

Contribution of wood burning to the exceedance of PM₁₀ limit values in North Rhine-Westphalia

Dedicated to Prof. Dr. Peter Bruckmann on occasion of his 65th birthday

U. Pfeffer, L. Breuer, D. Gladtko, T. J. Schuck

Abstract During the heating period from November 2011 until April 2012 measurements of PM₁₀ and levoglucosan as tracer for wood burning were performed at 21 stations of the air quality monitoring network LUQS in North Rhine-Westphalia (Germany). The levoglucosan concentrations were similar in different parts of the state. Nevertheless, local and/or regional sources of levoglucosan were clearly recognized. The highest daily averages were in the range of 2 to 2.3 µg/m³, corresponding to up to 3.5% of the PM₁₀ concentration. In a residential area with frequent wood burning in private homes measurements of PM₁₀ and levoglucosan were performed in order to determine a conversion factor between levoglucosan and related PM₁₀ concentrations. Using this factor, approximately 20 to 50% of the exceedances of the daily limit value for PM₁₀, as laid down in the European Directive 2008/50/EC, can be ascribed to the additional impact of wood burning during the six months from November 2011 until April 2012. Consequently, there is considerable potential to decrease urban PM levels by optimization of wood burning in stoves and fireplaces, e.g. by installation of filters, or by regulating usage of small combustion units during inversion periods accompanied by elevated PM levels.

Beitrag der Holzverbrennung zur Überschreitung von PM₁₀-Grenzwerten in Nordrhein-Westfalen

Zusammenfassung In der Heizperiode von November 2011 bis April 2012 wurden an 21 Stationen des nordrhein-westfälischen Luftqualitätsmessnetzes LUQS Messungen von PM₁₀ und Levoglucosan als Tracer für Holzverbrennung durchgeführt. Es zeigte sich, dass die Levoglucosankonzentrationen in den verschiedenen Teilen des Landes ähnlich waren. Lokale bzw. regionale Quellen von Levoglucosan waren jedoch klar erkennbar. Die höchsten Tagesmittelwerte lagen im Bereich von etwa 2 bis 2,3 µg/m³, entsprechend bis zu 3,5 % der PM₁₀-Konzentrationen. In einem städtischen Wohngebiet mit häufiger Nutzung von Einzelfeuerstätten in Privathäusern wurden Messungen von PM₁₀ und Levoglucosan durchgeführt, um einen Konversionsfaktor zu bestimmen, mit dem aus der gemessenen Levoglucosankonzentration die dadurch hervorgerufene PM₁₀-Konzentration ermittelt werden kann. Mithilfe dieses Faktors wurde abgeschätzt, dass im Zeitraum von November 2011 bis April 2012 zwischen etwa 20 und 50 % der Überschreitungen des PM₁₀-Tagesgrenzwertes der 39. BImSchV bzw. der zugrundeliegenden EU-Richtlinie 2008/50/EG dem zusätzlichen Beitrag von Holzfeuerungen zugeordnet werden kann. Somit besteht ein beträchtliches Potenzial zur Senkung der PM-Belastung in städtischen Gebieten durch die Optimierung von Einzelfeuerstätten, z. B. durch den Einbau von Filtern, oder Betriebsregulierungen für kleine Einzelfeuerstätten während austauscharmer Wetterlagen mit hohen PM-Konzentrationen.

Dr. Ulrich Pfeffer, Ludger Breuer, Dr. Dieter Gladtko,
Dr. Tanja J. Schuck,

Landesamt für Natur, Umwelt und
Verbraucherschutz NRW, Essen.

1 Background and introduction

During the last years, PM concentrations in North Rhine-Westphalia (NRW) were significantly reduced by various abatement measures, among others the implementation of air quality plans. Nevertheless, European limit values laid down in Directive 2008/50/EC [1] are still exceeded in NRW and many other regions in Europe, especially in years with unfavourable meteorological conditions like 2011. Therefore, it is of high interest to identify relevant sources of PM in addition to the well-known sources such as traffic and industrial installations.

Numerous studies have shown that biomass/wood burning contributes significantly to the PM burden. For example, Puxbaum et al. showed that during wintry conditions wood smoke may contribute more than 50% to organic material in the atmosphere at rural flat terrain sites in Europe [2]. Measurements in Tübingen (Baden-Württemberg, Germany) during the winter months 2008/2009 revealed a share of 25% of wood smoke in PM₁₀ concentrations [3; 4]. Piazzalunga et al. found that in the city of Milan wood smoke caused 20 to 25% of organic material and up to approximately 15% of PM₁₀ [5]. Recently, a study in Flanders showed that the number of days with PM₁₀ daily averages above 50 µg/m³ (EU limit value) could be reduced by 47% after "virtual" subtraction of the PM₁₀ mass due to wood burning [6].

