Air Pollution in Residential Areas from Wood-fired Heating

Md. Aynul Bari[1*], Günter Baumbach[1], Bertram Kuch[2], Günter Scheffknecht[1]

[1] *Department of Air Quality Control, Institute of Combustion and Power Plant Technology, Universität Stuttgart, Pfaffenwaldring 23, 70569 Stuttgart, Germany*
[2] *Institute of Sanitary Engineering, Water Quality and Solid Waste Management, Universität Stuttgart, Bandtäle 2, 70569 Stuttgart, Germany*

ABSTRACT

An important source of inhalable particles in residential areas, particularly in the winter season, is the biomass combustion when wood is used for domestic heating. This is a continuation of our previous investigation about wood smoke pollution in residential areas of southern Germany (Bari *et al.*, 2009). The target of this study was to characterise ambient levels of criteria pollutants, their risk assessment and find out influence of hardwood combustion on local air quality. Particle-phase PM_{10} samples were collected at a residential site Dettenhausen near Stuttgart during 2005/06 and winter 2009. Samples were analysed by gas chromatography–mass spectrometry for polycyclic aromatic hydrocarbons (PAHs) and other wood smoke tracer compounds (e.g., levoglucosan, methoxyphenols). High concentrations of PM_{10} and total PAHs were found during winter 2009 like winter 2005/06. Carcinogenic PAHs were detected in high concentrations and contributed 44% of the total PAHs in the ambient air. The significant concentrations of hardwood markers (i.e., syringaldehyde, acetosyringone) found in the ambient air suggest that the influence of hardwood combustion on ambient air quality is significant. Based on the emission ratio of hardwood markers and PM_{10}, it can be concluded that in the investigated residential site about 57% of ambient PM_{10} pollution can be attributed to hardwood combustion for winter heating.

Keywords: Wood combustion; Domestic heating; Inversion; PM_{10}; PAHs; Syringaldehyde; Acetosyringone.

INTRODUCTION

Wood as a renewable and worldwide available fuel is used for residential heating in small-scale fires during winter. In contrast to standard fossil fuels which generate greenhouse gas emissions contributing to global climate change, wood combustion is considered as a sustainable energy source. But wood smoke emissions can cause short and long-term health effects including acute respiratory infections, tuberculosis, asthma, chronic obstructive pulmonary disease, blindness, headache and reduced birth weight (Mishra *et al.*, 1999; Smith *et al.*, 2000; Mishra *et al.*, 2004; Schei *et al.*, 2004; Ekici *et al.*, 2005) Furthermore, significant concentrations of carcinogenic organic compounds emitted from wood combustion might contribute to the increasing risk of cancer. The use of wood for domestic heating is very familiar to the general public, especially in residential areas. Rising prices and uncertain availability of

fossil fuels for heating together with environmental concerns has resulted in the increased use of wood in many developing as well as in developed countries such as Germany.

In the residential villages of Germany, several households use log wood boilers, with and without a heat storage tank, for central heating and for additional heating including mainly manually fed chimney stoves, tiled stoves, and open fire places. Depending on the users' behaviour, such wood-firings can cause high particulate emissions (Johannson *et al.*, 2004; Tissari *et al.*, 2008). Wood combustion causes regional haze with high PM_{10} concentrations in the ambient air, resulting in considerable annoyance and nuisance with complains among the inhabitants. Even the inhabitants of such residential villages can often smell a distinct 'wood smoke odor' in the air (Bari *et al.*, 2009). Thus, it is evident that residential wood combustion has a direct influence on ambient air quality. So, due to public concern about the need of emission reduction and to justify the evidence, we investigated the air pollution caused by wood-fired heating in such residential areas.

This manuscript is a continuation of our previous study investigating on wood smoke pollution as a source of

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* Corresponding author. Tel.: 49-711-685-63489;
  Fax: 49-711-685-63491
  E-mail address: bari@ifk.uni-stuttgart.de

organic compounds in residential areas of southern Germany (Bari *et al.,* 2009). In that paper, we determined particle-phase organic compounds emitted from residential wood combustion, defined source fingerprints of polycyclic aromatic hydrocarbons (PAHs) for hardwood, softwood and other emission sources, characterised ambient levels of PAHs and methoxyphenols during winter months (November 2005 to March 2006) and finally applied receptor model positive matrix factorization to find out contribution of different emission sources including wood combustion to the ambient PM_{10}-bound organic compounds. In another continued study (Bari *et al.,* 2010a), we made a detailed characterisation of the identified wood smoke tracer compounds, their ambient levels and temporal variation, size distribution of ambient particles and determined the contribution of wood combustion to ambient PM_{10} pollution using emission ratio approach of general wood smoke tracer compound i.e., levoglucosan and PM_{10}. In this study, we reported criteria pollutants (e.g., NO_x, PM_{10}) in late winter and spring 2005, characterised ambient levels of PAHs during winter 2009 and compared results with winter 2005/06. Furthermore, we checked carcinogenic risk assessment of PAHs and finally used emission ratio approach to quantify the contribution of hardwood combustion to ambient PM_{10} pollution.

