneider et al., 2006). In the identified BBOA factor, the organic fragments m/z 60 and m/z 73 represented 4.2% and 1.7% of the total BBOA mass spectra, respectively. The contribution of m/z 60 is slightly higher than the reported values of 2-4% in Switzerland (Lanz et al., 2010). The fragment m/z 44 represented 3.3% of the total BBOA mass spectra. Based on the relation between the contribution of m/z 44 and the O/C ratio reported by Aiken et al. (2008) and Sun et al. (2009), a O/C ratio of 0.20 was obtained for the BBOA factor which is in agreement with previously reported values (Aiken et al., 2008; He et al., 2011). During the measurement period, the fragment $C_2H_4O_2^+$ represented 90% of the total m/z 60 and not surprisingly, the time series of the BBOA factor were strongly correlated to the $C_2H_4O_2^+$ (Fig. 3 and Fig. SI 4, $r^2 = 0.77$) as well as the m/z 73 fragment $C_3H_5O_2^+$ (Fig. SI 4, $r^2 = 0.93$). A small discrepancy between $C_2H_4O_2^+$ and BBOA could be observed once in a while which might be interpreted as a contribution of an organic compound other than BBOA to $C_2H_4O_2^+$. However, this was not the case for $C_3H_5O_2^+$. Potassium which can be emitted by biomass burning and was used as an inorganic biomass burning tracer (e.g. in Lee et al., 2005; Hudson et al., 2004) also presented a similar profile to the BBOA (Fig. 3 and Fig. SI 4, $r^2 = 0.83$). Both comparisons using reference mass spectra and comparisons using tracers confirmed our identification. Although the BBOA mass spectra showed similarities to

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mass spectra of pure levoglucosan, the comparison of BBOA concentration with the total concentration of MA measured on filters samples revealed that BBOA corresponds to the sum of MA (slope 1.14, $r^2 = 0.89$, Fig. 4) rather than to levoglucosan alone (slope 0.5, $r^2 = 0.65$). Therefore, the identified BBOA factor can be considered as a good tracer for the total amount of MA.

The identified HOA factor, commonly considered as corresponding to primary organic aerosols (fossil fuel combustion), had a mass spectrum similar to previously reported HOA factors (Lanz et al., 2007; Ulbrich et al., 2009) (Fig. SI 3). The HOA mass spectrum was also characterized by a large contribution of the fragment m/z 57 (3.6 % of the total HOA signal). A O/C ratio of 0.15 was estimated from the contribution of m/z 44 to the HOA mass spectra which agrees with previously reported values in urban areas (Aiken et al., 2008; He et al., 2011, Huang et al., 2011), but is higher than the reported O/C ratio of motor vehicle exhaust (0.03-0.04; Mohr et al., 2009). Compared to previous studies, the HOA time series correlated stronger with CO ($r^2 = 0.47$, Fig. SI 4) than NOx ($r^2 = 0.38$, Fig. 3 and Fig. SI 4), as previously reported (Lanz et al., 2010; Ulbrich et al., 2009; Zhang et al., 2005a). Unfortunately, no parallel online measurements of black carbon were performed and the daily averaged HOA concentration did not correlate with daily PM1 Elemental Carbon (EC). The absence of a linear relationship between HOA and PM1 EC could be explained by the low number of filter samples and the uncertainties in the EC measurements. As previously mentioned, wood combustion for domestic heating is used in approximately 30% of the houses in Seiffen. Therefore, other heating energy sources like liquid fuel have also to be considered (Saxony EPA agency data, Löschau, 2007). Consequently, during the cold period, heating systems using wood combustion or liquid fuel for domestic heating are emitted simultaneously. Thereby, two possible sources of the HOA factor have to be considered: a) vehicle exhaust and b) emissions from domestic heating. Considering a limited car traffic at Seiffen (less than 2000 vehicles per day, Saxony EPA agency data, Löschau, 2007), traffic emission should have a limited contribution to the HOA. The correlation between HOA and BBOA ($r^2 = 0.62$, Fig. SI 4) confirms the suggested

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contribution of the residential heating to the HOA. Moreover, a direct contribution of the biomass burning emissions to HOA cannot be completely excluded. For example, DeCarlo et al. (2010) reported a contribution of biomass burning to HOA of 49% over the region of Mexico, using a CO source apportionment model. Unfortunately, such information was not available and due to the limited gas phase measurements performed here, it is not possible to clearly attribute a quantitative contribution of biomass burning and liquid fuel combustion to the identified HOA factor. Nevertheless, it seems reasonable to assume that residential heating (independently of the energy) represents the largest contribution of HOA.