In order to access the share of wood or biomass burning in PM, the specific tracer substance levoglucosan (a monosaccharide anhydride) is used because this substance is almost exclusively produced by burning of cellulose-containing material. It is stable in the atmosphere for roughly ten days during wintry conditions [7]. Different concepts to derive a conversion factor relating levoglucosan concentrations to particle mass concentrations have been published. They are based e.g. on emission data, ambient air data or modelling results. This factor depends, inter alia, on the type of the combustion unit and its operation mode as well as on the type of wood. Therefore, the conversion factor can differ regionally. A study in Baden-Württemberg [3; 4] used a conversion factor of 8 µg PM₁₀/µg levoglucosan stemming from a Bavarian project [8; 9]. For Austria, an average conversion factor of approximately 11 was found in wood burning experiments [10]. A newer study by the same group of authors using more realistic burning conditions suggested that for manually fired stoves a conversion factor of 20 may be more appropriate (5% levoglucosan in particle emissions) [11]. In this context it is important to know that the number of single stoves and fireplaces sold per year in Germany increased significantly during the last years and reached a plateau in 2007 at approximately 400,000 installations per year. The total number of installations in Germany was esti-

Table 1. Total number of single stoves in European countries [12].

Country	Number of single stoves
Austria	238,000
Germany	5,816,000
Italy	1,268,000
France	2,505,000
EU-27	25,901,000

stations were represented (traffic, industry, urban, sub-urban, background, rural). Samples were generally taken every second day, at six stations daily (Table 2).

2.2 Analysis of levoglucosan and other monosaccharide anhydrides

Sampling of PM₁₀ is performed on quartz fibre filters (Pallflex TISSUQUARTZ TM 2500 QAT-UP) of 150 mm diameter according to DIN EN 12341 [14] using a Digital DH-80 device

Table 2. Measurement sites and sampling frequency. *u* = urban, *su* = sub-urban, *r* = rural, *b* = background, *i* = industry, *t* = traffic, *d* = daily, *sd* = every second day

Code	Station	Type	Sampling frequency
BIEL	Bielefeld-Ost	u-t	sd
BORG	Borken-Gemen	r-b	sd
BOTT	Bottrop-Welheim	u-i	sd
BUCH	Duisburg-Buchholz	su-b	sd
DDCS	Düsseldorf, Corneliusstraße	u-t	d
DMD2	Dortmund-Eving	u-b	sd
DUBR	Duisburg-Bruckhausen	u-i	d
DUM2	Duisburg, Kiebitzmühlenstraße	u-i	d
EIFE	Eifel, Simmerath	r-b	sd
GBFD *)	Grevenbroich-Frimmersdorf, Josef-Lützkirchen-Straße	u-i	sd
GELS	Gelsenkirchen-Bismarck	su-b	sd
GRGG	Grevenbroich-Gustorf	su-i	sd
KRES	Krefeld-Stahldorf	u-i	d
MHHS	Mülheim, Hofackerstraße	u-i	d
STYR	Mülheim-Styrum	u-b	d
VACW	Aachen, Wilhelmstraße	u-t	sd
VKTU	Köln, Turiner Straße	u-t	sd
VMGR	Mönchengladbach, Düsseldorfer Straße	u-t	sd
VWEL	Wuppertal-Gathe	u-t	sd
WALS	Duisburg-Walsum	u-i	sd
WAST	Warstein	u-i	sd

*) Since January 2012. Because the data from this station do not cover the complete evaluation period they were excluded from various calculations.

Table 3. Chromatographic conditions for the analysis of anhydrosugars.

Column	Metrosep Carb 1 – 150/4
Temperature	24 °C
Eluent	1.5 g NaOH and 0.4 g sodium acetate in 1 kg of water
Flow	0.7 ml/min at ca. 5.5 MPa
Loop	100 µl
Analysis time	15 min
Pulse	E1: +0.1 V; t1: 0.4 s; E2: +0.9 V; t2: 0.1 s; E3: -0.15 V; t3: 0.5 s; ts: 200 ms
Detection limit	10 ng/m ³

ated to be approximately 5.8 million in 2009. Table 1 shows the number of single stoves in some European countries.

2 Experimental

2.1 Sampling sites and PM₁₀ measurements

The measurements during the heating period from November 2011 until April 2012 were performed at 21 monitoring sites of the ambient air quality monitoring network LUQS in North Rhine-Westphalia (NRW) [15]. The sites were selected such that different regions of NRW and different types of

equipped with a PM₁₀ sampling head [15]. The sampled volume is about 720 m³ during 24 h. Up to six filter sections of 25 mm diameter are shaken for 60 min with 40 ml of water (ultrapur, electrical resistivity > 18 MΩ) in a 50 ml PE centrifuge tube with plug style cap. Analysis is carried out by ion chromatography, the conditions are summarized in Table 3. Extract work up and sample injection are combined using a sample processor (Metrohm 853), equipped with a polyamide inline filter (Metrohm

Ultrafiltration). The analytes are filled (1 ml during 2 min) into the sampling loop with a counter flow of the eluent (see Table 3). 100 µl of the solution are injected.