MATERIALS AND METHODS

Ambient Measurements and Sampling Procedure

Ambient PM_{10} samples were collected at a residential site in Dettenhausen (area 11 km^2, altitude 500 m above

sea level and 5389 inhabitants), which is located at the northern edge of the nature park area 'Schoenbuch', a large forest in the state of Baden-Wuerttemberg, Germany. The word "Schoenbuch" means "nice beeches" i.e., mainly beeches are growing in this forest and hardwood from these trees is the main solid biofuel used for domestic heating. Since people in this village can get hardwood easily from the forest, more than 90% of firewood used in this site is hardwood, mainly beech wood. This has been confirmed by the official local chimney sweeper (Decker Roman, personal communication, 2009). The other emission sources in this site are central heating with light oil and gas firing, but their emissions could be determined as very low due to good combustion quality (Bari *et al.*, 2009). Further sources are low traffic and regional background load, which are not dominant in this village. During winter under surface inversion conditions, the particulate matter (PM) load over the village could be visually detected coming from residential wood firings.

PM_{10} sampling was performed in two campaigns during 2005/06. The first campaign was conducted from 24 February to 30 June 2005 in one sampling site located in the village-centre (Rathaus) considered as background site. Another site was chosen in a residential area where the sampling period was from 2 March to 7 June 2005. The two selected sampling sites are depicted in Fig. 1. To identify the pollution load coming from the local emissions, a multi compound monitoring unit i.e., one measurement vehicle equipped with NO_x and TEOM instrument was installed in the village-centre. A second campaign was performed in the same residential site from 1 November

Fig. 1. Selected sampling sites at the residential village Dettenhausen.

2005 to 31 March 2006. During winter 2005/06, the average daily temperature was between –14 and 14°C and the area was covered with snow on most of the days in January and February 2006, which increased the demand for residential heating. Strong surface inversions were evident during 8–16 January and 25 January–6 February in southern Germany (Regierungspraesidium Stuttgart, 2006), which limited the dispersion of atmospheric pollutants. To check the influence of residential wood burning, PM_{10} samples were also collected during winter 2009 from 8 January to 23 February.

The gravimetric PM_{10} sampling was carried out using low-volume samplers (LVS3, Leckel, Germany) in accordance with the reference method defined in the European Standard EN 12341:1998 (CEN, 1998). The air inlet was located 2.5 m above ground level. The pumped volume flow rate was 2.3 m³/h. The membrane filters of cellulose nitrate (AE 98, Schleicher & Schuell, Germany) with 5 µm pore size and 47 mm diameter were used for collection of ambient particles during the first campaign (February to June 2005). In the winter 2005/06 campaign cellulose nitrate filters and as well as glass fibre filters (GF; diameter 50 mm; Whatman, UK) were chosen for parallel sampling in the residential area. To obtain sufficient material on the filter, the sampling duration was selected for 48 h during weekdays and 72 h over the weekends (including Friday). Prior to sampling, the glass fibre filters were baked at 300–550°C for at least 6 h to remove any traces of organics. After sampling, the filters were packed in petri dishes covered with aluminum foil to protect them from sunlight. In accordance with the protocol (CEN, 1998), the filters were conditioned in a weighing chamber for 48 h under controlled conditions before and after sampling. The weight of each filter was determined using an analytical balance (MC 210 P; Sartorius, Goettingen, Germany) with an accuracy of ± 0.02 mg and the filters were stored in a refrigerator (−18°C) until analysis.