The last factor had mass spectra similar to the mass spectra of oxygenated organic aerosol (OOA) reported by Zhang et al. (2005b) (Fig. SI 3) and to the reference mass spectra of fulvic acid (Fig. SI 3) which is considered to be a model substance for Humic Like Substances (HULIS). The mass spectra of the OOA factor was characterized by a larger contribution of the fragments m/z 44 (19%) and m/z 43 (11%) which is in agreement with the previously reported values (e.g. Lanz et al., 2010). As done previously, the O/C ratio (0.8) was estimated according to the contribution of the mass m/z 44 which is in agreement with previous measurements (Aiken et al., 2008; He et al., 2011; Ng et al., 2010) as well as with the presence of more oxygenated organic compounds in this factor compared to BBOA and HOA. The time series of the OOA factor correlated stronger with SO_4 ($r^2 = 0.68$, Fig. SI 5) than with NO_3 ($r^2 = 0.50$, Fig. SI 5). Based on the literature (Lanz et al., 2010; Ulbrich et al., 2009; Zhang et al., 2005a) this might be explained by long range transport or regional aerosols. Nevertheless, the OOA factor also correlated with CO ($r^2 = 0.61$, Fig. SI 5) which suggests a possible combustion origin. The emission of high molecular weight compounds during biomass burning was previously reported by several authors (e.g. Hoffer et al., 2006; Mukai and Ambe, 1986; Mayol-Bracero et al., 2002; Lin et al., 2010; Weimer et al., 2008). During their laboratory investigations, Weimer et al. (2008) observed a strong similarity between the organic mass spectra of wood burning emissions and the fulvic acid mass spectra, especially during the smoldering phase. Consequently, taking into

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account the sampling site environment leads to the assumption that at least a part of the OOA factor should have a direct wood combustion origin. However, relying on the available parallel measurements only, a quantified distinction between the long range transport OOA and the wood combustion related OOA was not possible.

Moreover, a part of the OOA might also be linked to the aging of the wood combustion emissions which leads to the formation of less volatile organics aerosols, for example due to photo-oxidation processes during day time (Capes et al., 2008; Grieshop et al., 2009a; Grieshop et al., 2009b). In addition, taking into account the high relative humidity during measurement period and regarding laboratory and simulation studies of the levoglucosan oxidation by OH-radical made by Hennigan et al. (2010) and Hoffmann et al. (2010), the aging of BBOA on deliquescence particles can be expected. The presence of SOA coming from the OH-oxidation of wood combustion VOCs during measurement periods was demonstrated by Iinuma et al. (2010). Indeed, using the PM1 filter measurements made during the entire project (i.e. from October 2007 to March 2008) and additional laboratory measurements in the simulation chamber, Iinuma et al. (2010) correlated the presence of methyl-nitrocatechols with the photooxidation of m-cresol directly emitted by wood combustion. In our study, the high resolution analysis of the AMS mass spectra showed a similar profile between the $C_xH_yO_zN_w+$ fragments (with z higher than 1) and the previously identified BBOA factor, besides a good correlation with identified methyl-nitrocatechols (Fig. 5). This is in agreement with the observation made by Iinuma et al. (2010). It is therefore reasonable to conclude that during our measurements, the $C_xH_yO_zN_w+$ fragments were related to methyl-nitrocatechol issues of the oxidation of m-cresol. This might also be considered as a possible source of the observed OOA factor. The distinction between the different sources of OOA appears to be challenging in our conditions. It was not possible to clearly distinguish the wood burning contribution from the long range transport. Hence, the OOA factor represented a mixture of these two main sources. The mean contributions of OOA, BBOA and HOA to total organics were approximately 62%, 20% and 17%, respectively. The contributions of BBOA and HOA are in the range of

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values observed by Lanz et al. (2010) in several places in the Alpine mountains (17-49% and 6-16% of OA for BBOA and HOA, respectively), but slightly lower than the values previously observed by Favez et al. (2010) (BBOA = 37%)”.

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.