Carbonate, silicate and borate reduce the retention time of the anhydrosugars and cause a reduction of the separation capacity of the column. Therefore, every contact of the eluent solution with carbonate, carbon dioxide (air), silicate and borate (glassware) has to be avoided carefully. The column can be regenerated with an eluent of 10 g NaOH (50% aqueous solution) and 50 g of sodium acetate per kg of water. The flow shall be 1 ml/min for 4 h to 10 h.

Calibration is carried out with arabitrol, manitol, levoglucosan, mannosan, galactosan and glucose at concentrations of 100, 400, 800 and 1,600 ng/g water. The solutions are analysed before, between and after each analysis batch; they have to be prepared weekly.

The precision of the analytical method was estimated according to DIN ISO 5725-2 [16]. 13 different extracts of one sample were analysed four times each. The relative repeatability standard deviation was 2.3%, the relative standard deviation due to extraction was 1.5%. Thus, the relative uncertainty of the extraction and measurement procedure is 2.7% and the expanded uncertainty at the 95% confidence level is 5.4%.

3 Results and discussion

Figure 1 shows daily averages of PM_{10} from four selected stations (see Table 2). Episodes with PM_{10} concentrations above $50 \mu\text{g}/\text{m}^3$ occurred especially in November 2011 and in January to March 2012 on days with unfavourable dispersion conditions.

3.1 Temporal and spatial variation of levoglucosan concentrations

Figure 2 shows levoglucosan concentrations (daily averages) at selected stations across NRW. The concentration patterns are similar, although these stations lie in different parts of the state within different surroundings (Table 2), demonstrating that levoglucosan can be considered ubiquitous and its concentration level is highly variable. The highest peaks were predominantly measured during episodes with unfavourable meteorological conditions and inversion heights of less than 400 m. The maximum daily average was encountered at the urban industrial station DUM2 with a value of $2.3 \mu\text{g}/\text{m}^3$.

At the station EIFE, in contrast, levoglucosan concentrations are less variable and do not show pronounced peaks. This site is located at a height of 572 m in the Eifel mountains, in a rural area remote from strong pollution sources. Owing to its elevation, the site is likely to be above inversion layers. Therefore, local sources and transport within the boundary layer may play a minor role at this site during inversion episodes, when high levels of levoglucosan and PM_{10} are observed at the other stations.

Whilst **Figure 2** exhibits absolute concentrations of levoglucosan in $\mu\text{g}/\text{m}^3$, **Figure 3** shows the relative share of levoglucosan in PM_{10} . The patterns of the relative concentrations are similar to those of the absolute concentrations. Thus, during winter, high PM_{10} concentrations are linked to high levoglucosan values as well as high percentages of levoglucosan in PM_{10} , clearly demonstrating that wood burning contributes an important share to the PM_{10}

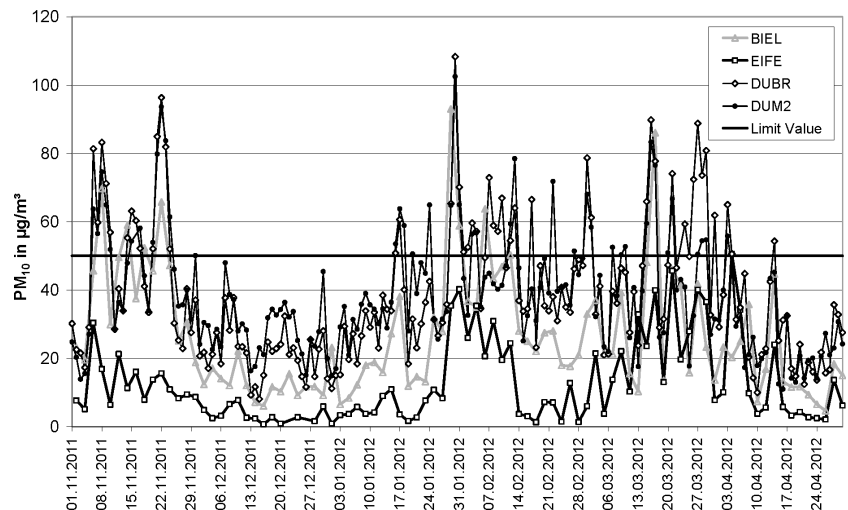


Figure 1. Daily averages of PM_{10} at different stations in NRW (November 2011 to April 2012).

