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particulate wood  
burning emissions**

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# Diurnal variations of residential particulate wood burning emissions and their contribution to the concentration of Polycyclic Aromatic Hydrocarbons (PAHs)

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## Abstract

Residential wood burning is becoming an increasingly important cause of air quality problems since it has become a popular source of alternative energy to fossil fuel. In order to characterize the contribution of residential wood burning to local particle pollution a field campaign was organized at the village of Seiffen (Saxony, Germany). During this campaign, an Aerosol Mass Spectrometer (AMS) was deployed in parallel to a PM<sub>1</sub> high volume filter sampler. The AMS mass spectra were analyzed using Positive Matrix Factorization (PMF) to obtain detailed information about the organic aerosol (OA). Biomass-burning organic aerosol (BBOA), Hydrocarbon-like organic aerosol (HOA), Low-volatility oxygenated organic aerosol (LV-OOA) and Semi-volatile oxygenated organic aerosol (SV-OOA) were identified. Additionally, Polycyclic Aromatic Hydrocarbons (PAH) were measured by the AMS and short term events of extremely high PAH concentration compared to the mean PAH value were observed during the whole measurement period. A comparison with the results from PM<sub>1</sub> filter samples showed that the BBOA factor and the AMS PAH are good indicators of the total concentration of the different monosaccharide anhydrides and PAH measured on the filter samples. Based on its correlation with CO and the low car traffic, the HOA factor was considered to be related to residential heating using liquid fuel. A clear impact of the week and weekend days on the diurnal profiles of the different OA components was observed. The weekdays were characterized by two maxima; a first one early in the morning and a stronger one in the evening; during the weekend days, the different OA components principally reached one maxima early in the afternoon. Finally, the PAH emitted directly from residential wood combustion was estimated to represent 1.5% of the total mass of BBOA factor and around 62% of the total PAH concentration measured at Seiffen. This result highlights the important contribution of residential wood combustion to air quality and PAH emissions at the sampling place which might have a significant impact on human health. Moreover, it also emphasizes the need for a better time resolution of the chemical characterization of toxic particulate compounds in order to provide more

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information about variations of the different sources through the days as well as to better estimate the real human exposure.

## 1 Introduction

Fine particulate pollution is suspected to directly affect mortality and morbidity (Pope, 2000; Ostro et al., 2007; Gurjar et al., 2010). Identifying the different sources of these fine particles is an important challenge in order to better estimate their impact on human health. It is well known that during winter time, residential wood combustion is an important local source of particles which directly influences local to regional air quality (Naeher et al., 2007). In the US, the particulate emission of residential wood combustion was estimated around  $1.1 \times 10^6$  t/year particulate matter (PM) (Nolte et al., 2001) and measurements made at different locations demonstrated that residential wood combustion can contribute significantly to the total PM mass fraction and/or organic aerosol (OA) fraction at rural and/or urban locations. For example, the contribution of residential wood combustion to total PM<sub>1</sub> organic fraction was estimated around 37–68% depending on the model used in the city of Grenoble (France, Favez et al., 2010), 35–40% in the urban background of Zurich (Switzerland, Lanz et al., 2010), 79% in the city of Prague (Czech Republic, Saarikoski et al., 2008), around 50% of the PM<sub>10</sub> mass in southwestern Germany (Bari et al., 2009), 10% of PM<sub>2.5</sub> in Vindingen (Denmark, Glasius et al., 2008) and 44–57% of the total mass concentration in wintertime in the size range 25–606 nm in Lycksele (Sweden, Krecl et al., 2008a). In the overview of central Europe measurements (Lanz et al., 2010), the estimated contribution of biomass burning aerosol to total PM<sub>1</sub> organic aerosol ranged from 17 to 49% during winter time.

For several years, wood combustion has been considered a good alternative to fossil fuel for domestic heating. A pragmatic reason is the increasing price of coal and oil. A more strategic motivation is increasing the interest in the development of renewable energy technologies to decrease greenhouse gas emission. Even though epidemiological

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5 observations demonstrated a direct contribution of wood smoke to respiratory systems (e.g. Bolling et al., 2009; Naeher et al., 2007; Zelikoff et al., 2002), the health effects of wood burning particles remain unclear. Although wood burning particles did not cause acute airway inflammatory responses in inhalation experiments (Sehlstedt et al., 2010), the authors also reported a protective adaptive response of the distal lung confirming that there are still many reasons to assume that wood combustion particles play a relevant role for indoor as well as outdoor particle exposure in areas where such combustion processes are relevant. As other combustion processes, wood burning emissions contain a variety of toxic pollutants and are thus expected to exert adverse effects upon human health. Different studies linked the contribution of wood combustion to an increase in troubles of the cardiovascular system and/or respiratory system (Naeher et al., 2007; Jalava et al., 2010). Aerosol particles from wood combustion are considered to be a carcinogenic and mutagenic agent (Boström et al., 2002; Lewtas, 2007). Nevertheless, the relation between particle emissions, composition and toxicity is extremely complex and there is a need to better understand the physicochemical properties of wood combustion smoke and to improve the estimation of their biological effects (Bolling et al., 2009; Naeher et al., 2007).

10 In the context of increasing residential wood combustion emissions and their possible effect on human health, an intensive field study was performed in the small town of Seiffen (Saxony, Germany) during the winter 2007/2008 (Müller et al., 2010). The goal was to evaluate the impact of residential wood combustion on the local aerosol concentration. During the winter time, wood combustion is commonly used for house heating in Seiffen and consequently, it is an important and significant source of aerosol particles. In this work, special attention is paid to the characterization of the organic particulate fraction and more specifically to the identification and quantification of the different monosaccharide anhydrides (MA) and polycyclic aromatic hydrocarbons (PAH) compounds. Measurements of MA and PAH were based on the analysis of 24 h high volume filter samples performed all 4 days. Additionally, in order to follow the diurnal variation with a high time resolution, an Aerodyne Aerosol Mass Spectrometer (AMS)

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was deployed in January 2008. In this investigation, the impact of residential wood combustion on local air quality was investigated regarding the AMS measurements and comparing with the analytical results from filter samples measurements performed simultaneously.

