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Correlation Between Biomass Burning Tracers in Urban and Rural Particles in Silesia—Case Study

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Abstract The major biomass burning tracers are thermal degradation products from the biopolymer cellulose, namely the didehydromonosaccharide derivatives levoglucosan, galactosan, and mannosan and the resin acid derivative dehydroabietic acid, with a minor contribution from β -sitosterol. Levoglucosan, galactosan, and mannosan were measured at two sites in Silesia, a rural (Rokitno) and industry region (Zabrze), during the winter of 2017/2018. The results showed that mean concentrations of the total tracers determined were 737 ng/m³ for Zabrze and 465 ng/ m³ for Rokitno. Levoglucosan was the most abundant tracer; it was 83.2% of the determined tracers in Zabrze and 78.1% in Rokitno. The relative proportions of levoglucosan to mannosan have been used for source reconstruction of combustion-derived byproducts in atmospheric aerosols. The levoglucosan to mannosan ratio for Zabrze was 8.9 and for Rokitno 5.3; the levoglucosan to sum of mannosan and galactosan ratio was 6.2 and 3.8 for Zabrze and Rokitno, respectively. The correlation between tracers is high (0.73 to 0.97) and shows linearity. In order to compare the fuel type (by the coefficient of divergence (CD)) between different sites, the results from a previous work in health resort Krynica were used. The CD

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between Krynica and Rokitno as well as Krynica and Zabrze was equal to 0.633 and 0.712, respectively. The CD between Rokitno and Zabrze was equal to 0.175. Despite the biomass burning tracer measurements are mostly local, they have a huge impact on air pollution and climate changes.

Abbreviations

LG Levoglucosan MN Mannosan GA Galactosan

1 Introduction

During the burning of biomass, which consist mainly of cellulose, at a temperature above 300 °C, levoglucosan (LG, 1,6-anhydro- β -glucopyranose) is created, as well as the levoglucosan isomers mannosan (MN) and galactosan (GA) which are products of hemicellulose burning. It is well known that levoglucosan, mannosan, and galactosan are biomass burning tracers. Those tracers are believed to be stable in the environment and resistant to photochemical oxidation (Jordan et al., 2006; Piot et al., 2012; Schkolnik & Rudich, 2006). However, it is shown that tracers are not inert and their lifetime depends on the OH radical concentration. At chamber experiments simulating typical summertime OH concentration (4.9×10^{10}) molecules cm^{-3} s), the levoglucosan was stable for 0.7 to 2.2 days (Hennigan et al., 2010, 2011). Because our study took place during the winter and the local biomass burning is a source of levoglucosan, it is assumed that LG is sufficiently stable to be used as a tracer. Sugar anhydrides are in the particular phase in the atmosphere because of their low vapor pressure and they can, for example, affect the earth radiation budget and act as cloud condensation nuclei (Janoszka & Czaplicka, 2019; Poor, 2002; Schkolnik & Rudich, 2006; Simoneit et al., 1999). Biomass burning emission can interfere with carbon bio- and geochemical cycling, the chemistry of the tropospheric, and furthermore, when deposited on glacier and ice sheets, reduce snow albedo and accelerate melting (Bhattarai et al., 2019; Xu & You, 2021; Galindo et al., 2021). Puxbaum et al. (2007) indicate that in the winter, the contribution of biomass combustion is higher than in the summer, because of lower temperatures which cause slower air mass exchange in the atmosphere and higher smoke emission. The biomass burning tracers may be used for determination of the products burned, for example, in domestic furnaces. Because the biomass smoke may have different sources, like wild fires, agricultural burns, and domestic heating as well as long-range transport, the concentrations of levoglucosan observed are characterized by high variability. For example, the average LG concentration in the Po Valley (Northern Italy) was 176 ng/m³ and 19.3 and 12.8 ng/m³ in MN and GA, respectively (Saarikoski et al., 2012). Mkoma et al. (2013) reported the values of 209 and 308 ng/m^3 for levoglucosan in the wet and dry season, respectively and 20 ng/m³ for mannosan in the wet and 30 ng/m^3 in the dry season.