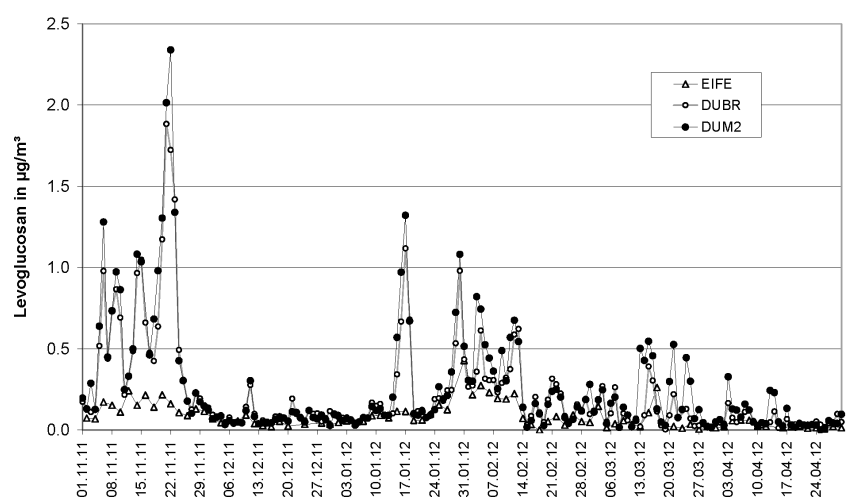
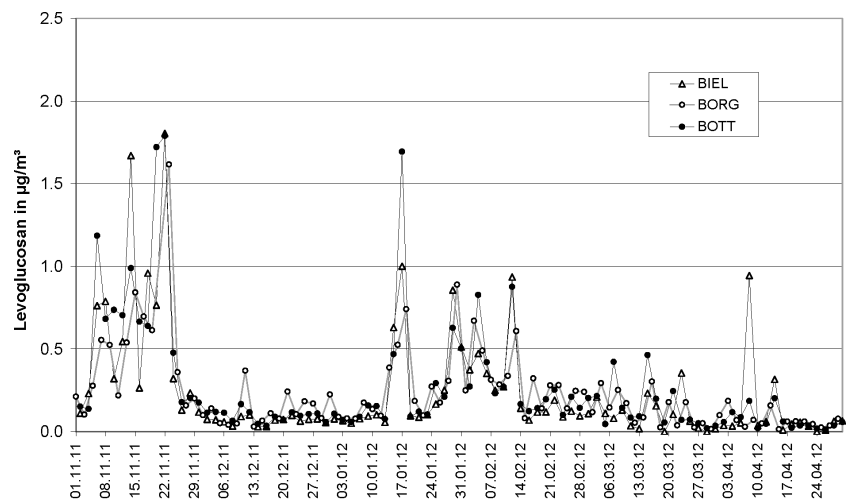


Figure 2. Daily averages of levoglucosan at different stations in NRW (November 2011 to April 2012).

load during pollution episodes. Again, station EIFE shows a specific behavior: the percentage of levoglucosan in PM_{10} is partly higher than at other stations, especially when the absolute values are very low (e.g. December 2011 to January 2012).

The highest absolute values of levoglucosan were measured in November 2011, a month with low temperatures and low