Response: It is true that the short events of high PAH concentrations were considered to correspond to a plume of a chimney exhaust of adjacent houses that due to a small change in wind direction moved to our sampling line. However, this statement did not consider that it is plume from wood burning emissions only. Therefore, we linked the high PAH concentration events to residential heating plume rather than pure wood burning emission. The value of the PAH/BBOA ratio of 0.03 was considered based on the slope between total PAH and total MA measured on PM1 filters during the measurement periods ($r^2 = 0.80$) which is in the range of the different values reported in the literature. Therefore, we considered the “red” point in our Fig. 7 as not correlated to wood burning emissions. Additionally, it was not possible to subdivide the “red”

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sample in two groups using different PAH/BBOA ratios. However, the “red” points did not systematically correspond to the period of high PAH concentrations. Based on the estimated contribution of BBOA to wood burning PAH emission, the PAH-wood burning concentration was calculated and compared to the measured PAH concentration. Consequently, considering the high PAH event as coming from liquid fuel emission only should lead to an under-estimation of the contribution of wood burning to total PAH.

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.

Response: It is true that the AMS results were compared to a limited number of filter samples. The following sentence was added in section 2.2.2.: “. Although the comparison between AMS and daily PM1 filters was successfully performed for different chemical compounds (see discussion below), it is important to mention that the precision of the resulting linear regression slopes is limited by the restricted number of filter samples.”

(B) Minor comments 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.

Response: The following details on Saxony’s wood consumption in the last years were added: “in the state of Saxony (Germany), the number of wood stoves (< 15 kW) increased by a factor close to 8 between 2002 and 2006 and the consumption of wood for residential heating was estimated around 8000 TJ in 2007 (Hausmann, 2010), illustrating the renewal of interest for this energy.”

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).

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Response: The following sentence was added “This is part of a global policy to reduce the human carbon footprint. Since the Kyoto Protocol has been accepted in 1997, the European Union intends to reduce its greenhouse gas emissions by at least 20% between 1990 and 2020 (EU, 2007). One approach employed to reach this goal is to promote wood combustion.”

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.

Response: The sentence was changed as follows “Several components of the wood combustion particles (e.g. PAH) are considered to be carcinogenic and mutagenic agents”.

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.

Response: We added the reference to Löschau, 2007.

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?

Response: The sentence was modified as follows: “To limit contribution of direct traffic emissions, the AMS was sampled at around 200 m away from the main street of the village”.

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

Response: Seiffen is a village with approximately 3000 inhabitants. This information was added in the text.

Section 2.1, page 1583: Add some words on the fuel share for space heating in resi-

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dential areas in Seiffen.

Response: At Seiffen, wood burning represents 32% of the smaller heating systems (less than 50kW) and 25% of the larger ones (up to 500kW). This was added in the presentation of Seiffen (section 2.1).

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

Response: Details on the meteorological station were added in a new subsection entitled “Other measurements”. The following description was added “Standard meteorological variables (Temperature, RH, atmospheric pressure, wind direction, and wind velocity) were also measured continuously (time resolution 30 min). The meteorological station was located near the Digital filter samplers and the measurements were performed on a mast of 8 m height which was still under roofs level of the surrounding houses.”

Section 2.2.1, page 1583, lines 21-22: Replace “January 10” by “10 January”.

Response: corrected

Add details on the cut-off size and sampling frequency of the AMS instrument.

Response: We added the following sentence: “The AMS is commonly considered to provide non-refractory near PM1 (NR-PM1) aerosol mass concentration (Canagaratna et al., 2007). During the measurement period, the AMS was sampled at a typical time resolution of 5 min in “V” mode collecting approximately 6700 samples (4900 during weekdays and 1800 during weekends).”

Include details on the CO and NO_x measurements conducted at the site and used in Figure 3.

Response: The following details were added: “In addition, standard gas phase measurements of NO_x and CO (Monitorlabs ML 9830) were performed from the same sample place as the weather station with a time resolution of 30 min. “

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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.

Response: To include the requested information, the sentence was modified as follows: "Every 4 days and during 24h from midnight to midnight (total sampling volume 720m³), PM₁ particles were sampled using a Digitel DHA-80 high volume sampler (Digitel Elektronik AG, Hegnau, Switzerland) and collected on pre-heated 15 cm diameter quartz fiber filters (105°C, 24h and then minimum 48h at 20°C and 50% RH) (Munktell Filter AB, Falun, Sweden)".