## 2 Instrumentation

### 2.1 Sampling station Seiffen

The sampling location is situated in the mid-level mountain area – *Erzgebirge* (50°38'50" N, 13°27'08" E and 647 m a.s.l.) around 50 km southwest of Dresden, Germany. The AMS instrument was installed in a non-heated room (mean temperature 16 °C) located at ground level in a house located in the centre of the village of Seiffen, at safe distance from the main street (around 200 m). The AMS was sampling through a PM<sub>2.5</sub> inlet around 2.5 m above ground level. It is important to mention that the ambient aerosol was not dried prior to its injection into the AMS. Additional filter sampling was performed in front of the house around 10 m away from the AMS sampling line. Meteorological parameters were measured on a mast of 8 m height, which was still below the level of the roofs of the surrounding village houses.

### 2.2 Instrumentation

#### 2.2.1 Real time particle characterization

In order to provide highly time-resolved measurements for chemical particle composition, an Aerodyne High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS, here simply referred to AMS, DeCarlo et al., 2006) was deployed from January 10 to 2 February 2008. Briefly, the AMS allows two alternative detection modes: one to measure the particle size distribution (PTOF-mode) and one to measure the chemical composition of the particle (MS-mode). Before detection by the time of flight mass

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spectrometer, the aerosol non-refractory (NR) components were flash-vaporized by impaction on a heated surface (600 °C) and the vapours were ionized by an electron impact ionization source at 70 eV.

The calculation of the AMS mass concentration required to estimate the collection efficiency (CE) of the instrument. The CE value is known to depend on several parameters (Huffman et al., 2005; Matthew et al., 2008), e.g. the nitrate content, particle acidity, particle phase (liquid or solid) and relative humidity (RH) in the sampling line. It is considered that in most of the cases, a CE of 0.5 represents a good assumption (e.g. Canagaratna et al., 2007). However, several authors have reported that CE can increase to 1 when the RH in the sampling line is high (Allan et al., 2004; Sun et al., 2010; Takegawa et al., 2009). As mentioned above, the sampling line was not dried during the present campaign and the outside RH was around 90 % most of the time. Therefore, an elevated RH in the sampling line and presence of liquid organic compounds were expected. Consequently, a CE value near 1 was expected. The actual comparison with the sulphate, nitrate and ammonium concentrations in PM<sub>1</sub> filters confirmed this hypothesis (see Fig. S11 in the Supplement).

## 2.2.2 Off-line measurements

Every 4 days and during 24 h from midnight to midnight, PM<sub>1</sub> particles were sampled using a Digital DHA-80 high volume sampler (Digital Elektronik AG, Hegnau, Switzerland) and collected on pre-heated quartz fibre filters (105 °C, 24 h and then minimum 48 h at 20 °C and 50% RH) (Munktell Filter AB, Falun, Sweden). After sampling, filters were conditioned at 20 °C and 50% RH for a minimum duration of 48 h before being weighed by a microbalance Mettler-Toledo (AT 261). The filters were then analyzed using Ion Chromatography (Metrohm System 690, Switzerland) for the main water soluble ions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup>) according to the analytical method of Brüggemann and Rolle (1998). OC/EC ratios were determined using a carbon analyzer type C-mat 5500 with a NDIR detector (Gnauk et al., 2008). In addition, a Curie Point Pyrolysis Gas Chromatography Mass Spectrometry (CPP-GC-MS, Neusüb

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et al., 2000) was used to identify and quantify the Polycyclic Aromatic Hydrocarbons (PAH), and a High Performance Anion Exchange Chromatography coupled to Pulsed Amperometric Detector (HPAEC-PAD) was employed to measure monosaccharide anhydrides wood smoke tracers (e.g. Levoglucosan) as previously described by Engling et al. (2006) and Iinuma et al. (2009).

### 3 Results

#### 3.1 AMS measurements overview

Figure 1 provides a basic overview of the local meteorological parameters (temperature, RH, wind direction and wind velocity) as well the mass concentrations of the main NR components (organics, nitrate, sulphate, ammonium and non sea-salt chloride). During the winter 2007/2008, the temperatures were generally mild, with temperatures rarely below 0°C even at Seiffen's altitude of 647 m. During the AMS measurements, the mean temperature was of  $0.9 \pm 3.7^\circ\text{C}$  ( $\pm 1\sigma$ ) with temperatures within a total range between  $-7.7$  and  $+9.6^\circ\text{C}$ . The ambient relative humidity had a mean value of  $89 \pm 10\%$ , with a total range between 37.7 and 100%. The wind blew mainly from two directions, either southeast or northwest at a mean wind velocity of  $2.02 \pm 1.16 \text{ m s}^{-1}$ , and within a total range between 0 and  $7.58 \text{ m s}^{-1}$ .

The AMS yielded a range of  $\text{PM}_1$  mass concentration between 2.6 and  $8.0 \mu\text{g m}^{-3}$ . The periods with the highest mass concentration were related to a combination of southeasterly winds and low temperatures, while periods with the lower mass concentrations were related to northwesterly winds and warmer temperatures (Fig. 1). These two cases corresponded to air masses of more continental and more maritime character, respectively. During the periods of high mass concentrations, a light temperature inversion could be detected between Seiffen and the higher-elevation Schwartenberg Mountain (952 m a.s.l.). The cool temperatures and the pollution trapping induced by the low winds and light temperature inversion are thought to be responsible for the local

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accumulation of the high mass concentrations.

From a composition point of view, the NR-PM<sub>1</sub> mass concentration contained mainly organic aerosol (OA) and sulphate aerosol (40 and 26%, respectively). The mass fraction of nitrate, ammonium and chloride were 21%, 12% and 1%, respectively (Table 1).