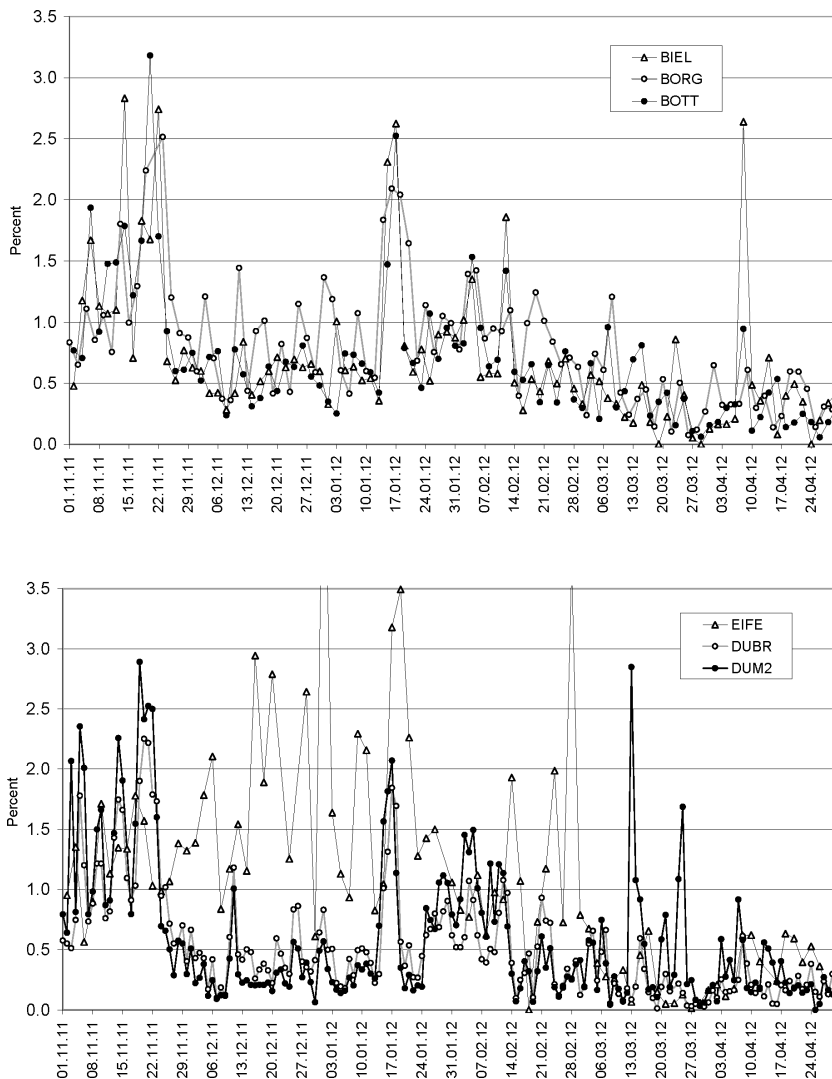


Figure 3. Relative shares of levoglucosan in PM₁₀ at different stations in NRW (November 2011 to April 2012).

wind speeds. Monthly averages for all stations varied between 0.4 and 0.7 µg/m³, except for the station EIFE which stood out with a value of only 0.14 µg/m³ (Figure 4). The low concentration of levoglucosan at this rural background station and the higher values in urban areas indicate that levoglucosan in urban areas mostly originates from regional and local sources, while long-range transport of aerosols contributes a minor share. The relative share of levoglucosan in PM₁₀ varied between 1.0 and 1.7 %. While the station EIFE behaved differently on a daily basis (see Fig. 3), this is not reflected in the monthly averages (see Fig. 4).

3.2 Conversion factor relating levoglucosan to PM₁₀ concentrations

During the heating period from November 2011 until April 2012, PM₁₀ measurements were conducted in a residential area in Essen at a site where frequent wood burning takes place in stoves and fireplaces, especially during evenings and weekends.

Two low volume samplers (LVS, 2.5 m³/h) with PM₁₀ sampling heads were operated as follows:

- Device A: 10 days sampling; from 18.00 to 24.00 (evening; e),
- Device B: 10 days sampling; from 10.00 to 16.00 (day; d)

Sampling over ten days ensured enough PM mass for the analysis. PM₁₀ was sampled on 47 mm Whatman GF 10 filters and analysed gravimetrically according to DIN EN 12541 [14], for the analysis of levoglucosan see Section 2.

This monitoring concept is based on the assumption that PM₁₀ emissions from wood burning are generally higher during evenings. Levoglucosan can be used as a tracer to quantify the contribution of wood burning to PM₁₀. It is assumed that this share can be calculated by multiplying the levoglucosan concentration by a factor F, Eq. (1):

$$PM_{10; \text{wood}} = \text{Levoglucosan (LG)} \cdot F \quad (1)$$

The concept requires the knowledge of the ratio X of “hypothetical PM₁₀ concentration without any influence of wood burning during evenings (PM_{10; e} - LG_e · F) to those during days (PM_{10; d} - LG_d · F), Eq. (2):

$$(PM_{10; e} - LG_e \cdot F) / (PM_{10; d} - LG_d \cdot F) = X \quad (2)$$

(e: evening; d: day; LG: levoglucosan)

If this ratio X is known and the concentrations of PM₁₀ and levoglucosan during days and evenings are measured the conversion factor F can be calculated from Eq. (2).

During summer months, almost no levoglucosan is found in PM₁₀ samples. Figure 5 shows the relation of evening values (18.00 to 24.00 h) to daytime values (10.00 to 16.00 h) of PM₁₀ continuously measured at 55 monitoring sites in NRW [15] over one year.

Wood burning does not contribute a relevant part to PM₁₀ mass during summer. Therefore the concentrations of levoglucosan are very

low at this time: from May to August the PM_e/PM_d ratio is very close to 1. Provided that the day/evening ratio of PM₁₀ without the contribution of wood burning is close to 1 also in winter, X in Eq. (2) can be considered to be approximately 1. Then the factor F can be calculated according to Eq. (3):

$$F = (PM_{10; e} - PM_{10; d}) / (LG_e - LG_d) = \Delta PM_{10} / \Delta LG \quad (3)$$

Taking all day/evening measurement data from November 2011 to April 2012 it was found that levoglucosan concentrations were always higher during evenings than during days. Using Eq. (3), the mean value of F is approximately 13 with a range from 6 to 27 for single periods.