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

Response: OC and EC correspond to Organic Carbon and Elemental Carbon, respectively. The sentence was changed to "Organic carbon to Elemental carbon ratios (OC/EC) were determined [...]."

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.

Response: It is well known that artifacts can have a large influence on the EC/OC measurements. To minimize these uncertainties, blank measurements were performed. During the Seiffen project, the mean blank values (and the standard deviations) of 8 laboratory measurements were $8.90 \pm 0.87 \mu\text{gC}$ for OC and $1.14 \pm 0.3 \mu\text{gC}$ for EC. The detection limits were $0.31 \mu\text{gC m}^{-3}$ for OC and $0.1 \mu\text{gC m}^{-3}$ for EC. This was added in the manuscript.

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

Response: corrected

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.

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Response: The AMS is usually considered to provide a near-PM₁ chemical composition. As the instrument cannot detect soot, crustal material and sea-salts, the total mass concentration provided by the AMS corresponds to the non-refractory PM₁. The PM₁ notation was corrected to NR-PM₁.

Section 3.1, page 11585, lines 23-25: Besides classifying the temperature inversion as light, report the temperature gradient.

Response: the text was changed to "The temperature inversion corresponds to a temperature at Seiffen 0.5°C colder than the expected neutral stratification between Seiffen and Schwartenberg".

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?

Response: The results presented in Table 1 correspond to the averaged Digitel and AMS values calculated over the entire AMS measurement periods. However, prior to any direct comparison of both instruments, the AMS data were averaged to match the sampling duration of the filter samples (i.e. averaged for each filter sampling day, from midnight to midnight). This approach was used for comparisons made in Figures 4, 5, 6 and SI-1. The following sentence was added in section 2.2.1 "Prior to any comparison between the AMS results and the PM₁ filters, the AMS data were averaged to match the sampling duration of the filter samples." Additionally, to avoid any confusion, we added the averaged AMS concentrations that corresponded to the exact filter sampling periods in table.1

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

Response: Based on the aerodynamic lens transmission efficiency, the AMS is referred

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to as a near PM1 instrument (Canagaratna et al., 2007) and the size cut of the AMS is considered to be similar to that of typical PM1 cyclones. Therefore, it is completely relevant to directly compare the AMS results with the PM1 filter samples.

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

Response: The following discussion was added: "The OM/OC ratio was calculated according to the elemental analysis method developed by Aiken et al. (2007) and Aiken et al. (2008). In agreement with the dominant primary emissions, a mean value of 1.56 ± 0.13 was obtained during the measurements. As our filter samples provided directly measurements of the OC concentration and as the AMS provided the OM concentration, using both instruments allowed us to estimate the OM/OC ratio, too. Using this method, an OM/OC ratio of 1.93 was obtained. The difference of these two ratios (1.56 vs. 1.93) might be explained by the uncertainties of each method as well as the limiting number of filter samples".

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.

Response: The notation was changed.

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.

Response: The conclusion on the diurnal profiles comparison was rewritten as follows: "Taking together, the BBOA, HOA and PAH diurnal profiles do not suggest pronounced differences in terms of emissions between weekdays and weekends. However, the significant differences found between the PAH concentrations during weekdays vs. weekends suggest differences in the burning conditions. As wood combustion emissions

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strongly depend on the burning conditions (Johansson et al., 2004), this difference in PAH concentration and not in HOA and BBOA might be interpreted as resulting from the difference in aerosol composition emitted from residential heating systems and bigger heating systems used by the artisanal factories. Nevertheless, further measurements are necessary to confirm this hypothesis".

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

Response: corrected

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

Response: The conclusion on the diurnal profiles comparison was rewritten as follows: "Taking together, the BBOA, HOA and PAH diurnal profiles do not suggest pronounced differences in terms of emissions between weekdays and weekends. However, the significant differences found between the PAH concentrations during weekdays vs. weekends suggest differences in the burning conditions. As wood combustion emissions strongly depend on the burning conditions (Johansson et al., 2004), this difference in PAH concentration and not in HOA and BBOA might be interpreted as resulting from the difference in aerosol composition emitted from residential heating systems and bigger heating systems used by the artisanal factories. Nevertheless, further measurements are necessary to confirm this hypothesis".