5 A clear difference of diurnal profiles of the NR-PM<sub>1</sub> components was observed between workdays and weekends (Fig. 2). During working days, organics presented two maxima: early in the morning around 08:00 and early in the evening between 19:00 and 20:00. Chloride concentration increased during the day and reached its maximum in the evening, slightly earlier than OA. Sulphate and ammonium presented relatively  
10 stable concentrations during the day whereas nitrate had a minimum concentration during day time. Compared to working days, the diurnal profiles of weekends differed significantly, showing lower concentrations for all compounds. Organics and chloride concentrations increased to a maximum at around 14:00 and then decreased, reaching a minimum during the night. Sulphate concentration strongly increased during the  
15 weekend whereas nitrate appeared to be the only compound with a similar trend during working and non-working days. The observed differences between working and non-working days could be linked to changes in human activity. During the week, morning and evening peaks might correspond to waking-up/going to work/coming back home; during the weekend, such activities might be reduced. Differences between working  
20 and non-working days could also be explained by the absence of activities in local wood decoration factories during the weekend and more certainly by the fact that two-thirds of weekends appended during lower particle concentration periods which directly affect the mean concentrations.

## 3.2 Organic aerosol composition

### 25 3.2.1 PMF analysis

The organic fraction of the NR-PM<sub>1</sub> particles was analyzed using factor analysis of AMS data (FA-AMS) developed by Lanz et al. (2007) and Ulbrich et al. (2009) and

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based on the positive matrices factorization (PMF) developed by Paatero and Tapper (1994). Four factors were identified to explain the organic signal; they corresponded to Biomass-burning organic aerosol (BBOA), Hydrocarbon-like organic aerosol (HOA), Low-volatility oxygenated organic aerosol (LV-OOA) and semi-volatile oxygenated organic aerosol (SV-OOA). The four factors were identified by the comparison of their mass spectra with reference mass spectra available on the AMS mass spectra database (<http://cires.colorado.edu/jimenez-group/AMSsd/index.html>) and by a comparison of their time series with time series of related compounds (Fig. 3).

The corresponding BBOA factor was identified by comparing its factor mass spectra with the mass spectra of wood combustion (Lanz et al., 2007) and its comparison with the high resolution potassium signal ( $r^2 = 0.33$ ) (Fig. S2 in the Supplement). Although the BBOA mass spectra showed similarities to mass spectra of pure levoglucosan, the comparison of BBOA concentration with the total concentration of MA measured on filters samples revealed that BBOA is more corresponding to the sum of MA (slope 1.14,  $r^2 = 0.89$ , Fig. 4) than to levoglucosan alone (slope 0.5,  $r^2 = 0.65$ ). The BBOA can thus be considered as a good estimate of the total amount of MA.

The HOA factor, commonly considered as corresponding to primary organic aerosols (fossil fuel combustion), has a mass spectrum similar to previously reported HOA factors (Lanz et al., 2007; Ulbrich et al., 2009). Comparing to previous studies, the HOA time series were weakly correlated to the time series of  $\text{NO}_x$  gas concentrations ( $r^2 = 0.33$ ) but better correlated to CO ( $r^2 = 0.43$ ). Unfortunately, no parallel online measurements of black carbon were performed and a comparison with daily  $\text{PM}_{10}$  Elemental Carbon (EC) was not conclusive.

In the small city of Seiffen, wood combustion for domestic heating is used in 30% of all houses; and other heating energies 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 at the simultaneously. Thereby, two possible sources of the HOA factor have to be considered: (a) vehicle exhaust and (b) emissions from domestic heating. Considering

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the limited car traffic at Seiffen which represent less than 2000 vehicles per day (Saxony EPA agency data, Löschau, 2007) and the similarities of the HOA and BBOA time series, we can reasonably considered that residential heating may be an important contributor of the HOA factor.

The factor corresponding to LV-OOA was successfully compared with the mass spectra of OOA-1 and present a similar time series than sulphate ( $r^2 = 0.73$ ), as determined by Zhang et al., 2005). LV-OOA stands for the highly oxygenated organic part of the aerosol and it is commonly associated with aged organic aerosol and long-range transport particles (Jimenez et al., 2009). However, the LV-OOA time series correlated also with CO ( $r^2 = 0.62$ ) and its mass spectra is also similar to the mass spectra of fulvic acid (Fig. S2 in the Supplement), which is considered to be a model substance for Humic Like Substances (HULIS). 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) reported during laboratory investigations, a strong similarity between the organic mass spectra of wood burning emissions and the fulvic acid mass spectra especially during the smouldering phase. Consequently, taking into account the sampling site environment leads to the assumption that at least a part of the LV-OOA factor should have a direct wood combustion origin.

The SV-OOA factor was successfully compared with mass spectra of OOA-2 and its time series with nitrate ( $r^2 = 0.57$ ), as previously reported by Lanz et al. (2007) and Ulbrich et al. (2009). SV-OOA is considered to consist of less oxygenated compound and an indicator of less aged secondary organic aerosols than LV-OOA. The presence of SV-OOA might be linked to a possible aging of the wood combustion aerosol which will lead to the formation of less volatile organics aerosols by 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 measurements 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), aging of BBOA on

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deliquescence particles can be expected. Moreover, 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 PM<sub>1</sub> filter measurements made during the entire project (i.e. from October 2007 to March 2008) and additional laboratory measurements in the simulation chamber, the authors correlated the presence of methyl-nitrocatechols with the photooxidation of *m*-cresol directly emitted by wood combustion. In our study, high resolution analysis of the AMS mass spectra (performed using Pika v.1.06 software from D. Sueper, University of Colorado-Boulder, USA, DeCarlo et al., 2006) showed a similar profile between the C<sub>x</sub>H<sub>y</sub>O<sub>z</sub>N<sub>w</sub> 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 the C<sub>x</sub>H<sub>y</sub>O<sub>z</sub>N<sub>w</sub> fragments are related to methyl-nitrocatechols issue of the oxidation of *m*-cresol which might also represent a possible source of the observed SV-OOA factor.

The mean contributions of LV-OOA, SV-OOA, HOA and BBOA to total organics were approximately 36%, 26%, 17% and 20%, respectively. Contributions of BBOA and HOA are in the range of values observed by Lanz et al. (2010) in several places over Alpine mountains (17–49% and 6–16% of OA for BBOA and HOA respectively) but slightly lower than values previously observed by Favez et al. (2010) (BBOA = 37%).

The diurnal variations of the identified organic components (BBOA, HOA, LV-OOA and SV-OOA) during workdays and non-workdays are presented in Fig. 2. During workdays, tracers of fresh/primary organic aerosol (POA) emissions (i.e. BBOA and HOA) show two maxima: a first around 08:00 and a second around 19:00; and in accordance with the main observation made for the total OA, their concentrations were relatively stable during day time and decreased during the night. 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), indicating a stronger emission in the evening.