This experimentally derived conversion factor of 13 is in good agreement with other studies (see Section 1) and was therefore applied in all following evaluations.

3.3 Contribution of wood burning to average PM₁₀ concentrations

Figure 6 shows daily PM₁₀ averages measured at the station Mülheim (MHHS). The share of the PM₁₀ concentrations which can be attributed to wood burning is marked in black; the other sources as traffic, industry or secondary aerosols are summarized in the hatched columns. If the horizontal

line for the daily limit value of $50 \mu\text{g}/\text{m}^3$ [1] crosses the black parts of the columns, the exceedances of the limit value could – theoretically – have been avoided without the additional PM_{10} share caused by wood burning, at the station MHHS for example on eleven days.

3.4 Contribution of wood burning to exceedances of daily PM limit values

Figure 7 summarizes the results of 20 stations in the period from November 2011 to April 2012. The columns show the number of days with daily PM_{10} averages above $50 \mu\text{g}/\text{m}^3$. Again, the black parts of the columns stand for the number of exceedance days which can be ascribed to the additional PM_{10} input caused by wood burning. Apart from the station in the Eifel mountains (EIFE), this fraction varies between 20% at the traffic station DDCS in Düsseldorf and 55% at the station in Duisburg Walsum (WALS) which is situated in a residential area with some influence of industrial sources. Even at the highly polluted stations DUBR (Duisburg-Bruckhausen) and DUM2 (Duisburg-Marxloh) in surroundings with heavy industries, 13 to 14 exceedance days could theoretically have been avoided without the additional impact of PM_{10} by wood combustion.

Because the conversion factor from levoglucosan to PM_{10} has a considerable uncertainty (see Section 3.2), a sensitivity analysis was performed using conversion factors between 6 and 15 (Figure 8). A factor of 6 is the lowest single value found in this study (see Section 3.2) whilst a factor of 15 is still clearly below the value of 20 proposed recently [11].

As expected, the number of exceedances related to the additional contribution of wood combustion clearly depends on the conversion factor used. But even using the lowest factor of 6, this number remains considerable: more than 35% of exceedance days could be attributed to the additional burden caused by wood burning in the highly polluted November 2011.

4 Summary

- During the heating period from November 2011 until April 2012 levoglucosan as tracer for wood burning was measured at 21 stations of the North Rhine-Westphalian air quality monitoring network LUQS. Concentration levels were similar across the state, even at different types of stations.

- On days with high levoglucosan concentrations also the relative share in PM_{10} is elevated. This underlines the influence of wood burning on the PM_{10} levels.

- The difference between the levoglucosan concentrations at a rural background site and at sites in urban agglomerations points out the importance of local and regional sources.

- Highest daily averages of levoglucosan are in a range be-

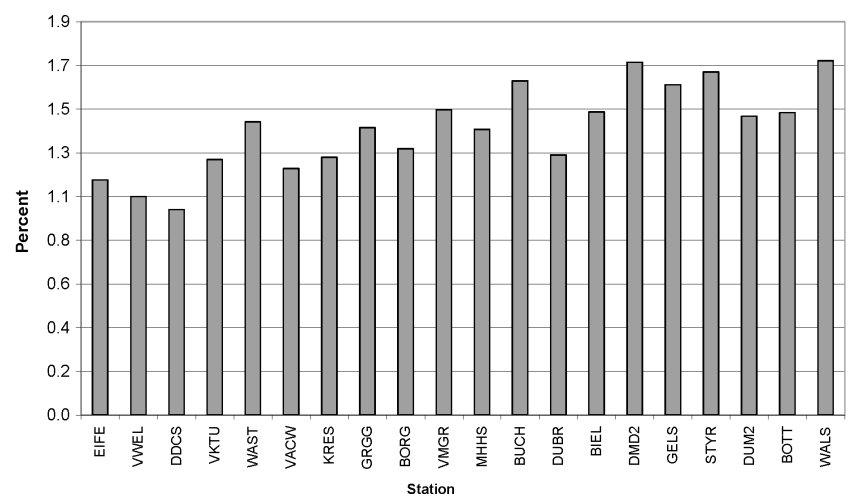
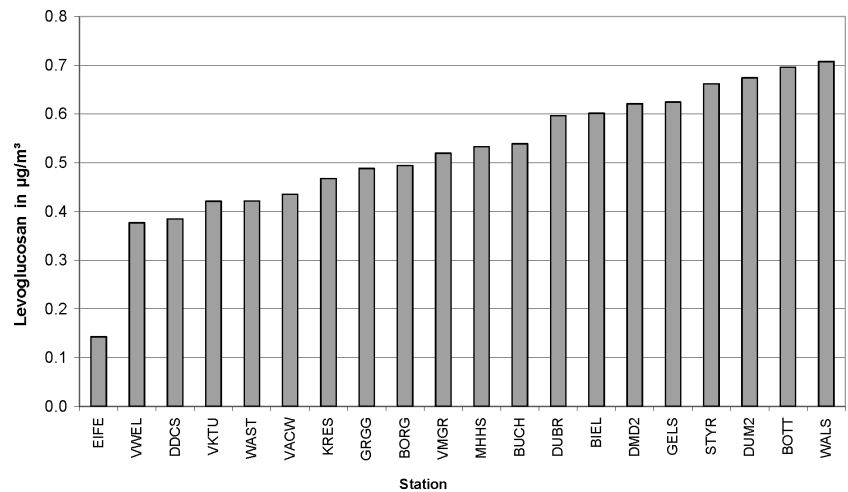


