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Atmospheric
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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.

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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.

Poulain et al. paper presents a comparison of on-line and off-line measurements on NR-PM1 aerosol in a mid-level mountain area in Saxony (Germany). The source apportionnement study is performed using HR-TOF-AMS data and PMF analysis together with tracers from filters. The main focus is in the impact of residential wood burning and PAHs in the NR-PM1 fraction. The work is based on a classic approach of PMF analysis and external tracers. There is an interesting comparison between off-line and on-line data (as total monosaccharide anhydrides and methyl-nitrocathecols). The experimental section and the PMFanalysis are briefly treated. More details are needed in those two sections. The written English is quite poor in some paragraphs and should be revised. A good number of typos should be corrected (end of my comments).

General comments and questions:

1) More details are needed in 2.2.1 section. Page 11584 line 12-15. The ambient humidity was on average 89%. This does not mean that the same RH is found in the sampling line and before the AMS orifice. Which is the residence time of the particles in the sampling line, which is the humidity just before the inlet? This is important in choosing CE values. The RH can highly change if the sampling flow is slow and the outdoor and indoor temperatures are very different. Please comment on this and add details.

Response: It is true that the RH in the sampling line did not correspond to the outside value. The sampling line was not dried during the present campaign, the outside RH was around 90% most of the time and the instrument was installed in a room at around 16°C. Therefore, based on the difference of temperature between outside and inside, the lowest values of relative humidity could be expected to range between 30 and 40% in the sampling line. The particles do not instantaneously reach the equilibrium with the RH. Several seconds are needed. For example, based on the comparison of 6 different HTDMAs, Duplissy et al. (2009) suggests that particles have to be dried at least 10s at 15% RH before entering to the first DMA. In our sampling line, the residence time was estimated to be around 10s. Consequently, the presence of water on the particles could be expected, suggesting a CE value near 1. This was added in the text.

2) More details are needed in the PMF section. They are presented without any type of quality details (Q/Qexp, errors matrix calculation and so on). There is a total lack of

discussion about the many possible solutions from PMF analysis. How many FPEAKs and SEEDs have been tested? Which solutions have been chosen and why? Need of some critical discussion about the many possible PMF solutions.

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 “PMF analysis” and “Factor identification”.

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 fPeak 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 Organic Aerosols (OOA) from a mixture of primary organic aerosol (POA) with high contributions of masses m/z 55, 57 and 60 (Q/Qexp = 5.16). The 3 factors solution splits the POA into Hydrocarbon-like Organic Aerosol (HOA) and Biomass-Burning Organic Aerosol (BBOA) (Q/Qexp = 4.35). The 4 factors solution additionally splits the OOA (Q/Qexp =3.59) and the 5 factors solution results from further splitting of the OOA (Q/Qexp = 2.94). The 3 factors solution was retained after having investigated the fPeak 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 fPeak value of 0.0 which was used for Seed and bootstrap investigations. The Q/Qexp and the factors obtained for different fPeak 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 (NOx 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".

3) Table 1 and figure 1. NR-PM1 sulphate is 26% and nitrate 21%. This means that they equally contribute to the inorganic PM1 loading. Sulphate is not dominating the inorganic fraction as the authors state.

Response: We changed the sentence in the following way: "The compositions of the NR-PM1 mass concentration mainly contained organic (OA) aerosol (40%). The mass fraction of sulfate, nitrate, ammonium, and non sea-salt chloride were 26%, 21%, 12%, and 1%, respectively".

4) Figure 2. The section about diurnal profiles is quite confusing and not convincing. Organic loading is almost the same during working days and week-ends from fig2 left plots. I would expect some difference, since human activities are enhanced during the working days. Your BBOA with higher values in the early morning and late evening should not be connected to residential wood-burning instead of wood decoration factories, unless those factories have highest productivity at 8h00 and 19h00-20h00. If HOA is associated to both vehicle exhaust and residential heating, should it be possible to differentiating the two using different tracers correlation plot? (ex. NOx, CO and so on. See DeCarlo, ACP, Milagro, 2010). Which is the time series correlation (regression

plot) between BBOA and HOA data? Could you use this to separate the two HOA contributions? How can the authors state that m-cresol could be related to SV-OOA when they do not measure it at all? You measure only CxHyOzNw which is associated with BBOA.

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. With respect to the profiles of HOA, BBOA, OOA and PAH, the 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 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 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."

It was not possible to separate the HOA fraction associated to biomass burning from the fraction resulting from liquid fuel combustion. This is now mentioned in the text: "The correlation between HOA and BBOA (r^2 = 0.62, Fig. SI 4) confirms the suggested 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".

We agree that the discussion on the contribution of wood burning aging and SV-OOA was not fully relevant. Due to the few arguments available to properly justify the presence of two OOA (LV-OOA and SV-OOA), we decided to consider the 3 factor solutions only. "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". The discussion on the contribution of CxHyOzNw to OOA was modified

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as follows: “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 CxHyOzNw+ fragments (with z higher than 1) and the previously indentified 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 CxHyOzNw+ 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.”

5) Since a HR-TOF AMS allows evaluating O/C ratio why the authors did not presented them for the different factors and compared to published data?