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A completely different diurnal variation was observed during non-working days; the BBOA concentrations increased slowly during the morning, reached a maximum around 16:00 and then slowly decreased to a minimum during the night with a small peak at 22:00. Although the HOA diurnal profile is similar to the diurnal profile of the BBOA, the increase of HOA during the day is not as constant as the increase of BBOA; small hourly changes were observed. However, during working days, BBOA and HOA concentrations mainly followed a profile similar to the total OA. Most of the time HOA and BBOA profiles correlate. This leads to the suggestion that the HOA emissions are more correlated with house heating emission than with the vehicle exhaust. This is in agreement with the low car traffic mentioned above, even if this conclusion seems to be less marked during weekend. Moreover, similarities between POA and OA diurnal patterns indicated that in Seiffen, POA is certainly the most important source of OA. Similar diurnal variations and comparisons between week and weekend were previously made by Krecl et al. (2008b), confirming that even if residential wood combustion emissions depend on the location and type of heaters; diurnal profiles and difference between week and weekend days can be more and less generalized. LV-OOA profile is close to the one of BBOA during both working and non-working days confirming a possible contribution of residential wood combustion to the LV-OOA. Finally, the diurnal variation of SV-OOA seems to be also very different during working and non-working days. During the week, the SV-OOA slowly increases during the day and reaches a maximum in the evening, while during the weekend, SV-OOA show a maximum at 14:00.

### 3.2.2 Polycyclic aromatic hydrocarbons (PAH)

Wood combustion is known to be a source of Polycyclic aromatic hydrocarbon (PAH) produced by the pyrolysis of cellulose and lignin. PAH were identified on the AMS organic mass spectra according to the fragmentation table described by Dzepina et al. (2007). The PAH time series is presented in Fig. 6, and a mean value of  $10 \pm 22 \text{ ng m}^{-3}$  was obtained for the whole campaign. Although the AMS is not able to

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provide PAH speciation as filters analysis made by CPP-GC-MS; a comparison of the AMS PAH concentration with the sum of the 18 PAH identified by filter samples show a high correlation (slope = 0.77,  $r^2 = 0.97$ ; Fig. 6 insert panel). Taking into account the uncertainties of 35–38% when identifying the PAH by AMS (Dzepina et al., 2007); we conclude that the measured AMS PAH concentration represents a good assumption of the total particulate PAH content. The mean PAH mass concentration measured by both methods ( $10 \text{ ng m}^{-3}$  for AMS PAH and  $12 \text{ ng m}^{-3}$  for filters, respectively) is in good agreement with residential wood combustion PAH concentrations previously reported for similar locations ( $10.2 \text{ ng m}^{-3}$  Glasius et al. (2008); 30 to  $22.9 \text{ ng m}^{-3}$  Bari et al. (2010a) and Bari et al. (2009)) and with the mean PAH concentration observed by Marchand et al. (2004) in urban and near-city background sites in French alpine valleys (range  $2.3\text{--}22.8 \text{ ng m}^{-3}$ ). However, it is two times higher than the mean concentration found by Favez et al. (2010) in the city of Grenoble (France). PAH contributes  $0.5 \pm 0.01\%$  to total organics aerosol which is slightly higher than values reported for Switzerland ( $0.1 \text{ ng m}^{-3}$ , Lanz et al., 2010).

In addition, the PAH time series (Fig. 6) shows several short-term events (lasting approximately 20–45 min) with concentrations up to  $500 \text{ ng m}^{-3}$ , which are extremely high compared to the mean concentration of  $10 \text{ ng m}^{-3}$ . Marchand et al. (2004) reported such high concentrations of PAH (up to  $150 \text{ ng m}^{-3}$ ) using 12 h sampling in two French alpine valleys during anticyclonic conditions with low temperature. However, the short-term of these high concentrations PAH events were observed during the whole campaign and their frequencies did not seem to be directly related to the meteorological conditions. Considering their durations, these high PAH events might be caused by strong local sources of PAH, e.g. a plume of a chimney exhaust of neighbouring houses. Nevertheless, due to their longer sampling time, filter samplers cannot show these peaks. This highlights the importance of a time resolution higher than 12 or 24 h in order to identify strong sources of PAH.

As done previously for the different NR-PM<sub>1</sub> and organic components, diurnal variation of PAH was calculated for working and non-working days (Fig. 2). During working

days, the diurnal variation of PAH shows two maxima at the same time as previously observed for organics, BBOA and HOA. However, during the weekend, the PAH profile shows a peak in the middle of the morning at the same time as HOA. It is therefore very likely that these two peaks are caused by the same source. The PAH variations seem to be directly connected to the BBOA and HOA factors. In the next section, the contribution of each factor (BBOA and HOA) to the total PAH will be investigated.

### 3.2.3 Contributions of the residential wood combustion to total PAH

As previously mentioned, the main interest of this field campaign was to quantify the impact of residential wood combustion on the local air quality in Seiffen. Using the high time resolution of the BBOA, HOA and PAH concentrations, the contribution of wood combustion to total PAH concentration was estimated. In order to do so, the ratio PAH/BBOA was calculated (Fig. 7 upper panel). Depending on the nature of the wood fuel as well as on the burning conditions, different values of the emission ratio PAH/levoglucosan or PAH/MA were reported (e.g. PAH/MA ranging from 0.0007 to 0.014 (Iinuma et al., 2007) and from 0.06 to 0.113 (Dhammapala et al., 2007)). According to this, a setting value of PAH/BBOA = 0.03 was considered to separate PAH directly related to wood combustion emissions (ratio < 0.03; referred to PAH<sub>wb</sub> here) from PAH not correlated to BBOA but coming from other PAH sources like car exhaust and/or domestic liquid fuel heaters (ratio > 0.03; referred to PAH<sub>no-wb</sub> here). Then, the PAH<sub>no-wb</sub> could be connected to the HOA factor. As presented in the bottom panel of Fig. 7, PAH<sub>wb</sub> (in purple) is related linearly to BBOA (slope = 0.015,  $r^2 = 0.87$ ), indicating that PAH<sub>wb</sub> emissions represent about 1.5% of the emitted mass of BBOA (e.g. total MA compounds). This is in agreement with the range of 0.7 to 2.2% defined by Weimer et al. (2008) during laboratory measurements of emissions from different wood species during flaming and smoldering modes.