Organic Chemical Analysis

The collected filter samples were analysed by gas chromatography mass spectrometry (GC-MS). The detailed extraction procedure, GC configuration and temperature profile are described in Bari *et al.* (2009). Standards of PAHs were purchased from LGC Promochem (Wesel, Germany), and Sigma-aldrich (Steinheim, Germany). The isotope labelled PAH standard containing naphthalene-d_8, acenaphthene-d_8, acenaphthylene-d_{10}, fluorene-d_{10}, phenanthrene-d_{10}, anthracene-d_{10}, fluoranthene-d_{10}, pyrene-d_{10}, chrysene-d_{12}, benzo[a]anthracene-d_{12}, benzo[k]fluoranthene-d_{12}, benzo[b]fluoranthene-d_{12}, benzo[a]pyrene-d_{12}, dibenzo[a,h]anthracene-d_{14}, indeno[1,2,3-cd]pyrene-d_{12}, and benzo[ghi]perylene-d_{12} was used for quantification. Retene was quantified based on seven-point standard calibration curves using authentic standard. Each species was identified and quantified based on its retention time and integrated area of its molecular ion fragments compared to that of the appropriate standard. Compounds for which standards were not available were identified comparing their mass spectra and interpretation of mass spectrometric fragmentation patterns to the standards for the similar compounds and by comparison to the National Institute of Standards and Technology (NIST) libraries. The quantification of these compounds was based on the response factors for the authentic standards of closely related compounds. For blank filter samples, measurement results, especially for naphthalene and phenanthrene, were generally below the quantification limit; therefore, no blank correction was applied to the measurement results.

RESULTS AND DISCUSSION

The result of continuous measurements of NO_x at the village-centre (Rathaus) is shown in Fig. 2. The values are
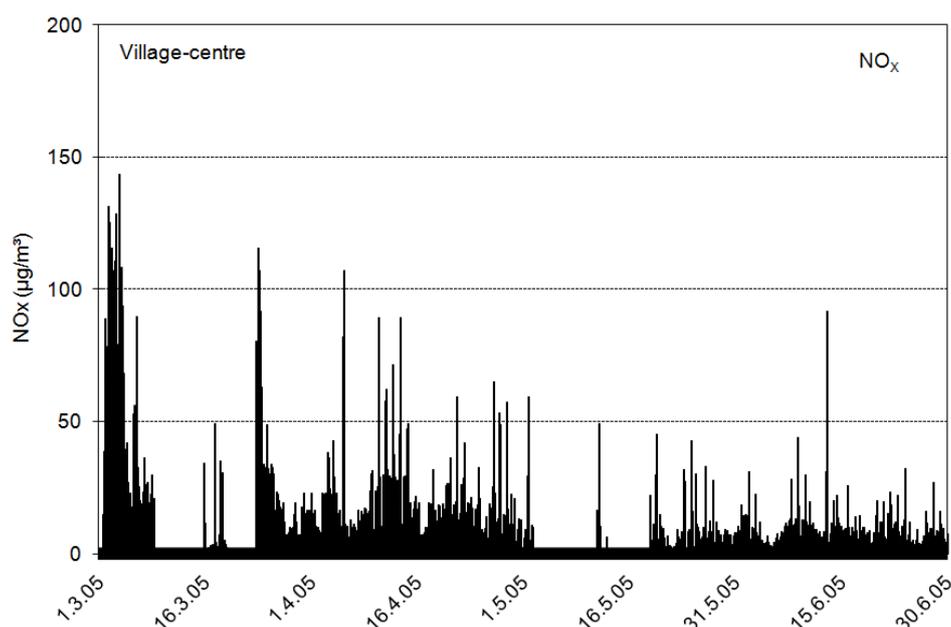
Fig. 2. Temporal concentration profiles of NO_x at the village-centre during March to June 2005.

presented as hourly averages for March to June 2005 and show a low pollution load emitted from the combustion sources during this campaign. The gravimetric PM_{10} concentrations at the background site village-centre and the residential area are shown in the Fig. 3. It was found that there was no significant difference of PM_{10} levels in both sites. High concentrations were observed during February and March 2005, which was assumed to be caused by residential wood-fired hearting. While at the end of the heating season and during the spring months i.e., from April to June 2005, comparatively low PM_{10} concentrations were evident. It was found that PM_{10}

concentrations obtained from TEOM measurements were lower than the gravimetric method and a correlation coefficient of 0.61 was found between the TEOM and gravimetric measurements, with an intercept of 5 $\mu g/m^3$, which might be the result due to the loss of some semi-volatile compounds in the inlet of TEOM.