The ratios of levoglucosan to mannosan and levoglucosan to the sum of mannosan and galactosan may be used to distinguish between different types of fuel burned when analyzing atmospheric aerosols. However, caution must be taken in the interpretation of environmental samples because of smoke mixing and high values of LG/MN ratios for grasses and different hardwood (Fabbri et al., 2009; Schmidl et al., 2008). For example, the LG/MN ratio in the case of combustion of American beech was 17 and for White spruce 4 (Piot et al., 2012).

The authors indicate a correlation between air pollutants and the type of emission source. One of

these dependences is the coefficient of divergence (CD), a self-normalizing parameter for comparison of chosen species concentration. Tan et al. (2014) used the CO, NO_x , and SO_x concentration to compare sampling sites in term of pollutants. Jiang et al. (2018) and Kong et al. (2011) used $PM_{2.5}$ and PM_{10} concentration.

Considering the need to monitor air quality for the presence of pollutants and their impact on climate change, it is important to have knowledge of the biomass burning tracer concentrations in the air. It is known that due to their physicochemical properties, they affect aerosol grain sizes and cloud formation processes (Dixon & Baltzell, 2006; Gao et al., 2003; Graham et al., 2002; Oros & Simoneit, 2001; Schkolnik & Rudich, 2006). In Poland, there is a lack of research on biomass burning tracer concentrations in atmospheric aerosols. The knowledge about the local biomass burning, like in densely populated area of Upper Silesia Agglomeration, may have an impact on the larger area of the country. Moreover, there are more and more regulations in Poland enforcing higher requirements for heat sources. The Upper Silesian Agglomeration and the whole Silesian Voivodeship are regulated by an anti-smog law. It obligates the inhabitants to replace the heat sources with ecological ones. The schedule assumes replacement of all out-of-class boilers or those older than 10 years by the end of 2021. The modern boilers with feeders (eco-pea hard coal, pellet) at least fifth eco-design class, gas boilers, heat pumps, or electric boilers are allowed (Resolution of the Sejmik no. V/36/1/2017). The most promoted solutions are heat pumps and pellet boilers. Similar resolutions are in force in most of the country. Further monitoring will assess the impact of the ordinances on atmospheric pollution.

Therefore, within the framework of this article, the objectives were to (i) determine the level of biomass burning tracer concentrations in air samples for two sites, industry region and rural area, during the winter season 2017/2018, (ii) investigate the relationship between biomass burning tracers, and (iii) estimate the coefficient of divergence.

2 Experimental and Methodology

Atmospheric aerosol samples were collected on quartz filters at measurement stations situated in

southern Poland, in Zabrze (industry region, urban background, located in the center of Upper Silesia Agglomeration, $\varphi = 50^{\circ}18'53''$ N, $\lambda = 18^{\circ}46'17''$ E, h=254 m above sea level, a.s.l.,) and Rokitno (rural area, rural background, open area near small village situated close to the zinc and lead ores, $\varphi = 50^{\circ}26'09''$ N, $\lambda = 19^{\circ}25'29''$ E, h = 344 m a.s.l.) from December 2017 to March 2018 (Fig. 1). In order to compare the results between different sites, the measurements from a previous work in health resort Krynica (rural background, slope of the mountain valley, $\varphi = 49^{\circ}24'28''$ N, $\lambda = 20^{\circ}57'39''$ E, h = 582 m a.s.l.) were used. Samples were collected in accordance with the methodology previously described (Janoszka et al., 2020; Klejnowski et al., 2017) with the use of a low flow sampler with a PM₁₀ separating head (Atmoservice PNS3D15/LVS3d) and a stabilized flow of 2.3 m³/h at a 24-h cycle on 47-mm Whatman QMA quartz filters.

PN-EN 12,341:2014 (ambient air) was used for sample collection and gravimetric. A standard gravimetric measurement method for the determination of the PM_{10} mass concentration of suspended particulate matter of every sample filter was used. Conditioning, weighing (Mettler Toledo microbalance with resolution 2 µg), and storage of exposed and non-exposed filters were done at a weighing room with control conditions (dryer and humidifier) with temperature 20 ± 1 °C and humidity $45 \pm 5\%$ RH.