According to this, the PAH<sub>wb</sub> concentration was estimated from the BBOA time series and compared to the total PAH concentration. Consequently, during the AMS measurements period in Seiffen, wood combustion was assumed to contribute about

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62% to the total particulate PAH mass concentration. This results confirms the impact of residential wood combustion on the local PAH concentration reported by Boström et al. (2002), Mandalakis et al. (2005) and Sheesley et al. (2009) in Sweden ( $PAH_{wb} = 50\text{--}80\%$ ). However, Mandalakis et al. (2005) who compared the contribution of  $PAH_{wb}$  in three different European background sites (Sweden, Croatia and Greece) reported large variations of the  $PAH_{wb}$  fractions (e.g. only 10% in southern European atmosphere) and justified this finding by differences in the contribution of residential wood combustion to total house heating systems.

Moreover, residential wood combustion seems to be the main source of individual PAH. Sheesley et al. (2009) estimated that in Lycksele (Sweden) 71–87% of individual PAH may have a wood combustion origin and Nielsen and Illerup (2006) argued that in some locations in Denmark 80 to 90% of some individual PAH compounds like B[a]P were caused by wood combustion. These results are in accordance with our estimation that  $PAH_{wb}$  contribution is a main source of PAH at Seiffen (ranging from 2 to 100% of total PAH). Moreover, Bari et al. (2010a) reported a contribution of 44% of the potential carcinogenic PAHs to total particulate PAH pointed out potential impact on human health. It is important to link these results to the actual European legislation and more specially to the European Directory 2004/107/CE, imposing to all European Member States from the 31 December 2012 a value of  $1\text{ ng m}^{-3}$  for the total content of B[a]P in the  $PM_{10}$  fraction averaged over a calendar year. The B[a]P was chosen because of its toxicity and because it is considered to be a good indicator of the total PAH content in the air. As such, according to our results and previous discussion, we conclude that an annual value might be representative for total PAH concentration, but not useful enough to estimate the real human exposition to PAH and seasonal variations.

Finally, based on the high time resolution of the AMS, our results outline the need to carefully monitor PAHs in the particles as well as in the gas phase to provide a better understanding of human exposure. For this kind of approach, parallel AMS and filters samples are promising instruments to provide high time resolution total PAH concentration and a clear fingerprint of the PAH composition.

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## 4 Conclusion

In order to study the impact of residential wood combustion on local PM in the small town of Seiffen (Saxony, Germany), an AMS was deployed during January 2008 in complement to a longer period of daily PM<sub>1</sub> filter measurements. While filter samples provided a daily detailed chemical composition of PAH and anhydrous monosaccharide compounds, the AMS provided a highly time resolved (5 min) information about the daily variation of the NR-PM<sub>1</sub>. Using FA-AMS analysis, the AMS organic signal was decomposed in 4 factors – LV-OOA, SV-OOA, BBOA and HOA - corresponding to 36%, 26%, 20% and 17% of the total OA respectively. In addition, PAH concentration extracted from the organic mass spectra (Dzepina et al., 2007) represented 0.5% of total OA. The presence of short-term events of extremely high PAH concentrations were observed and directly attributed to an emission plume chimney exhaust from a house located close to the point of sampling. BBOA factor and AMS PAH were successfully compared to daily PM<sub>1</sub> filter samples, leading to the conclusion that they are good estimates of the total particle content in MA and PAH, respectively. Moreover, according to its similarity with fulvic acid mass spectrum (a model substance for HULIS) and its correlation with the CO time series, a possible contribution of wood combustion to LV-OOA was suggested. Oxidation of wood combustion VOCs leading to SOA formation was previously observed at Seiffen and linked to the presence of methyl-nitrocatechols (Iinuma et al. 2010) which highly correlates with the detected C<sub>x</sub>H<sub>y</sub>O<sub>z</sub>N<sub>w</sub> fragments. Oxidation of emitted wood combustion compounds might be a good explanation for the presence of SV-OOA. HOA was also found to be mainly linked to residential heating considering the low car traffic of the village and its better correlation with CO than NO<sub>x</sub>. Although wood combustion was performed in only one-third of the houses in Seiffen, it represents the main primary source of particles. We thus conclude that POA emissions at Seiffen mainly correspond to general residential heating during the measurement period. Because of the high time resolution of the AMS measurements, it was possible to observe diurnal profiles of the identified aerosol components. A clear difference was

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observed between workdays (with maxima early in the morning and in the evening) and weekend days (maximum during the afternoon). PAH emitted by residential wood combustion was estimated around 1.5% of the total mass of BBOA, representing approximately 62% of the total PAH measured. The large contribution of the residential wood combustion to the PAH concentration highlights its possible impact on human health, as considered in previous toxicological studies. It also emphasizes the need to decrease the particulate emissions of residential wood combustion in the aim of decreasing atmospheric PAH concentrations in agreement with the future European regulation concerning PAH (European Directory 2004/107/CE).

Our measurements draw attention to advantages of a combination of AMS measurements and filter samplers. While the AMS provides information about changes in the concentration during the day, filter samplers provide the chemical finger print of the particles during the whole day. The filter samples can for example, segregate between softwood and hardwood combustion for example (Bari et al., 2009, 2010b; Schauer and Cass, 2000). Combining approaches using off-line and on-line measurements in the next field campaigns will be a real improvement in helping to understand the changes of the aerosol chemical composition as well as the human exposure to toxic chemical compounds as polycyclic aromatic hydrocarbons.

Finally, comparing three different European background sites, Mandalakis et al. (2005) reported a large variation of the PAH<sub>wb</sub> contribution between the countries. Therefore, the impact of residential wood combustion on the air quality not only depends on the types of wood burned or heaters used but also on the local policy, which can promote this source of energy or not. In the current context of a large development of residential wood combustion and governmental wishes to improve local air quality, our results confirm the requirement of highly controlled domestic heating emissions including wood combustion in addition to the actual regulations mainly focusing on the reduction of industrial and traffic emissions.