The PM_{10} levels at the residential site during winter 2005/06 from two parallel measurements using two filter media (i.e., glass fibre and cellulose nitrate) are shown in Fig. 4. The PM_{10} measurements using cellulose nitrate filters were carried out up to 3 February 2006. It was found that there was no significant difference of PM_{10} concentrations
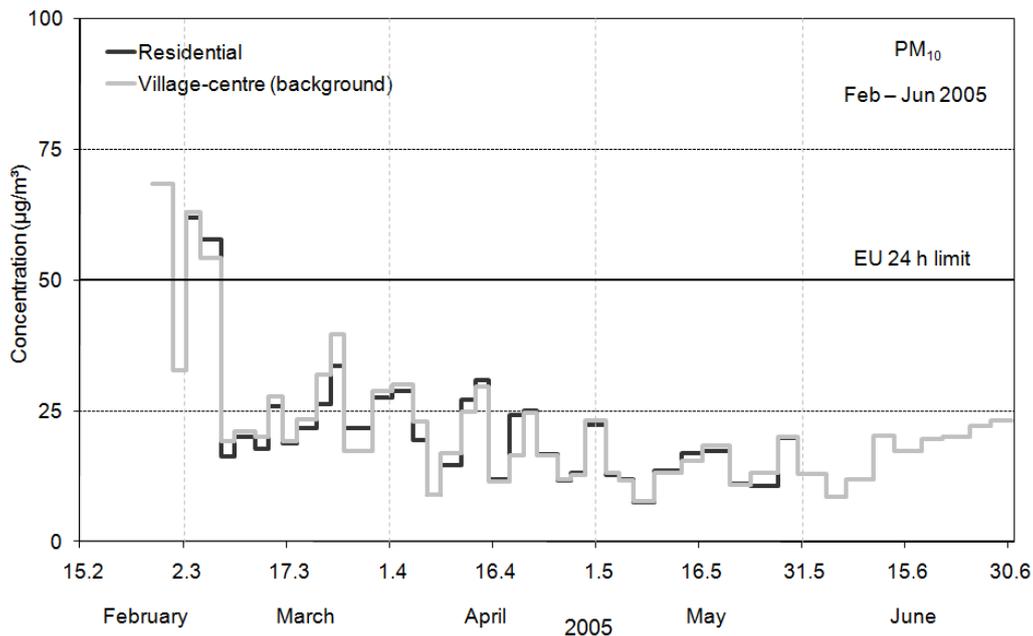
Fig. 3. Temporal ambient concentration profiles of PM_{10} at the background and the residential site (February to June 2005).
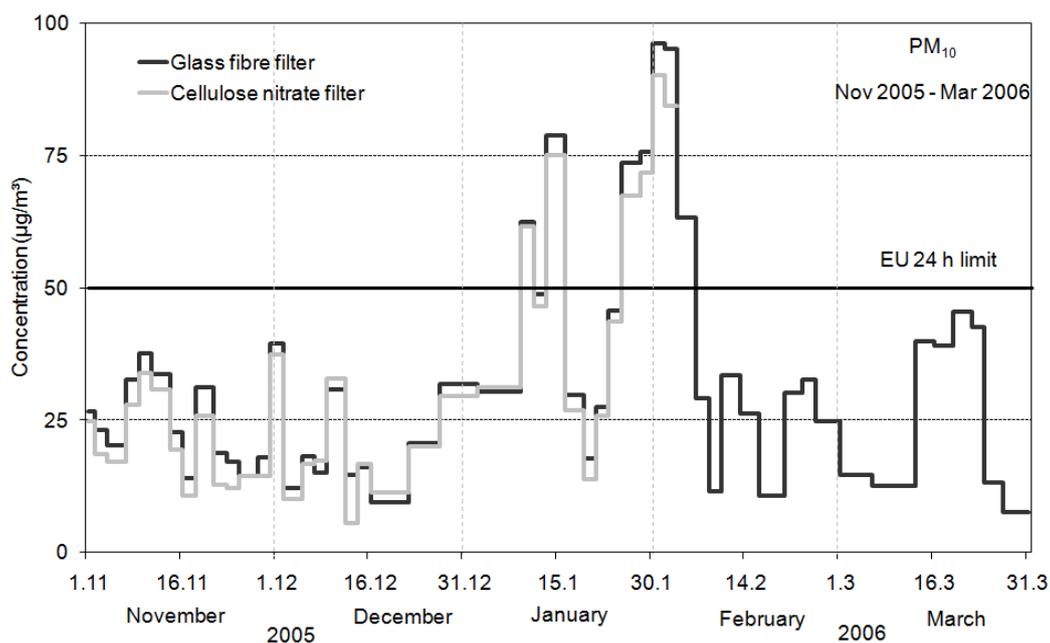
Fig. 4. Temporal concentrations of PM_{10} at the residential site during winter 2005/06.

