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Temporal variation and impact of wood smoke pollution on a residential area in southern Germany

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ABSTRACT

This paper is a continuation of our previous publication (Bari, M.A., Baumbach, G., Kuch, B., Scheffknecht, G., 2009. Wood smoke as a source of particle-phase organic compounds in residential areas. Atmospheric Environment 43, 4722–4732) and describes a detailed characterisation of different particle-phase wood smoke tracer compounds in order to find out the impact of wood-fired heating on ambient PM_{10} pollution in a residential area near Stuttgart in southern Germany. The results from previous flue gas measurements help distinguishing different tracer compounds in ambient PM_{10} samples. In the residential area, significant amounts of hardwood markers (syringaldehyde, acetosyringone, propionylsyringol, sinapylaldehyde) and low concentrations of softwood markers (vanillin, acetovanillone, coniferyldehyde, dehydroabietic acid, retene) were found in the ambient air. The general wood combustion markers Levoglucosan, mannosan and galactosan were detected in high concentrations in all particle-phase PM_{10} samples. To find out the size distribution of ambient particles, cascade impactor measurements were carried out. It was found that more than 70% of particulate matter was in the particle diameter of less than 1 μm. Using emission ratio of levoglucosan to PM_{10}, it can be demonstrated that during winter months 59% of ambient PM_{10} pollution could be attributed to residential wood-fired heating.

1. Introduction

This study is a continuation of our previous investigation about wood smoke pollution in residential areas in southern Germany (Bari et al., 2009). In that study, we determined particle-phase organic compounds emitted from wood stoves, investigated source fingerprints of polycyclic aromatic hydrocarbons (PAHs) of wood combustion and other emissions, characterised ambient levels of PAHs and methoxyphenols in detail and finally implemented the positive matrix factorization model to find out emission sources as well as to quantify the wood smoke contribution to the ambient PM_{10}-bound organic compounds in the residential area. In this continued study, we made a detailed characterisation of the identified wood smoke tracer compounds, including the ambient levels of wood smoke compounds, the size distribution and composition of ambient air particles, and the temporal variation of wood smoke tracer compounds. Furthermore, we determined the impact of wood-fired domestic heating on ambient PM_{10} pollution in the investigated residential site of southern Germany.

In residential villages of Germany, located in forest-rich areas, several people use log wood boilers with and without a heat storage tank for central heating. For additional heating, commonly used combustion appliances are manually fed chimney stoves, tiled stoves, and open fire places. Depending on the operational parameters (e.g., fuel seasoning, fuel distribution inside chamber, kindling approaches etc.), such wood-firings can cause high particulate emissions (Tissari et al., 2008; Johannson et al., 2004). Wood combustion causes regional haze with high PM_{10} concentrations in the ambient air, resulting in considerable annoyance and nuisance with complaints among the inhabitants. Even the inhabitants of such residential villages can often smell a distinct ‘wood smoke odor’ in the air. 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, it was necessary to find out the impact of wood-fired heating on ambient PM_{10} pollution in such residential areas. One common approach is to characterise the organic composition of wood smoke emissions and using some of these compounds as potential tracers (Rogge et al., 1993; Schauer et al., 1996; Zheng et al., 2002). In our previous study (Bari et al., 2009), we determined the organic composition of wood smoke emissions and identified wood smoke tracer compounds. Here in this study, we characterise different identified tracer

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compounds and try to find out the impact of wood smoke on ambient air quality.

In the previous study, we considered wood smoke compounds from the thermal degradation of lignin, cellulose and hemicellulose. As pyrolysis products of wood lignin, methoxylated phenolic compounds (methoxyphenols) have been suggested as potential wood smoke tracers in multivariate source apportionment models to determine the contribution of urban particulate matter (PM) derived from wood combustion (Schauer and Cass, 2000). Smoke from residential wood stoves has been reported to contain syringol (2,6-dimethoxyphenol) and its derivatives (syringaldehyde, acetosyringone, propionylsyringol, sinapylaldehyde) in large amounts in hardwood combustion but no significant quantities are detected in softwood emissions. Whereas guaiacol (2-methoxyphenol) and its derivatives are emitted from both softwood and hardwood combustion but the emission rates of individual guaiacol derivatives (e.g., vanillin, acetovanillone, guaiacylacetone, coniferylaldehyde) are very different for the two types of wood (Schauer et al., 2001; Rogge et al., 1998; Hawthorne et al., 1989). We found similar results in our previous study (Bari et al., 2009).