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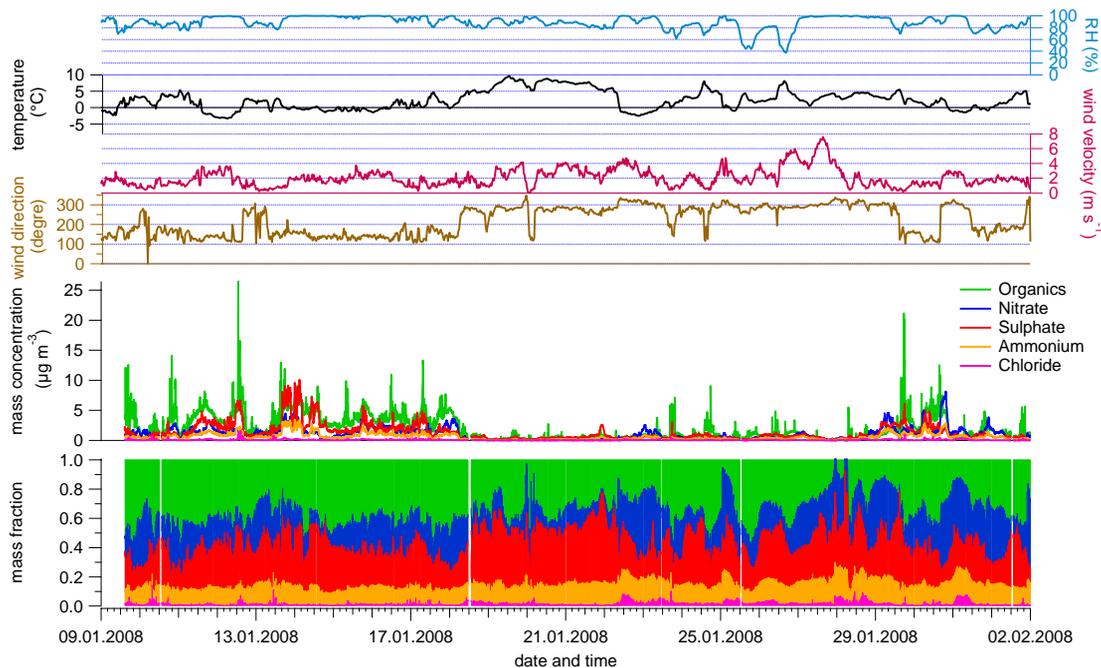

**Table 1.** Summary of the mean concentrations measured on PM<sub>1</sub> filter samples and AMS.

	PM <sub>1</sub>		AMS	
	Mass (μg m <sup>-3</sup> )	Mass (μg m <sup>-3</sup> )	Mass fraction (%)	
Total	15.18±6.11	5.05±4.42		
Organics	1.18±0.55 <sup>1</sup>	2.02±2.00 <sup>1</sup>	40	
Nitrate	1.27±0.94	1.05±1.02	21	
Sulphate	1.59±0.74	1.29±1.33	26	
Ammonium	1.08±0.53	0.61±0.54	12	
Chloride	0.03± 0.02	0.07±0.08	1	

<sup>1</sup> The PM<sub>1</sub> organics corresponds to OC and the AMS organics corresponds to OM.

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**Fig. 1.** Campaign overview including local meteorological parameters (relative humidity (RH), temperature, wind direction and wind velocity), and time series mass concentration and mass fraction of the main NR-PM<sub>1</sub> aerosol components during the AMS campaign.