using the two different filter media and a good correlation ($R^2 = 0.99$) of PM_{10} concentrations was evident. The purpose of using glass fibre filters was to use PM_{10} filter samples for extraction and analysis with gas chromatography and mass spectrometry (GC-MS) in order to find out organic compounds such as polycyclic aromatic hydrocarbons (PAHs) and other compounds e.g., levoglucosan (Shafizadeh, 1968; Locker, 1988; Simoneit *et al.*, 1999; Fraser and Lakshmanan, 2000; Jordan *et al.*, 2006) and methoxyphenols (Simoneit *et al.*, 1993; Fine *et al.*, 2002, 2004), which are the organic tracers for the identification of residential wood combustion for domestic heating during the winter time. It was found that at the beginning of winter months (November, December), the average PM_{10} concentrations varied from 10 to 40 μg/m³. The highest peak concentrations were observed from middle of January to the early February due to the strong barrier layer of the surface inversions which limited the dispersion of air pollutants.

The mean, median, minimum and maximum values for each PAH compound and total PAHs during the winter 2009 at the residential site are shown in Table 1. Altogether 19 PAHs were quantified in 16 ambient particle-phase PM_{10} samples in Dettenhausen during winter 2009. The results are compared with the measurements of PAHs during winter 2005/06. It was found that during the winter 2009, the average value of total PAHs was 30.1 ng/m³ (median 16.5 ng/m³), with a maximum value of 94.2 ng/m³ and a minimum of 2.95 ng/m³. The most dominant compounds found in the PM_{10} samples from ambient air were fluoranthene (Flut) (16.8%), pyrene (Py) (16.4%), benzo[a]anthracene (BaA) (9.5%), benzo[a]pyrene (BaP)

(7.2%), chrysene (Chr) (7.0%), phenanthrene (Phe) (6.8%), benzo[b]fluoranthene (BbF) (6.4%), benzo[k]fluoranthene (BkF) (5.8%), benzo[ghi]perylene (BghiP) (5.2%) and the other 9 compounds represent about 13% of the total PAHs concentration. Due to the two months measurement in winter 2009, the average concentrations of Phe, Flut, Py and BaP were found higher than the values in winter 2005/06. To check the influence of carcinogenic PAHs in this residential site during winter 2009, the relative contribution of each carcinogenic PAH to the total PAHs was calculated. It was found that carcinogenic PAHs contributed 44% of the ambient total PAHs, which was similar to the contribution found in winter 2005/06. It was evident that during winter 2009 most of the particle-phase carcinogenic PAHs especially BaA, BaP, Chr, BbF and BkF were found in significant concentrations in the residential site. The winter average of most carcinogenic compound BaP was 2.2 ng/m³, which was higher than our previous measurement during winter 2005/06 (1.6 ng/m³). While compared to other studies influenced by residential wood combustion, average BaP concentration of this study was higher than in winter 2006 in Kurkimäki, Finland (1.3 ng/m³) (Hellen *et al.*, 2008) and in winter 2006/07 in a residential village Slagslunde (1.7 ng/m³) in the north of Copenhagen, Denmark (Wåhlin *et al.*, 2010) but lower than in winter 2009 in a residential site (13.3 ng/m³) of Tianjin, China (Shi *et al.*, 2010), dominated by coal combustion for heating and wood combustion for cooking and heating.

In this study, we calculated carcinogenic risk of nine carcinogenic PAHs during winter 2009, and compared with winter 2005/06 values using toxic equivalent factors (TEF) and cancer unit risk from the following relationship:

Table 1. Mean, median, minimum and maximum concentrations (in ng/m³) of each PAH compound at the residential site Dettenhausen.