Response: The PMF analysis was performed using the unit mass spectra resolution. However, based on the correlation between the fragment m/z 44 and the O/C ratio reported by Aiken et al. (2008) and Sun et al. (2009), an estimation of the O/C ratio was done for each PMF factor. The following discussion was added:” 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). [. . .]A O/C ratio of 0.15 was estimated from the contribution of m/z44 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 2010), but is higher than the reported O/C ratio of motor vehicle exhaust (0.03-0.04; Mohr et al., 2009). [. . .]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 aerosol in this factor compared to BBOA and HOA."

6) P. 11588 last paragraph: the authors state that OH chemistry can explain the SVOOA. If OH is processing semi-volatile species you should see a diurnal cycle of this fraction and a SVOOA type material arising in the middle of the day (if you had enough photochemistry and OH around). It is not the case during working days. How much Ox (O3+NOx) and photolysis were present in March? The discussion and diurnal pattern of LVOOA do not confirm this. You say that is it probably produced during burning processes and this is why it has the same time evolution of BBOA. I think there is some contradiction between what you show in fig. SI2, what you say in p. 11588 and 11590.

Response: We agree that the discussion on the SV-OOA factor was rather confusing and we have to admit that we don't have enough arguments to completely justify the presence of LV-OOA and SV-OOA and their link with aerosol aging (for example, no ozone measurement was performed at Seiffen, so it is not possible to estimate Ox). Therefore, we decided to limit our PMF analysis to 3 factors, means to consider total OOA only. Moreover, the following discussion on the expected contribution of wood burning aging aerosol to OOA was added: "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 CxHyOzNw+ fragments (with z higher than 1) and the previously indentified 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 CxHyOzNw+ 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.

7) P. 11591 line 16-27. If the PAHs arise from residential heating why they exhibit 20-45 minutes short-term events? Heating a house should take longer? More details are needed.

Response: We agree that heating a house takes longer than 20-45 minutes. We did not want to state that the events of high PAH concentrations corresponded to the duration of a house heating. We rather considered that the PAH peaks might be attributable to a period during which the measurement station was influenced by a plume of adjacent houses that due to a small change in wind direction moved to our sampling line. Unfortunately, as the wind direction was measured below the roof level, with a time resolution of 30 min only (see section 2.2), it was not possible to attribute these peaks to a precise house/source. This was added in the text.

8) P. 11592 line 16. How did the authors decided that PAH/BBOA=0.03 is a good ratio for separating PAHs from wood burning and other PAHs sources?

Response: Different aspects lead to the decision that PAH/BBOA = 0.03. As mentioned in the manuscript, the literature reports a large range of values for the PAH/BBOA ratio. According to this and regarding the slope of 0.03 (r^2 = 0.80) between total PAH and total MA obtained during the measurement period from the filters, a setting value of PAH/BBOA = 0.03 was considered to separate PAH directly related to wood combustion emissions. This is now mentioned in the text.

Small details and typos: 1. Table 1 there must be an error in the total PM1 filters the sum should be 5.15 and not 15.18 as written in the first line.

Response: This is not a typing error. The total mass measured by filters was effectively higher than the total AMS mass. The following discussion was added: “However, the total masses provided by the two instruments differed strongly. A possible explanation for this discrepancy is the presence of AMS refractory compounds, e.g. black carbon. EC, potassium, calcium, magnesium, and sodium concentrations measured by filter samples represented approximately 1 μg m-3 only and therefore cannot explain the entire difference. Another reason might be the presence of water in the filters that artificially enhances the total mass concentration. It is extremely difficult to completely subtract this water contribution from the total particulate mass measured in the filters, even if the filters were conditioned for a minimum duration of 48h at 20°C and 50% RH before being weighed”

2. P. 11586. “Two-thirds of weekend appended during lower particle concentration periods” should be rephrased. Append=add. I guess the authors wanted to say “happen or occur”.

Response: corrected

3. Page 11587. Line 9-11. It is a little confusing. I will rephrase it as follows “ the corresponding BBOA factor was identified by comparing its factor mass spectrum with the one published by Lanz et al 2007 (Fig. S2 in the Supplement) and with the time series of the AMS-HR K+ signal (Fig.3)”

Response: The description of the BBOA factor was rewritten as follow: “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. Alfarra 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 C2H4O2+ represented 90% of the total m/z 60 and not surprisingly, the time series of the BBOA factor were strongly correlated to the C2H4O2+ (Fig. 3 and Fig. SI 4, r^2 = 0.77) as well as the m/z 73 fragment C3H5O2+ (Fig. SI 4, r^2 = 0.93). A small discrepancy between C2H4O2+ and BBOA could be observed once in a while which might be interpreted as a contribution of an organic compound other than BBOA to C2H4O2+. However, this was not the case for C3H5O2+. 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”.

4. P. 11587 line 28: “ using wood burning and liquid fuel for domestic heating are emitted at the simultaneously” delete “at the” in bold

Response: deleted

5. P. 11588. Take away a dot before Weimer et al.

Response: done

6. P. 11590 line 8. Rephrasing suggested “Most of the time HOA and BBOA profiles

correlate, suggesting that HOA is mostly correlated with residential heating".

Response: corrected as follows:" The correlation between HOA and BBOA (Fig. SI-4, $r^2 = 0.62$) confirms the suggested contribution of the residential heating to the HOA. Moreover, a direct contribution of the biomass burning emissions to HOA cannot be completely excluded."

7. P. 11590 line 15: take away the ; and put a comma before diurnal.

Response: corrected

8. P. 11592 line 28. Rephrasing suggested ". . .PAHwb was assumed to contribute to about 62% of the total particulate PAHs mass concentration".

Response: corrected as suggested

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