In order to determine the concentration of biomass burning tracers (levoglucosan, mannosan, and galactosan), a sample of atmospheric aerosol collected on a quartz filter was extracted and derivatized at the same time according to the methodology previously



Fig. 1 Sampling site position

described (Janoszka et al., 2020). Briefly, the sample was placed in a 4-ml dark glass vial; 2 ml of pyridine (POCH) and 50 µl of a derivatizing agent and a mixture of N,O-bis(trimethylsilyl)trifluoroacetamide and trimethylchlorosilane (BSTFA:TMCS, 99:1, Supelco) were added. The vial was closed, shaken for 1 min, and placed in an oven at 40 °C for 30 min. After reaction, 1 ml of the extract was filtered through a syringe filter (0.22 µm) and analyzed using a Shimadzu GC-2010 gas chromatograph coupled with a mass spectrometry detector equipped with an HP-5MS column (30 m, 0.25 mm, 0.25 µm). The limit of detection (LOD) was equal to 3.33 ng/ml for LG, 1.50 ng/ ml for MN, and 1.53 ng/ml for GA. The limit of quantification (LOQ) was equal to 9.99, 4.51, and 4.58 ng/ ml for LG, MN, and GA, respectively. The precision expressed as a relative standard deviation was equal to 19%.

3 Result and Discussion

The mass concentration of PM₁₀ and biomass burning tracer concentrations (levoglucosan, mannosan, and galactosan) are statistically summarized in Tables 1 and 2 and Figs. 2 and 3.

Table 1 Concentration of PM₁₀ by months and for the entire sampling period at both sites

Month	Rokitno , µg/m ³	Zabrze , μg/m ³
December	20.4	58.8
January	23.2	55.1
February	30.5	69.9
March	25.2	75.3
Average	24.8	64.8

The average mass concentration of PM_{10} (Table 1) was 24.83 μ g/m³ at the rural area Rokitno which is higher than at a rural background site in Vindeln, Sweden with about 5 μ g/m³ of PM₁₀ (Hedberg et al., 2006). The average PM_{10} concentration value at the industrial area Zabrze was $64.78 \,\mu\text{g/m}^3$. This is higher than the winter average PM_{10} concentration value of 43 µg/m³ for Launceston, Australia (Jordan et al., 2006), and 56.83 μ g/m³ for Lhasa, Tibet (Yin et al., 2019), but more than 2 times lower than in urban Beijing with a mean PM_{10} concentration of 169 µg/m³ (Zhang et al., 2008).

The level of average PM₁₀ concentration in most months in Rokitno was from 20.41 to 30.53 μ g/m³. In Zabrze, the mean concentration for each month is higher, from 55.13 to 75.29 μ g/m³.

Biomass burning tracers are frequently detected in terms of atmospheric pollution. For example, the average concentration of the tracers at an urban background site in Helsinki during winter 2008-2009 was 90 ng/m³ (Saarnio et al., 2010) and 630 ng/m³ for Beijing and Shijiazhuang, China, from 1 to 12 November 2014 (Zhang et al., 2017).

The average tracer concentrations during the measurement campaign were 465 ng/m³ for Rokitno and 736 ng/m³ for Zabrze. In the case of levoglucosan, the average campaign concentration was 366 and 606 ng/m³ for Rokitno and Zabrze, respectively. The mannosan average campaign concentration was 66.6 ng/m³ for Rokitno and 96.9 ng/m³ for Zabrze. For galactosan, the average concentration was 33.0 and 33.5 ng/m³ for Rokitno and Zabrze, respectively (Fig. 4).