PAHs ng/m³	Ring	Winter 2005/06 (n = 52)[a]			Winter 2009 (n = 16)		
		Mean	Median	Range	Mean	Median	Range
Naphthalene (Nap)	2	0.08	0.07	0.02–0.4	0.11	0.14	0.01–0.2
Acenaphthene (Ace)	3	0.05	0.03	0.004–0.3	0.06	0.05	0.02–0.2
Fluorene (Fl)	3	0.1	0.08	0.01–0.4	0.18	0.16	0.07–0.3
Phenanthrene (Phe)	3	0.95	0.5	0.13–6.7	2.03	1.11	0.07–7.2
Anthracene (An)	3	0.2	0.07	0.004–1.3	0.33	0.13	0.05–1.0
Retene (Ret)	3	0.7	0.4	0.02–3.4	0.58	0.37	0.03–1.9
Fluoranthene (Flut)	4	2.9	1.4	0.22–19.07	5.05	3.45	0.13–15.9
Pyrene (Py)	4	2.3	1.5	0.18–23.8	4.92	2.76	0.11–18.0
Chrysene (Chr)	4	1.8	1.1	0.07–8.7	2.1	1.24	0.09–6.2
Benzo[a]anthracene (BaA)	4	2.3	1.4	0.19–10.5	2.87	1.77	0.35–8.2
Benzo[k]fluoranthene (BkF)	5	1.9	1.2	0.15–7.5	1.75	1.14	0.31–5.3
Benzo[b]fluoranthene (BbF)	5	1.9	1.3	0.14–8.3	1.92	1.32	0.30–6.1
Benzo[j]fluoranthene (BjF)	5	0.5	0.3	0.02–2.6	0.76	0.45	0.06–2.5
Benzo[e]pyrene (BeP)	5	1.5	0.8	0.12–6.4	1.8	1.25	0.33–5.0
Benzo[a]pyrene (BaP)	5	1.6	0.9	0.05–7.4	2.17	1.27	0.12–7.1
Perylene (Prl)	5	0.3	0.2	0.01–1.4	0.38	0.23	0.03–1.2
Dibenzo[a,h]anthracene (DBA)	5	0.2	0.2	0.02–0.9	0.26	0.16	0.05–0.7
Indeno[1,2,3-cd]pyrene (IP)	6	1.1	0.7	0.09–4.8	1.34	0.91	0.22–3.9
Benzo[ghi]perylene (BghiP)	6	1.6	0.9	0.12–7.4	1.56	1.03	0.24–4.6
Total PAHs		23	13.3	1.6–121.4	30.1	16.5	2.95–94.2

[a] Supplemented from Bari *et al.* (2010).

Carcinogenic risk = $[PAH]_i \times TEF_{PAHi} \times UR_{BaP} = BaP_{eq} \times UR_{BaP}$ (1)

UR_{BaP} is the inhalation cancer unit risk factor of BaP (= 1.1×10^{-6} $(ng/m^3)^{-1}$) estimated from cancer potency factor (CPF) using the following relationship:

$UR = CPF \times 20 \ m^3/(70 \ kg \times CV)$ (2)

where cancer unit risk (UR) represents the excess cancer risk associated with an inhalation exposure to a concentration of 1 $\mu g/m^3$ of a compound, the cancer potency factor (CPF) (= 3.9 $(mg/(kg\text{-}day)^{-1})^{-1}$ for BaP) indicates the excess cancer risk associated with exposure to 1 mg of a compound per kilogram of body weight, 70 kg is the reference human body weight, 20 m^3 is the reference human inspiration rate per day, and CV is the conversion factor from mg to ng (= 1×10^6) (OEHHA, 2005).

The total carcinogenic potency as estimated by total BaP_{eq} and total carcinogenic risk calculated from ambient particle-phase concentrations of nine PAHs are presented in Table 2. It can be seen that total BaP_{eq} concentration in the village Dettenhausen during two months measurements in winter 2009 was 3.4 ng/m^3, which was comparatively similar to the previous results found from winter 2005/06 (2.7 ng/m^3). From the risk estimates, it was found that total carcinogenic risk during winter 2009 in Dettenhausen was found comparatively similar like winter 2005/06. From this risk assessment, we can underline that wood smoke particles emitted from manually fed batch combustion and uncontrolled combustion of modern pellet oven and log wood boiler for residential heating might contribute potential health risk to the people living in the rural areas. Therefore, these results suggest an important concern of controlling emissions from wood-fired heating during winter in the residential areas.

To quantify the contribution of wood combustion to ambient PM_{10} pollution, levoglucosan has been suggested as tracer due to its high atmospheric stability. Several researchers have adopted different approaches to find out the contribution of particulate matter originating from wood combustion. Schmidl *et al.* (2008) considered relationships of levoglucosan with organic carbon (OC) and PM from wood smoke in mass per volume units based on the factors derived from the emissions. Another common approach to assess the contribution of wood combustion to ambient PM concentrations is to use the emission ratio of levoglucosan to PM (mass levoglucosan to mass PM) (Yttri *et al.*, 2005; Wang *et al.*, 2007). In our study, we adopted a semi-quantitative approach using average mass fraction of lignin breakdown products i.e., hardwood marker compounds from emissions and ambient air samples. In our previous investigation (Bari *et al.*, 2009), syringaldehyde and acetosyringone were found as most dominant compound in

Table 2. Total carcinogenic risk and BaP equivalent values (ng/m^3) in the residential areas.