Monosaccharide anhydrides (MA) such as levoglucosan, mannosan and galactosan are the predominant organic compounds in wood smoke emissions, which are emitted in high quantities from the smouldering stage of combustion and formed from the thermal degradation of cellulose and hemicellulose and have been considered as the candidate tracers for residential wood combustion (Shafizadeh, 1968; Hornig et al., 1985; Locker, 1988; Simoneit et al., 1999; Jordan et al., 2006). Due to certain chemical properties i.e., low vapour pressure with high molecular weight, the partition of levoglucosan is in the particulate phase (Fine et al., 2001; Locker, 1988). Another important property that enables the use of levoglucosan as a potential tracer for particle emissions from wood burning is its resistance to chemical degradation. It is found stable in the atmosphere over a period of 10 days in aqueous solutions (Locker, 1988; Fraser and Lakshmanan, 2000). The levoglucosan to particle emission fraction 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 (Gullett et al., 2003; Fine et al., 2001, 2004a; Hedberg et al., 2006; Jordan and Seen, 2005). Levoglucosan emitted from wood stoves and fireplace combustion, ranged 0.8–32% of $PM_{2.5}$ emissions (Fine et al., 2001; Schauer et al., 2001; Jordan and Seen, 2005). In central Europe comparatively similar results were found from domestic tiled stoves ranging 4–15% levoglucosan in PM_{10} emissions (Schmidl et al., 2008). Levoglucosan has been used extensively to address the impact of wood combustion on local air quality in the USA not only from the wood-fired winter heating, but also from other forms of biomass burning such as wild fires especially in the west and prescribed burning for wide spread forest land management in the east. It is found in high concentrations in the ambient air in several US cities (Simoneit et al., 1999; Fraser and Lakshmanan, 2000; Nolte et al., 2001, 2002) as well as in Europe (Zdráhal et al., 2002; Carvalho et al., 2003; Yttri et al., 2005; Puxbaum et al., 2007; Saarikoski et al., 2008; Caseiro et al., 2009).

To distinguish between softwood and hardwood emissions, resin acids and retene have been suggested as tracers for softwood smoke (Fine et al., 2001, 2002; Standley and Simoneit, 1994; Simoneit et al., 1993; Ramdahl, 1983). During the combustion of coniferous wood, tricyclic resin acids are released due to volatilisation by steam in either their unaltered form, partially altered or completely combusted, where the common altered resin acid is dehydroabietic acid. Dehydroabietic acid may be unstable in the ambient air and it degrades in water after treatment with ultraviolet light (Corin et al., 2000). In our study, we characterised the identified wood smoke tracer compounds to assess the impact of wood-fired domestic heating on ambient PM_{10} pollution in a residential site of southern Germany.

2. Methodology

2.1. Site description

Ambient PM_{10} samples were collected at the residential site in Dettenhausen as we did in our first investigation (Bari et al., 2009), which is located at the northern edge of the nature park area “Schoenbuch”. 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.

2.2. Sampling procedure

2.2.1. Ambient and emission sampling

The ambient PM_{10} sampling was carried out from 1 November 2005 to 31 March 2006. To distinguish between hardwood and softwood tracer compounds, 6 beech wood and 5 pine wood combustion experiments were carried out. The procedure of ambient and emission sampling is described in detail in Bari et al. (2009).

2.2.2. Particle size distribution

To get different size ranges of ambient particles, gravimetric sampling with cascade impactor was carried out with 8-stage nonviable particle size sampler (Andersen Instrument Inc., USA) during February and March 2006. For the particle separation, aluminium foils have been used as collection plates and glass fibre filters (GF10 Schleicher and Schuell, diameter 90 mm) as backup filter. The impactors are operated at 28.3 $m^3\,min^{-1}$ and the effective cut-off diameter of the impactor stages are 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 5.8, 9.1 and 10.0 μm for particles with the assumed density of 1.0 $g\,cm^{-3}$. The change in weight for each stage in the cascade impactor including the backup filter was determined by using an analytical balance. According to the specification of the European Standard EN 12341:1998 (CEN, 1998), filters were equilibrated in the weighing chamber for 48 h under controlled temperature and humidity conditions before and after sampling to prevent hydration of filters' surface.

2.3. Organic chemical analysis

The collected forty-two ambient PM_{10} filter samples were analysed to measure wood smoke tracer compounds by gas chromatography mass spectrometry (GC-MS) using a Hewlett Packard 6890 Gas Chromatograph coupled with a Agilent 5973 Mass Selective Detector (MSD) operating in full scan mode. The GC configuration, temperature profiles as well as filter extraction procedure and quantification of organic tracer compounds are described in detail in another study (Bari et al., 2009). The identified methoxyphenols and other tracer compounds in the wood smoke and ambient samples are not detected in the filter blanks.