Figure 4. Monthly averages and shares of levoglucosan at different stations in NRW in November 2011.

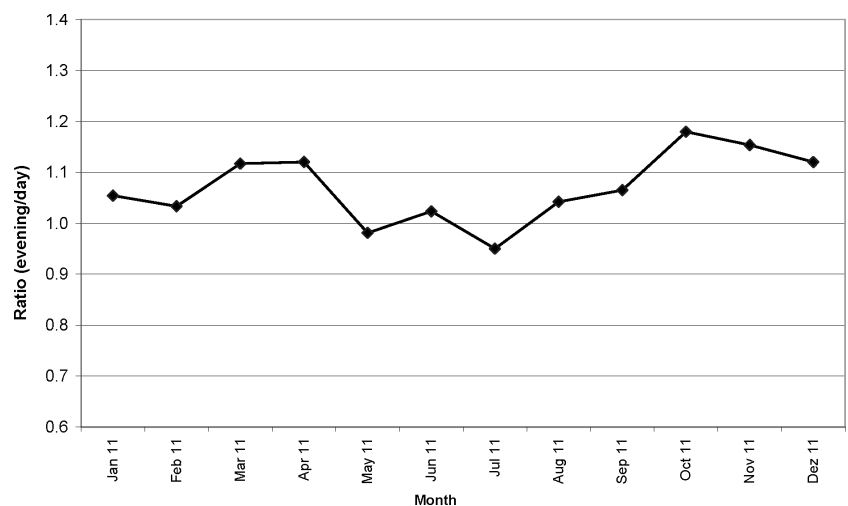


Figure 5. Ratio of PM_{10} concentrations during evenings and days in 2011 (average calculated from 55 stations).

tween 2 and $2.5 \mu\text{g}/\text{m}^3$ (up to 3.5% of the PM_{10} concentration).

- In November 2011, monthly averages of levoglucosan were generally in the range from 0.4 to $0.7 \mu\text{g}/\text{m}^3$, corresponding to 1 to 1.7% of PM_{10} (except station Eifel with only $0.14 \mu\text{g}/\text{m}^3$, corresponding to 1.2%).

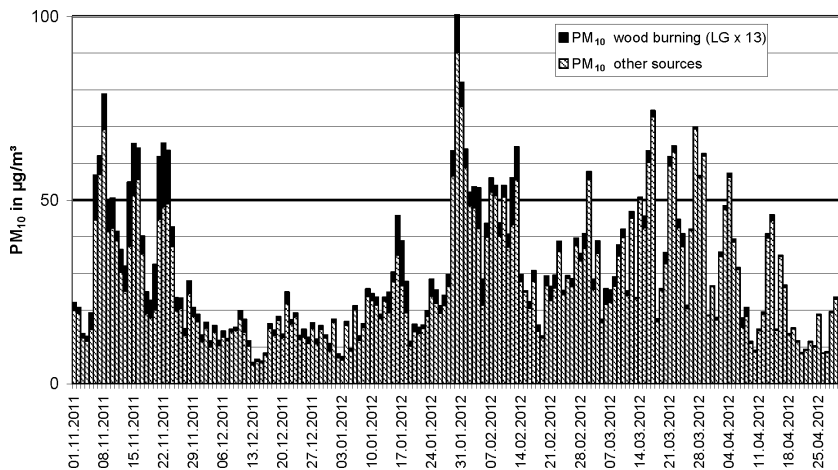


Figure 6. Daily averages of PM₁₀ and contribution of wood burning measured at the station Mülheim (MHHS) in winter 2011/2012.

