Comparison of biomass burning tracer concentrations between two winter seasons in Krynica Zdrój

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Abstract

Biomass burning is one source of air pollution that emits soot, smoke, particulate matter, free radicals and other substances, affecting human health, air quality, cloud formation processes and climate change. During biomass burning, organic biomass burning tracers are emitted, such as levoglucosan and its isomers mannosan and galactosan, which are formed at temperatures above 300 °C during cellulose and hemicellulose pyrolysis. The objective of this research is the measurement of biomass burning tracer concentrations in PM₁₀ samples in Krynica during two winter seasons from 01.12.2017 to 31.03.2018 and from 01.12.2018 to 26.03.2019. The average concentrations of organic carbon were 9.48 µg/m³ in the 2017/2018 season and 8.79 µg/m³ in the 2018/2019 season. The average concentration of levoglucosan in the PM₁₀ samples in 2017/2018 was 137 ng/m³, while in 2018/2019, the average concentration reached 245 ng/m³. Levoglucosan was the dominant compound among the determined biomass burning tracers, accounting for 88.6% in 2017/2018 and 72.4% in 2018/2019. The conducted measurements show that the levoglucosan to mannosan ratio was equal to 10.5 and 3.9 in 2017/2018 and 2018/2019, respectively. The biomass smoke organic carbon was approximately 10.6% in 2017/2018 and 20.5% in 2018/2019. These results correspond to the contribution of biomass smoke to organic carbon, which was equal to 14.4% and 28.4% in the 2017/2018 and 2018/2019 seasons, respectively.

Keywords Biomass burning · Tracers · Levoglucosan · Mannosan · Galactosan

Introduction

Biomass burning is a global phenomenon resulting from forest fires and slashes and the burning of forests for agricultural, agricultural waste burning and residential uses. Biomasses, such as straw; wood in the form of pellets; granules or sawdust; and waste, such as scales, can be used as fuel during direct combustion or co-firing. Biomass burning (BB) is a source of air pollution that emits particulate matter, free radicals and other substances, which affect human health, air quality, cloud formation processes and climate change (Schkolnik and Rudich, 2006). In recent years, several organic molecular markers have been widely used in terms of monitoring environmental pollution with biomass burning. During biomass burning, organic biomass burning tracers are emitted, such

as levoglucosan (LG) and its isomers mannosan (MN) and galactosan (GA), which form at temperatures above 300 °C during cellulose and hemicellulose pyrolysis. These three tracers are most suitable and recognized due to their high abundance and relatively long stability under specific environmental conditions (Fabbri et al. 2009; Zhang et al. 2010; Simoneit et al. 1999; Simoneit 2002). As levoglucosan is the most abundant among all mentioned biomass burning tracers (Schmidl et al. 2008; Simoneit et al. 1999), the relative abundance of LG to MN and GA in atmospheric particulate matter (PM) is affected by the type of biomass burnt (Fabbri et al. 2009; Schmidl et al. 2008). The LG/MN ratio is higher than 50 for a poor quality brown coal, while for softwood and hardwood/grass combustion, it is approximately 5 and 10–20, respectively. The number of grasses is approximately 25 (Fabbri et al. 2009; Schmidl et al. 2008), and the burning of rice straw is 26.6 (Engling et al. 2009). The LG/MN ratio can be used to distinguish wood type burnt, but it makes it difficult to separate emissions from hardwood and crop residual burning (Cheng et al. 2013). The concentration of biomass burning tracers can be determined by different techniques, but the most popular techniques are gas chromatography with mass

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spectrometry (GC/MS) and high-performance liquid chromatography with pulsed amperometric detection (HPLC/PAD) (Schkolnik and Rudich 2006; Janoszka and Czaplicka 2019). In this research, the concentration of biomass burning tracers was determined by GC/MS.

In Poland, the levels of biomass burning tracers are poorly documented. Consequently, the European Union has funded research on air quality to determine biomass burning tracer concentrations. To address this shortcoming, this report focuses on the following objectives: (i) determination of the level of biomass burning tracer concentrations in PM₁₀ samples from the health resort Krynica during two winter seasons, 2017/2018 and 2018/2019; (ii) determination of individual biomass burning tracer percentages from the total sum of tracers in PM₁₀; (iii) determination of LG/MN and LG/(MN+GA) ratios; and (iv) estimation of organic carbon contribution from biomass burning in the total OC.