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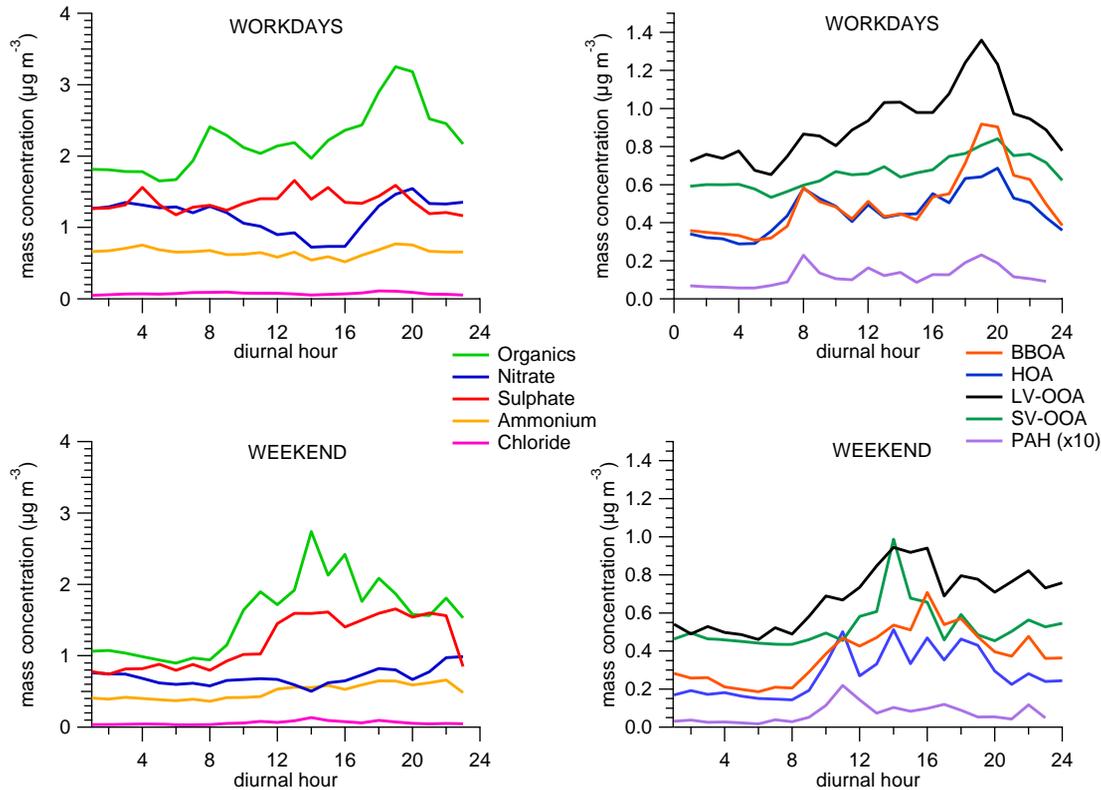
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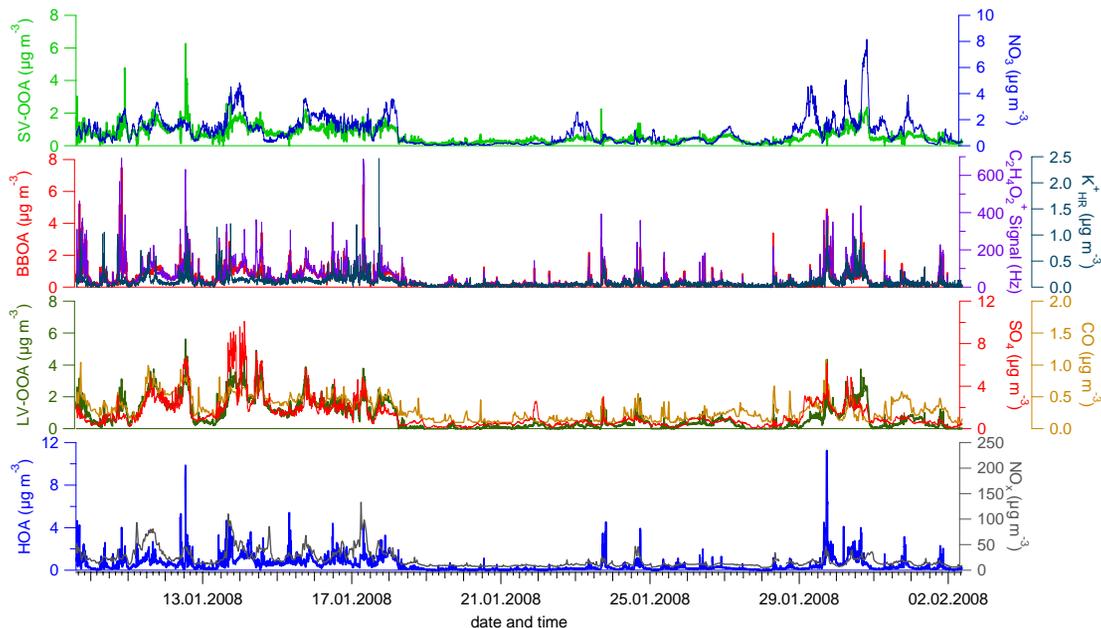




**Fig. 2.** Diurnal variations of the aerosol composition during workdays (top) and weekend days (bottom).

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**Fig. 3.** Time series of the 4-factors and their selected tracer species.

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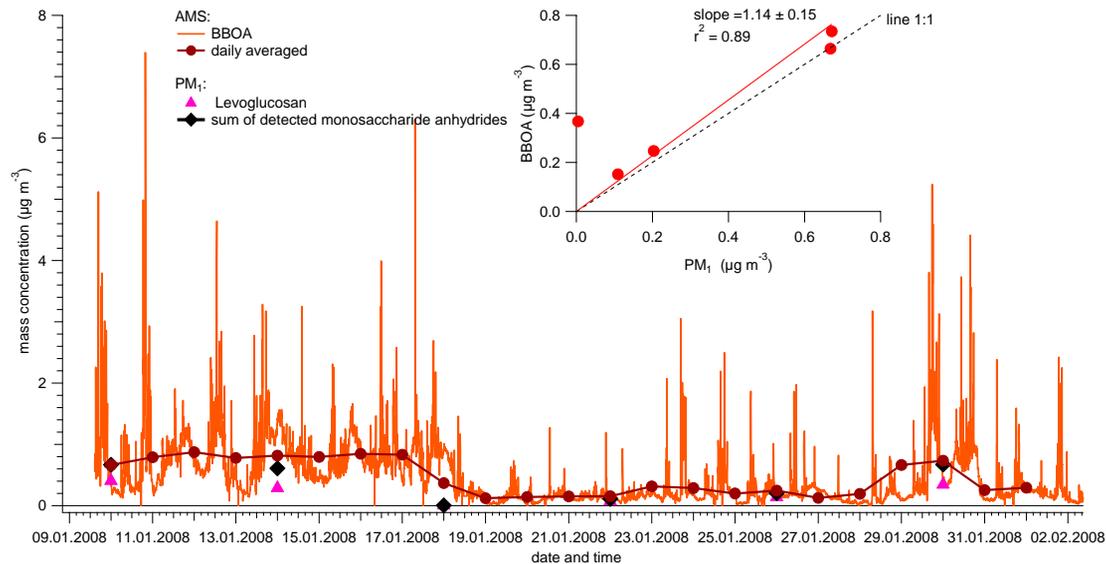
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**Fig. 4.** Time series of BBOA factor and comparison with the sum of PM<sub>1</sub> MA identified (levoglucosan, arabitol, mannosan, mannitol, mannose, galactosan, glucose and sucrose).

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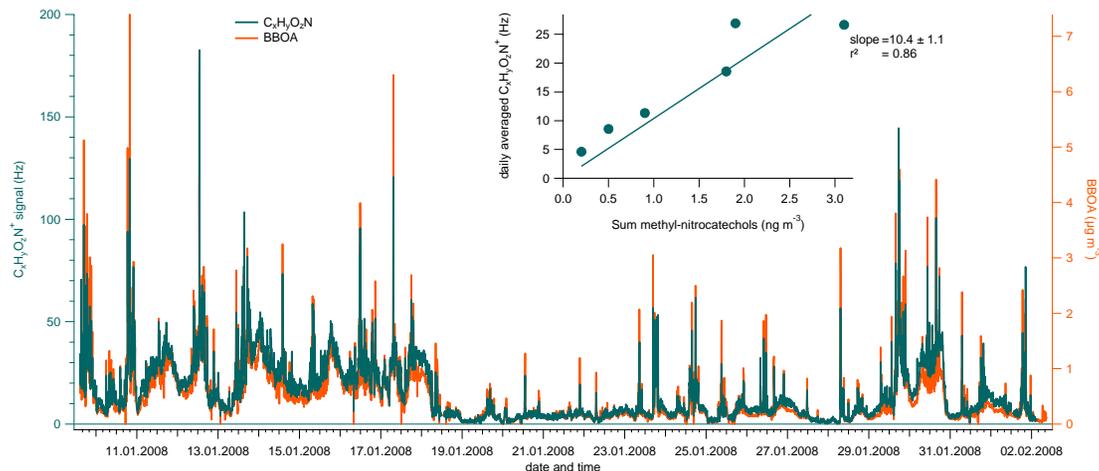
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**Fig. 5.** Comparison of the  $C_xH_yO_zN_w$  fragments with BBOA factor and daily  $\text{PM}_{10}$  methyl-nitrocatechols.