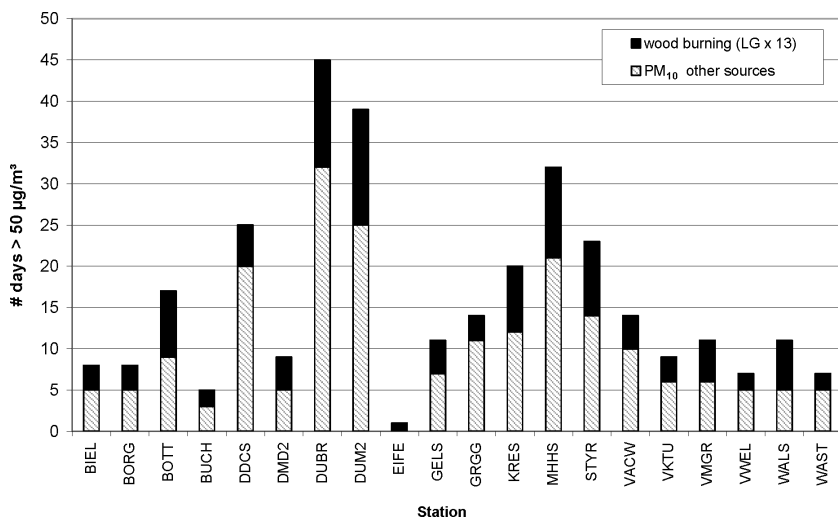


Figure 7. Number of days with exceedances of the daily PM₁₀ limit value of 50 µg/m³ (November 2011 to April 2012).

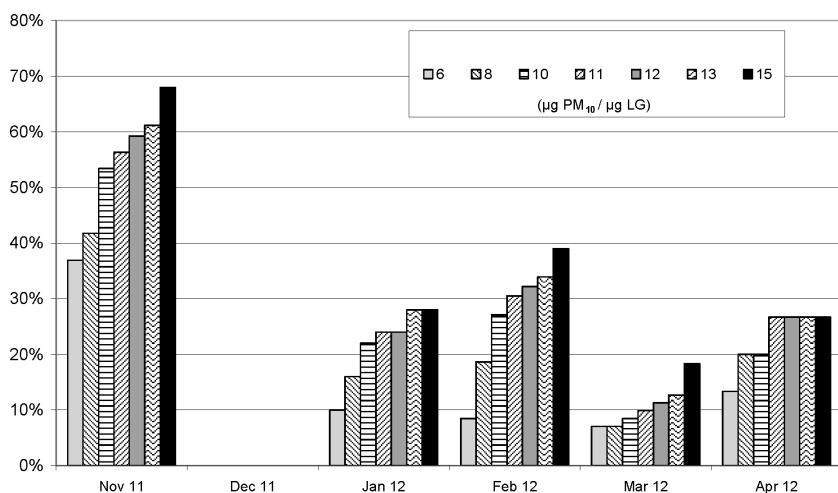


Figure 8. Percentage of days with exceedances of the daily PM₁₀ limit value of 50 µg/m³ in winter 2011/2012 calculated with conversion factors between 6 and 15 for 21 sites in NRW.

- The experimental determination of a conversion factor relating the PM concentrations stemming from wood burning to levoglucosan levels is difficult, because reference stations which are unaffected by wood combustion during the cold season are not available. Therefore, additional measurements were performed.

- From variations of the levoglucosan concentrations between day and evening a conversion factor of 13 was derived. The application of this factor suggests that between approximately 20 and 50% of exceedance days for the PM₁₀ daily limit value can be ascribed to the additional impact of wood burning during the six months from November 2011 until April 2012.

- The contribution of wood burning to the PM₁₀ burden is relatively low as annual average. However, it is concentrated on days and episodes during the winter season when PM concentrations are already elevated due to unfavourable meteorological conditions.

5 Conclusions and outlook

Although the quantitative determination of the PM₁₀ share caused by wood burning is uncertain, a sensitivity analysis shows that this contribution is significant. Consequently, there is a considerable potential for the decrease of PM levels by optimizing wood burning in stoves and fireplaces (e.g. installation of filters) or regulating the use of small combustion units during periods with bad dispersion conditions and elevated PM levels. Studies have shown that emissions from fire places and wood stoves depend crucially on the burning material, but even more on the construction and on the operating procedures of these installations. Single stoves and fire places are particularly relevant because they produce higher particle emissions than automatic pellet stoves [11].

Levoglucosan is predominantly present in the particulate phase. During winter time, the lifetime of levoglucosan is of the order of ten days and thus close to the lifetime of fine particles in the atmosphere [7]. Consequently, levoglucosan particles can be transported over long distances as other fine particles. Whether such long range transport is of importance under real conditions will be investigated in further studies.

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