Materials and methods

Samples of PM₁₀ were collected over a 24-h cycle on 47 mm Whatman QMA quartz filters at the measurement station Krynica (health resort $\varphi = 49^\circ 24' 28''$, $h = 20^\circ 57' 39''$, $h = 582$ m above sea level) from 01.12.2017 to 31.03.2018 (118 samples) and from 01.12.2018 to 26.03.2019 (112 samples). Samples were collected in accordance with the methodology described previously (Klejnowski et al. 2017) with the use of a low flow sampler with separating head PM₁₀ Atmoservice PNS3D15/LVS3d, with a stabilized flow of 2.3 m³/h. Blank samples were checked before every month (8 in total).

The concentrations of biomass burning tracers were determined based on the method described by Poore (2002). Levoglucosan, mannosan and galactosan were determined by placing a sample of PM₁₀ collected on a quartz filter (one half of the 47-mm filter) in a 4-ml dark glass vial. The following were added to the sample: 2 ml of pyridine (POCH) as a solvent and 50 μ l of a mixture of N,O-bis(trimethylsilyl)trifluoro-acetamide and trimethylchlorosilane (BSTFA: TMCS, 99: 1, Supelco) as a derivatizing agent. The vial was shaken for 1 min after closure and placed in an oven at 40 °C for 30 min. The extracts (1 ml) were analysed after filtration through a syringe filter (0.22 μ m) using a Shimadzu GC-2010 gas chromatograph coupled with a mass spectrometry detector equipped with an HP-5MS column (30 m, 0.25 mm, and 0.25 μ m). The limit of detection (LOD) was equal to 0.25 ng/m³ for LG, 0.11 ng/m³ for MN and 0.12 ng/

m³ for GA. The limit of quantification (LOQ) was equal to 0.76, 0.34 and 0.35 ng/m³ for LG, MN and GA, respectively. Method precision expressed as a relative standard deviation was equal to 19%, and recovery was 95%. Pure levoglucosan was purchased from Sigma-Aldrich, Germany. Mannosan and galactosan were obtained from LGC, Germany. The validation parameters are presented in Table 1.

The analysis of organic carbon (OC) was performed with the use of a thermal-optical organic and elemental carbon analyser, Sunset Laboratory Inc., using the EUSAAR_2 protocol, which was developed as a proposed standard method for European measurement stations under the European Supersites for Atmospheric Aerosol Research project (EUSAAR project, CEN/TR 16243, 2011). For each real sample batch/series, the blank samples were analysed to verify the presence of OC. Based on the results of the 1.0 cm² pieces from the 25 blank samples, the LOQ was calculated as 0.94 μ gC/cm² for OC. Repeatability was estimated from 25 independent measurements of the same sample under the same measuring conditions (slice of quartz filter with a glucose standard solution of concentration 4 μ gC/ μ L). Repeatability determined that the relative standard deviation for OC was 1.6%.

Results and discussion

The mass concentrations of the analysed chemical species, biomass burning tracers, PM₁₀ and OC for health resort Krynica are summarized in Tables 2 and 3 and Figs. 1, 2 and 3.

The average mass concentration of PM₁₀ was 28.4 μ g/m³ in the 2017/2018 season and 24.6 μ g/m³ in the 2018/2019 season. These results correspond to the results from Gothenburg and Umeå with a PM₁₀ concentration of 21 μ g/m³ (Ferm and Sjöberg 2015) and a regional background of approximately 20 μ g/m³, as determined by Forsberg et al. (2005) in Sweden, e.g., Trelleborg and Malmö. A similar annual average of 24.5 μ g/m³ was reported by Monteiro et al. (2018) in Porto. The results are higher than those of Riccione, Italy, with a concentration of 8 μ g/m³ (Vassura et al. 2014), but lower than those of Cracov and Brzezina, Poland, with a mean value of approximately 100 μ g/m³ (Samek et al. 2013).