	Carcinogenic group	TEF[a]	Winter 2005/06 Dettenhausen		Winter 2009 Dettenhausen	
			Mean ng/m^3	BaP_{eq} ng/m^3	Mean ng/m^3	BaP_{eq} ng/m^3
Naphthalene (Nap)	2B	0.001	0.08	0.00008	0.11	0.00011
Acenaphthene (Ace)	3	0.001	0.05	0.00005	0.06	0.00006
Fluorene (Fl)	3	0.001	0.10	0.00010	0.18	0.00018
Phenanthrene (Phe)	3	0.001	0.95	0.0009	2.03	0.00203
Anthracene (An)	3	0.001	0.15	0.00015	0.33	0.00033
Fluoranthene (Flut)	3	0.01	2.92	0.0292	5.05	0.05050
Pyrene (Py)	3	0.001	2.93	0.0029	4.92	0.00492
Chrysene (Chr)	2B	0.001	1.77	0.0018	2.1	0.00210
Benzo[a]anthracene (BaA)	2B	0.1	2.32	0.2323	2.87	0.28700
Benzo[k]fluoranthene (BkF)	2B	0.1	1.94	0.1943	1.75	0.17500
Benzo[b]fluoranthene (BbF)	2B	0.1	1.87	0.1868	1.92	0.19200
Benzo[j]fluoranthene (BjF)	2B	0.1	0.54	0.0540	0.76	0.07600
Benzo[e]pyrene (BeP)	3	0.01	1.48	0.0148	1.8	0.01800
Benzo[a]pyrene (BaP)	1	1	1.62	1.6152	2.17	2.17000
Perylene (Prl)	3	0.001	0.27	0.0003	0.38	0.00038
Dibenzo[a,h]anthracene (DBA)	2A	1	0.24	0.2419	0.26	0.26000
Indeno[1,2,3-cd]pyrene (IP)	2B	0.1	1.11	0.1112	1.34	0.13400
Benzo[ghi]perylene (BghiP)	3	0.01	1.61	0.0161	1.56	0.01560
Retene (Ret)	D		0.7		0.58	
Total carcinogenic PAHs			11.20		13.30	
Total BaP equivalent (BaP_{eq})				2.70		3.39
Carcinogenic risk				2.97×10^{-6}		3.73×10^{-6}

1: cacinogenic to humans; 2A: probably carcinogenic to humans; 2B: possibly carcinogenic to humans; 3: not classifiable as to its carcinogenicity to humans; D: inadequate information to assess carcinogenic potential, [a] TEF adopted from Nisbet and LaGoy (1992).

hardwood combustion emissions and they were not detected in softwood emissions. These were also found in significant amounts in the ambient air of the residential site, suggesting that these compounds can be used as tracers for hardwood combustion in the urban and rural atmosphere, although long-term stability of these compounds has not yet been defined.

Using the emission ratio of syringaldehyde and acetosyringone to PM_{10}, the contribution of hardwood combustion to ambient PM_{10} pollution can be quantified according to the following equation:

$$\text{Contribution} = (\text{Syringaldehyde}/PM_{10})_{ambient}/(\text{Sringaldehyde}/PM_{10})_{emission} \times 100$$
$$= (\text{Acetosyringone}/PM_{10})_{ambient}/(\text{Acetosyringone}/PM_{10})_{emission} \times 100 \quad (3)$$

The emission factors of syringaldehyde and acetosyringone may vary due to different types of wood species, different wood combustion appliances, burn rate, different air flow settings and moisture content in the fuel (Fine *et al.*, 2001, 2002; Gullett *et al.*, 2003; Fine *et al.*, 2004; Jordan and Seen, 2005; Hedberg *et al.*, 2006). In fine particle emissions from wood stoves and fireplace combustion, syringaldehyde and acetosyringone were found as most abundant hardwood marker compounds accounting for 0.26–3.6% of $PM_{2.5}$ (Fine *et al.*, 2001, 2002, 2004). In our investigation the emission factors (mass fraction) of syringaldehyde and acetosyringone were in the range of 0.03–0.37 mg/g PM_{10} and 0.71–2.47 mg/g PM_{10}, respectively. Since it is quite difficult to measure the average mass fraction of syringaldehyde and acetosyringone from all wood stoves used in the residential site, we followed an approach where we investigated the linear regression between the ambient

concentrations of syringaldehyde and acetosyringone with PM_{10} as depicted in Fig. 5. From the least-square fits of the ambient concentrations of syringaldehyde and acetosyringone with PM_{10}, average mass fractions of syringaldehyde and acetosyringone were 0.7 mg/g PM_{10} (i.e., 0.07% of PM_{10}) and 2.5 mg/g PM_{10} (i.e., 0.25% of PM_{10}), respectively. Significant correlations were found between syringaldehyde and PM_{10} ($R^2 = 0.79$) as well as between acetosyringone and PM_{10} ($R^2 = 0.78$).