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.

Response: During the 22 days of the campaign, the AMS collected approximately 6700 samples (4900 during weekdays and 1800 during weekends). AMS data were analyzed using the standard data analysis software packages (Squirrel version 1.49 and PIKA version 1.08), downloaded from the TOF-AMS-Resources webpage (<http://cires.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/>). The quan-

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titative results were calculated using the following relative ionization efficiency (RIE) values: 1.4 for organics, 1.2 for sulfate, 1.1 for nitrate, 1.3 for chloride, and 4 for ammonium (Canagaratna et al., 2007). The PMF analysis was performed using the matrixes (organic mass spectra matrix and the corresponding error matrix) obtained from Squirrel in the range of 12 to 100 m/z. This was added in the text.

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.

Response: No linear relationship was observed between the daily averages of HOA and EC measured from the filter. This can be explained by the low number of filter samples as well as the uncertainties of the EC measurements. The following text was added: "Unfortunately, no parallel online measurements of black carbon were performed and the daily averaged HOA concentration did not correlate with daily PM1 Elemental Carbon (EC). The absence of a linear relationship between HOA and PM1 EC could be explained by the low number of filter samples and the uncertainties in the EC measurements".

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

Response: The sentence was corrected as follows: "Consequently, during the cold period, heating systems using wood combustion or liquid fuel for domestic heating are emitted simultaneously"

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 modeled series.

Response: The correlation plot between HOA and BBOA ($r^2 = 0.62$) was added.

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

Response: OOA-I corresponds to the oxygenated-like organic aerosol type I, as defined by C7355

defined by Ulbrich et al. 2009.

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

Response: OOA-II corresponds to the oxygenated-like organic aerosol type II, as defined by Ulbrich et al. 2009.

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)"?

Response: We replaced "the authors" by "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.

Response: The sentence was rewritten as follows: "Although this suggests larger emissions in the evening, the concentrations could also be enhanced by local meteorological conditions like a lower mixed layer height limiting the dilution process during the night."

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 et al., 2008).

Response: We agree that the comparison was wrong. Therefore, the discussion was removed and our conclusion was rewritten as follows: "Taking together, the BBOA,

HOA and PAH diurnal profiles do not suggest pronounced differences in terms of emissions between weekdays and weekends. However, the significant differences found between the PAH concentrations during weekdays vs. weekends suggest differences in the burning conditions. As wood combustion emissions strongly depend on the burning conditions (Johansson et al., 2004), this difference in PAH concentration and not in HOA and BBOA might be interpreted as resulting from the difference in aerosol composition emitted from residential heating systems and bigger heating systems used by the artisanal factories. Nevertheless, further measurements are necessary to confirm this hypothesis”.

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

Response: We added the following references: Rogge et al., 1998; Dhammapala et al., 2007; Mandalakis et al., 2005; Kozinski and Saade, 1998.

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.

Response: We added the following sentence “due to the limiting time measurements (4 weeks), our results cannot be considered as representative for all winter conditions. Longer campaigns including on-line and daily filter sampling taking every day to increase the number of samples are needed for better statistical analyses.”

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

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(see comment Section 3.2.1, page 11589, lines 25-29).

Response: The sentence was changed as follows “Although residential wood combustion emissions strongly depend on burning conditions, type of wood (e.g. Johansson et al., 2004), and/or local meteorological conditions, the promotion of this source of energy by the local policy also contribute to increase the impact of residential wood burning on local air quality.”

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

Response: OM corresponds to organic mass. We changed the sentence as follows: “The PM1 organics corresponds to the organic carbon (OC) and the AMS organics corresponds to organic mass (OM)”. Numbers on the table are mean values ± standard deviation. The table title was changed to “Summary of the mean concentrations (± standard deviation) measured on PM1 filter samples and AMS (using the same sampling time and days than PM1 (Mass synchronized to PM1) and for the entire campaign).”

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

Response: The font sizes of the legends and the labels of the figures were enlarged.

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

Response: Fig. 1 is corrected

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

Response: In all figures, the presented AMS results have a time resolution of 5 min and the gas measurements have a time resolution of 30 min. Precisions of the time resolutions of each instrument are now added in the Figure labels.

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<http://www.atmos-chem-phys-discuss.net/11/C7337/2011/acpd-11-C7337-2011-supplement.pdf>

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

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