Table 1 Validation parameters for levoglucosan, mannosan and galactosan

Biomass burning tracers	LOD ng/m ³	LOQ ng/m ³	Calibration lines	Regression coeff.
LG	0.25	0.76	$y = 87,429x - 44,445$	0.999
MN	0.11	0.34	$y = 76,601x - 52,178$	0.998
GA	0.12	0.35	$y = 189,952x - 81,840$	0.997

Table 2 Average, minimum and maximum concentrations of biomass burning tracers (ng/m³), PM₁₀ (μg/m³) and OC (μg/m³) in aerosols in the two winter campaigns

	2017/2018			2018/2019		
	Average (SD)	Min.	Max.	Average (SD)	Min.	Max.
LG	137 (178)	0.6	972	245 (250)	21.0	1520
MN	13.0 (19.5)	0.8	99.8	62.1 (39.9)	7.9	226
GA	4.7 (14.4)	0.2	80.9	31.1 (20.5)	0.5	95.2
PM ₁₀	28.4 (16.6)	6.71	106	24.6 (15.7)	4.98	77.9
OC	9.48 (5.93)	2.43	36.5	8.79 (6.72)	1.54	33.6

The average temperature during the winter season in 2017/2018 was -2.2 °C, and in the season 2018/2019, it was -1.4 °C.

The mean biomass burning tracer concentrations from Krynica in the two winter seasons of 2017/2018 and 2018/2019 were 155 and 338 ng/m³, respectively. The average concentrations of the three biomass burning tracers in Europe are very diverse, e.g., 379 ng/m³ in rural backgrounds in Seiffen, Germany (Linuma et al. 2009); 506 ng/m³ and 562 ng/m³ in urban backgrounds in Ghent, Belgium (Pashynska et al. 2002; Zdráhal et al. 2002); and 772 ng/m³ in winter in suburban sites in Elverum, Norway (Yttri et al. 2007). In Brno and Šlapanice, the average concentrations of tracers in winter in PM₁ were 273 and 646 ng/m³, respectively (Křůmal et al. 2015).

For PM₁₀ biomass burning, tracer concentrations in Krynica in 2017/2018 were 137 ng/m³ (from 0.6 to 972 ng/m³) for LG, 13.0 ng/m³ (from 0.8 to 99.8 ng/m³) for MN and 4.7 ng/m³ (from 0.2 to 80.9 ng/m³) for GA. In the 2018/2019 season in Krynica, the tracer concentrations were higher, reaching 250 ng/m³ (from 21.0 to 1520 ng/m³), 62.1 ng/m³ (from 7.9 to 226 ng/m³) and 31.1 ng/m³ (from 0.5 to 95.2 ng/m³) for LG, MN and GA, respectively. These results are comparable with biomass burning tracers determined in Ghent (urban), Belgium, where the concentration of LG was 477 ng/m³ (from 121 to 1133 ng/m³), that of MN was 65.9 ng/m³ (from 17.3 to 153 ng/m³) and that of GA reached 19.6 ng/m³ (from 4.4 to 44.2 ng/m³) (Zdráhal et al. 2002). In the work of Pashynska et al. (2002), the concentrations of

Table 3 Percentages of biomass burning tracers in the total sum of compounds in atmospheric aerosols, and the LG/MN and LG/(MN+GA) ratios in the two winter seasons of 2017/2018 and 2018/2019

Biomass burning tracers	2017/2018	2018/2019
LG	88.6	72.4
MN	8.4	18.4
GA	3.0	9.2
LG/MN	10.5	3.9
LG/(MN+GA)	7.7	2.6

Fig. 1 The average concentrations of biomass burning tracers in 2017/2018 (black with dots) and 2018/2019 (chequered pattern)

biomass burning tracers were 420 ng/m³ (from 96 to 1900 ng/m³) for levoglucosan, 61 ng/m³ (from 10 to 290 ng/m³) for mannosan and 25 ng/m³ (from 4.9 to 115 ng/m³) for galactosan. Higher results were observed in Porto, with a winter mean of LG equal to 774 ng/m³ (Monteiro et al. 2018) and Elverum (suburban) winter means of 605, 167 and 4.0 ng/m³ for LG, MN and GA, respectively (Yttri et al. 2007). Biomass consists of several main components, such as cellulose with 40–50% dry weight of wood; hemicellulose with 20–30% dry weight of wood; and lignins, combined with tannins, terpenes and other substances. The relative amount of tracers is determined by the type of biomass (softwood, hardwood, and grass) (Simoneit 2002; Janoszka and Czaplicka 2019) and the proportions of the components (Pereira et al. 2017).