In December 2017 and January and March 2018, the average concentration of biomass burning tracers in atmospheric aerosol was higher at the sampling point in Zabrze being 902, 646, and 449 ng/

ng/m^3 GA, ng/m^3
30.1
7.5
80.0
14.2
55.2
32.9
32.4
13.6
r



Fig. 2 Variations of PM₁₀ concentrations at the sampling sites

 m^3 , respectively. In February, the higher total tracer concentration in Rokitno was equal to 1005 ng/m^3 (Table 2).

Zabrze is an industrial city with 170,000 inhabitants surrounded by single-family houses with individual furnaces. Therefore, the higher biomass burning tracer concentrations are caused by higher emissions from biomass combustion and domestic heating. Rokitno is a village in a rural area with about 900 inhabitants and characterized with lower tracer concentration.

The results for single tracers can be very diverse. For example, in Malaysia the concentration of levoglucosan varies from 10 ng/m³ at Taman Negra up to $34,000 \text{ ng/m}^3$ at the University of Malaya (Petaling Jaya) at night (Abas et al., 2004).

In the case of the rural area Rokitno, the mean concentration varies from 192 ng/m³ in March to 1005 ng/m³ in February (Fig. 5). The highest average

concentration of biomass burning tracers for the industry region in Zabrze was measured in February and was equal to 946 ng/m³, while the lowest concentration of 449 ng/m³ was determined in March (Fig. 6).

Of the determined biomass burning tracers, the dominant compound was levoglucosan. The mean LG concentration determined during the measurement campaign varies from 142 to 806 ng/m³ for Rokitno and from 403 to 846 ng/m³ for Zabrze. In the case of mannosan, the mean values for Rokitno vary from 27.9 to 119 ng/m³ and for Zabrze vary from 31.8 to 177 ng/m³. The galactosan is the least abundant of the determined tracers and it varies from 7.5 to 80.0 ng/m³ and from 13.6 to 55.2 ng/m³ for Rokitno and Zabrze, respectively. These results correspond to the previous work by Hedberg et al. (2006) where the mean LG concentration at urban Lycksele, Sweden, equaled to 896.6 ng/m³,



Fig. 3 Variations of the sum of the biomass burning tracer concentrations at the two sampling sites



with the minimum and maximum value of 16.5 and 2335.4 ng/m³, respectively. The result in the Po Valley, Italy, for LG was 176 ng/m³, for MN 19.3 ng/m³, and for GA 12.8 ng/m³ (Saarikoski et al., 2012). In the case of Morogoro, Tanzania, East Africa LG and MN mean concentrations in the wet season were equal to 209 and 20 ng/m³, respectively, and in

the dry season were 308 and 30 ng/m^3 , respectively (Mkoma et al., 2013).

In the case of Rokitno, the total determined tracer concentrations vary from 1.2 ng/m^3 determined on 12.03.2018 to 1710 ng/m^3 on 10.02.2018. For Zabrze, the lowest total biomass burning tracer concentration was 33.7 ng/m^3 determined on 18.03.2018, and the

Fig. 4 The average concen-

tration for the sum of the biomass burning tracers at each site by month





Fig. 6 Average monthly concentration of biomass burning tracers in atmospheric aerosol in Zabrze

highest value of total concentration was 3165 ng/m^3 on 17.12.2017.

In the case of the rural area Rokitno, the mean percentage of individual tracers in the total sum was 78.1%, 15.5%, and 6.4% for LG, MN, and GA, respectively. In the industry region (Zabrze), the LG was on average 82.8%, MN was 12.7%, and GA was 4.4% (Table 3).

Galactosan is the least occurring tracer and levoglucosan is the dominant biomass burning tracer in each measurement point. In the case of Zabrze, the LG percentage is above 80%. The galactosan percentage is less than 10%. The mannosan percentage of the sum varies from 12.7 to 15.5%. A similar relationship was observed by Saarikoski et al. (2012) in the Po Valley, Italy, with LG percentage of 84.4%, MN of 9.56%, and GA of 6.0%.

The relative proportion of levoglucosan to mannosan (LG/MN) and levoglucosan to the sum of mannosan and galactosan (LG/(MN+GA)) may be utilized to distinguish the smoke emission for different fuel types. The results obtained by Rodrigues et al.