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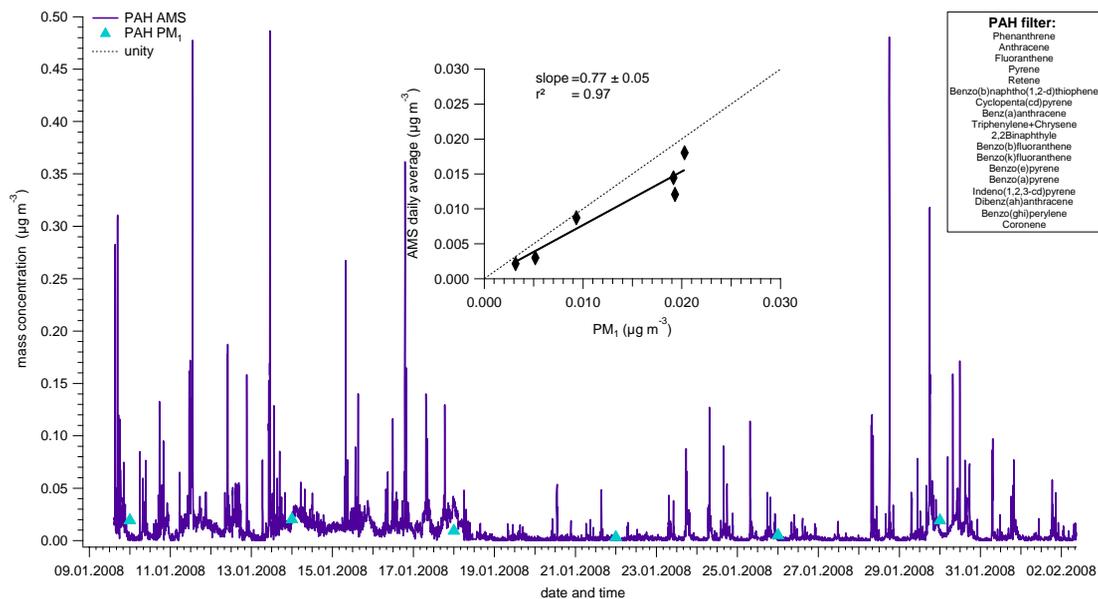
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**Fig. 6.** AMS-PAH time series and comparison with daily  $PM_{10}$  filters.

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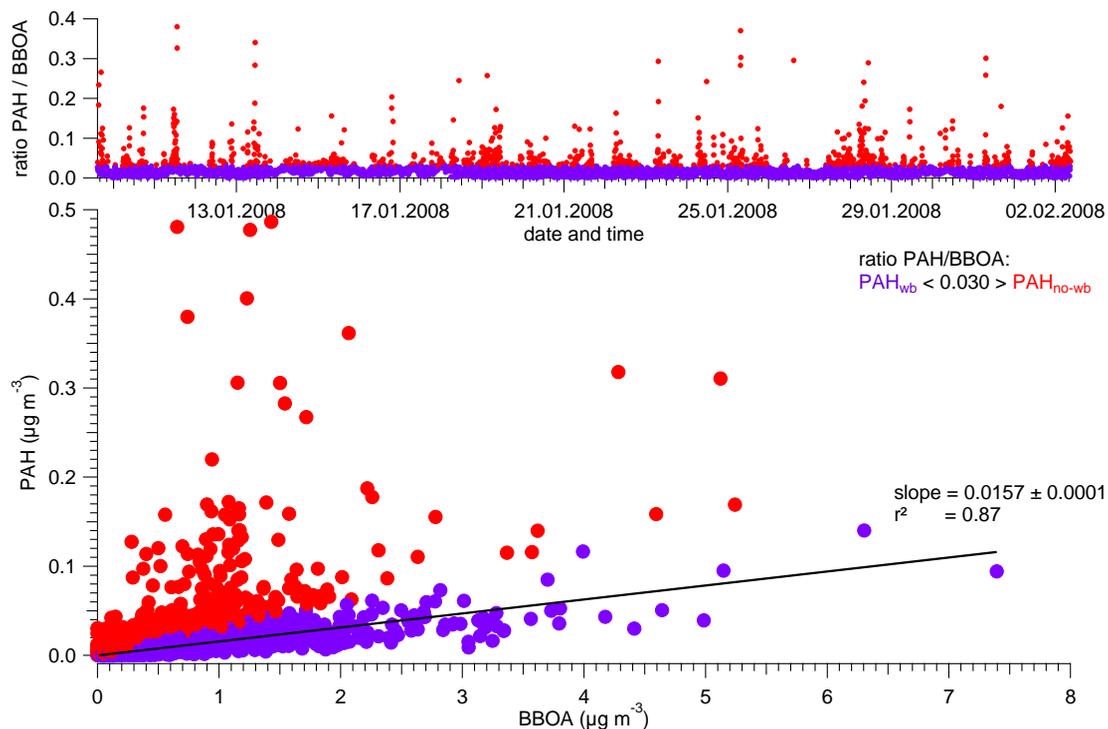
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**Fig. 7.** Estimation of the biomass burning contribution to total PAH concentration. The upper panel represents the time series of the ratio PAH/BBOA and the bottom panel shows the correlation between PAH and BBOA. Colours correspond to the differentiation between PAH correlated to wood combustion (purple) and not correlated to wood combustion (red).

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