The differences between the results in the 2017/2018 and 2018/2019 seasons may have been caused by various temperatures. The correlation between the levoglucosan concentration and temperature for the 2017/2018 and 2018/2019 seasons is presented in Figs. 1 and 2, respectively. In the 2017/2018 season, the mean temperature was -2.2 °C, including 8 days from 25.02.2018 to 04.03.2018 where temperatures were under -10 °C. However, the maximum peaks in the biomass burning tracers may result from local conditions such as wind direction and humidity together with temperature. In the 2018/2019 season, the mean temperature was -1.4 °C, and there were only 5 days with temperatures under -10 °C. From 19.01.2019 to 24.01.2019, the temperature was approximately -10 °C, which correlates with the peak biomass burning tracers. This difference could be caused by people who used coal and other types of biomass, such as straw or miscanthus pellets, instead of wood in domestic heating more in 2017/2018 than in 2018/2019.

Table 3 shows the percentage of individual tracers in the total sum of determined compounds based on average values. Among the determined biomass burning tracers, LG is the

Fig. 2 Variations in average levoglucosan concentrations and temperatures during the winter season of 2017/2018

most dominant. The levoglucosan percentage in the total biomass burning tracer sum was 88.6% in the 2017/2018 season and 72.4% in the 2018/2019 season. The mannosan percentages were 8.4% and 18.4% in the 2017/2018 and 2018/2019 seasons, respectively. The least abundant was galactosan, with percentages of 3.0% in the 2017/2018 season and 9.2% in 2018/2019. Zdráhal et al. (2002) observed a similar correlation with an LG percentage of 84.9%, MN of 11.8% and GA of 3.3%. The same relationship was observed for PM_{10} in Brno and Šlapanice in the Czech Republic, with 80% LG, 13% MN and 7% of GA when breaking down total amount of tracers (Křůmal et al. 2015).

To distinguish smoke emissions from different fuel types, relative abundances may be used: levoglucosan to mannosan (LG/MN) and levoglucosan to the sum of mannosan and galactosan (LG/(MN+GA)). In Krynica Zdrój, the mean LG/MN ratio for the 2017/2018 season was 10.5, and that for the 2018/2019 season 2018/2019 was 3.9. The mean LG/(MN+GA) values were 7.7 and 2.6 for the 2017/2018 and 2018/2019 seasons, respectively. The results may indicate that in

the 2017/2018 season in Krynica, a co-combustion of hardwood and straw or miscanthus pellets with coal occurred, and in the 2018/2019 season, softwood burning was dominant. A similar result for the LG/MN ratio in the 2018/2019 season was reported by Cordell et al. (2016) for Leicester; GB ranged from 2.6 to 4.6, and the LG/(MN+GA) ratio ranged from 2.1 to 3.0, which is typical for softwood. For Lille, France, the LG/MN ratio varies from 2.8 to 6.7, and the LG/(MN+GA) ratio varies from 2.0 to 4.3, possibly indicating a higher hardwood contribution. However, the results of the LG/MN ratio from standard reference material on urban aerosols in Washington, USA and Prague, Czech Republic were equal to 9.4 and 4.9, respectively, suggesting that of the mixture of hardwood and softwood input into the Washington samples and Prague samples, softwood was predominant (Louchouart et al. 2009). Caseiro and Oliveira (2012) reported the winter average LG/MN ratio in Birmingham, GB, with an urban background of 7 for fine fraction $PM_{2.5}$ and 18 for core fraction $PM_{2.5-10}$. The ratio in Okinawa varies from 12.3 to 13.9 in winter, suggesting hardwood, softwood and

Fig. 3 Variations in average levoglucosan concentration and temperature during the winter season of 2018/2019

hardwood mixtures or softwood and plant straw mixture burning (Zhu et al. 2015). In the 2017/2018 season in Krynica, the LG/MN ratio was higher compared to the data in the literature, which may be the result of different biomass burnings other than softwood and hardwood, such as straw or miscanthus pellets with coal. Because the atmospheric aerosol samples contain a spectrum of particulate matter, the observed biomass burning ratios must be considered carefully.

The amount of organic carbon derived from the burning of biomass can be calculated from Equation (1), which was proposed by Puxbaum et al. (2007) and Gelencsér et al. (2007), where an emission factor of 7.35 was based on the OC/LG mass emission ratio:

$$\text{Biomass smoke OC} = \text{levoglucosan} \times 7.35 \quad (1)$$

The data are presented in Table 4.