It was considered that all particles in PM_{10} emitted from wood smoke emissions were dispersed equally, and wood combustion was the only major source of ambient PM_{10} pollution. Assuming syringaldehyde as 0.07% and acetosyringone as 0.25% of PM_{10} in wood smoke emissions, the estimated relative contribution of hardwood firing to wintertime ambient PM_{10} pollution was found to be 57 ± 45% and 51 ± 42%, respectively. From our previous investigation (Bari *et al.*, 2010a), using levoglucosan as general wood combustion tracer, the impact of wood smoke was found as 59% of ambient PM_{10} pollution. From local statistics we know that people burn mainly hardwood for heating (more than 90% of firewood). This indicates that the results found in this study seem to be plausible and are in good agreement with the reality. Therefore, a significant impact of hardwood combustion to ambient PM_{10} pollution was found in the investigated residential site.

CONCLUSIONS

Wood as renewable fuel is considered as sustainable energy source for domestic heating. If incomplete combustion takes place from manually fed wood stoves with batch combustion in residential villages, considerable
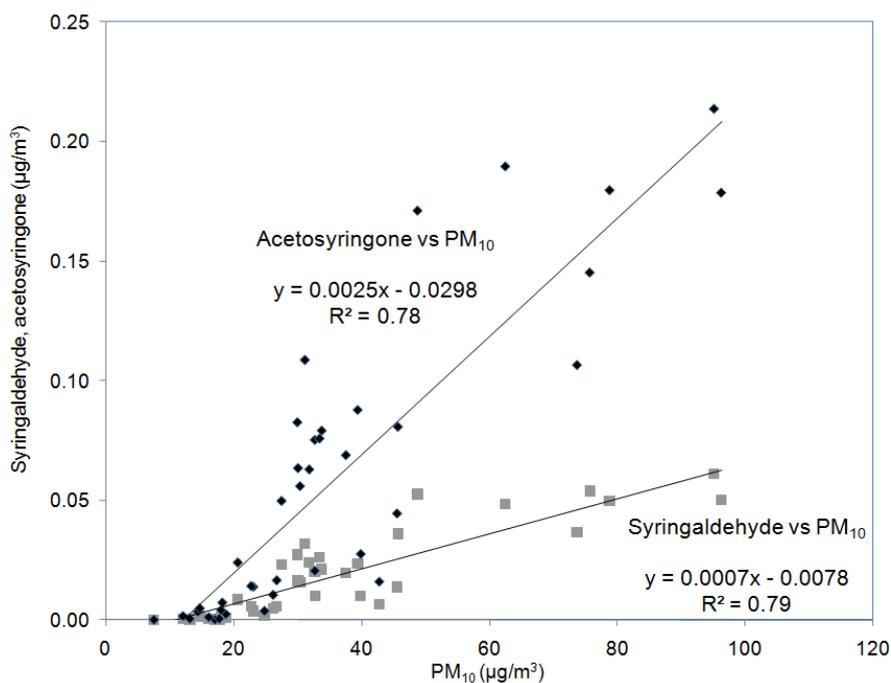
Fig. 5. Correlation of ambient concentrations of syringaldehyde and acetosyringone with PM_{10}.

amounts of particles can be emitted. In this study, high concentrations of PM_{10} and total PAHs were found to be higher during the wintertime. These high concentrations occurred during poor atmospheric exchange conditions (inversions). It was found that during winter 2009, 44% of the total particle-bound PAHs in the ambient air have carcinogenic potential, indicating reasonable concentrations of carcinogenic PAHs in the air which have to be considered and avoided to prevent health risks. From risk assessment it can be seen that total carcinogenic risk during winter in Dettenhausen was found similar during winter 2009 like winter 2005/06. In ambient PM_{10} significant amounts of hardwood smoke markers (i.e., syringaldehyde, acetosyringone) were found which suggests that hardwood smoke has a considerable contribution to ambient PM_{10} pollution in residential areas. Using emission ratio of hardwood markers and PM_{10}, it can be concluded that during winter months the contribution from hardwood combustion to ambient PM_{10} pollution is reached to 57% in the investigated residential site. This confirms that wood-fired house heating can be a dominant source of particulate air pollution in residential villages of southern Germany, when wood is used as main or additional fuel for heating.

ACKNOWLEDGMENTS

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