In the winter season of 2017/2018, the OC equalled 9480 ng/m³, and in the season of 2018/2019, it was 8790 ng/m³. These results are comparable to those obtained by Zhang et al. (2010) from Guangzhou in China, where the OC concentration was 7930 ng/m³. In PM₁, the OC contents in Brno and Šlapanice in the winter season of 2009 were 4.73 and 6.00 µg/m³, respectively, and in winter 2010, they were 9.97 and 8.74 µg/m³, respectively (Křůmal et al. 2015). The biomass smoke OC values calculated by Equation (1) were equal to 1010 ng/m³ and 1800 ng/m³ in the seasons of 2017/2018 and 2018/2019, respectively. The results are different from those reported by Puxbaum et al. (2007) in winter at several sites, such as 48 ng/m³ in Azores; 134 ng/m³ in Puy de Dôme, France; 7739 ng/m³ in Aveiro, Portugal; and 4800 ng/m³ in K-Puszt, Hungary. However, the results from Krynica are comparable with those presented by Caseiro and Oliveira (2012) in Oporto in the winter season, with a roadside value of 1.5 µg/m³ and an urban background value of 1.1 µg/m³. The biomass smoke OC according to Equation (1) was 10.6% of organic carbon measured in the 2017/2018 season and 20.5% of that measured in the 2018/2019 season.

The LG concentration was also used to calculate the contribution of biomass burning to OC by Equation (2), as proposed by Sang et al. (2011):

$$\text{BB to OC} = \frac{\text{Levoglucosan} - 17.5}{\frac{1000 \cdot \text{OC}}{0.082}} \cdot 100\% \quad (2)$$

The contributions of biomass smoke to OC in Krynica, according to Equation (2), in the winters of 2017/2018 and 2018/2019 were 14.4% and 28.4%, respectively. These results are compatible with the data obtained by Sang et al. (2011) from Hot Tsiu, China, with contributions in the range of 6.5 to 11%, and Zhang et al. (2008) from Beijing, with contributions ranging from 14 to 32% in PM₁₀ aerosols. However, the results from Krynica are lower than those reported by Caseiro et al. (2009), whereas those from Vienna range from 31.3 to 39.5%, those from Graz range from 38.4 to 58.9%, and those from Salzburg range from 34.4 to 70.2%. For Brno and Šlapanice in the winter of 2009, the relative contributions of biomass smoke to OC were 34.2 and 51.7%, respectively, and 24.1 and 20.2% in the winter of 2010, respectively (Křůmal et al. 2015).

Conclusions

The obtained results showed the following:

1. The average biomass burning tracer concentration in the winter season of 2017/2018 was lower than that in the winter season of 2018/2019.
2. Levoglucosan, the most abundant tracer, accounted for 88.6% of the total determined biomass burning tracers in the 2017/2018 season, while in the 2018/2019 season, it accounted for 72.4%.
3. Galactosan, the least abundant tracer, comprised 3.0% of the sum of tracers in 2017/2018 and 9.2% in 2018/2019.
4. The LG/MN ratios were 10.5 and 3.9 for 2017/2018 and 2018/2019, respectively, while the LG/(MN+GA) ratios were 7.7 and 2.6 for 2017/2018 and 2018/2019, respectively. In the 2017/2018 season in Krynica, the co-firing of hardwood and straw or the co-firing of miscanthus pellets with coal was dominant, and in the 2018/2019 season, softwood burning was dominant.
5. The biomass smoke OC calculated according to Puxbaum et al. (2007) and Gelencsér et al. (2007) was equal to 1010 ng/m³ in the 2017/2018 season and 1800 ng/m³ in the 2018/2019 season, which is 10.6% of the organic carbon measured in the 2017/2018 season and 20.5% of the organic carbon measured in the 2018/2019 season. However, the results of the contribution of biomass smoke to OC according to Sang et al. (2011) were 14.4% and 28.4% in the 2017/2018 and 2018/2019 seasons, respectively.

Table 4 Concentration of biomass smoke OC and the OC levels in two winter seasons (2017/2018 and 2018/2019), ng/m³

ng/m ³	2017/2018		2018/2019	
	Biomass smoke OC	OC	Biomass smoke OC	OC
Average	1010	9480	1800	8790
Maximum	7150	36,500	11,200	33,800
Minimum	4.0	2430	154	1540

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Author' contributions Katarzyna Janoszka performed the experiments and co-wrote the paper; Marianna Czaplicka analysed the data and co-wrote the paper; Krzysztof Klejnowski collected the samples.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interest.

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