	Month	LG %	MN %	GA %
Rokitno	December	74.2	18.9	6.8
	January	84.2	12.5	3.3
	February	80.2	11.9	8.0
	March	73.8	18.8	7.4
Zabrze	December	74.4	19.5	6.1
	January	77.7	17.2	5.1
	February	89.4	7.2	3.4
	March	89.9	7.1	3.0

 Table 3
 Percentages of biomass burning tracers in the total sum of compounds by month at each site

Table 4 The LG/MN and LG/(MN+GA) ratios as well as the average temperature by month for each site

	Month	LG/MN	LG/ (MN+GA)	Tem- perature, °C
Rokitno	December	3.9	2.9	0.8
	January	6.7	5.3	0.2
	February	6.8	4.0	-4.4
	March	3.9	2.8	-0.4
Zabrze	December	3.9	2.9	2.5
	January	4.5	3.5	3.0
	February	12.5	8.4	-2.9
	March	12.7	8.9	2.1

0.40

0.35 0.30

0.25 0.20 0.15 0.10 0.05 0.00

0.00

0.20

0.40

0.60

0.80

MN, ng/m³

Fig. 7 The levoglucosan concentration plotted against the mannosan concentration at Rokitno



1.00

LG, ng/m³

1.20

1.40

1.60

1.80

(2020) with a LG/MN ratio equal to 5.68 and a LG/ (MN+GA) ratio of 2.85 indicated softwood burning in Mt. Wellington, (Hobart, Tasmania, Australia); the results of 10.19 and 4.25 for LG/MN and LG/ (MN+GA), respectively, indicate higher hardwood contribution in Gingin (Western Australia). Mkoma et al. (2013) reported LG/MN ratio values equal to about 10 to 13 which suggested hardwood and crop residue burning.

The mean value of the LG/MN ratio for the rural area Rokitno was 5.3 and for the industry region Zabrze was equal to 8.4. The mean value of the LG/(MN+GA) ratio was equal to 3.8 and 5.9 for Rokitno and Zabrze, respectively (Table 4). The results indicate that in Rokitno softwood burning was dominant and in Zabrze the co-combustion of softwood with hardwood.

The correlation coefficient of the mannosan and levoglucosan ratio was 0.79 for Rokitno and 0.60 for Zabrze. The correlation coefficient of the GA and LG ratio was 0.81 for Rokitno and 0.60 for Zabrze. In the case of the galactosan and mannosan ratio, the result was 0.78 and 0.96 for Rokitno and Zabrze, respectively. The correlation coefficient for the tracer ratios shows high correlation with linear regression (Figs. 7, 8, 9, 10, 11, and 12).

These results correspond to that obtained by Sullivan et al. (2014) who observed GA/LG correlation coefficient from 0.80 to 0.88 and MN/LG from 0.97 to 0.99. Levoglucosan and mannosan strongly correlated in measurements from Tanzania and the Po Fig. 8 The levoglucosan

against the galactosan con-

concentration plotted

centration at Rokitno



Fig. 9 The mannosan concentration plotted against the galactosan concentration at Rokitno



Valley, Italy, with R^2 values ranging from 0.91 to 0.98 (Mkoma et al., 2013) and 0.98 to 0.99, respectively (Saarikoski et al., 2012).

The tracer sum to PM_{10} concentrations ratio was 1.77×10^{-2} for Rokitno and 1.21×10^{-2} for Zabrze. In the case of the LG/PM₁₀ ratio, the result was 1.41×10^{-2} and 1.00×10^{-2} for Rokitno and Zabrze, respectively. In the case of the MN/PM₁₀ ratio, the result was 2.63×10^{-3} and 1.74×10^{-3} for Rokitno and Zabrze, respectively. The GA/PM₁₀ concentration ratio was 1.27×10^{-3} for Rokitno and 5.83×10^{-4} for Zabrze (Table 5). Mkoma et al. (2013) reported a LG/PM ratio (%) ranging from 0.20 to 0.78. The PM from wood burning mean estimated concentration can be calculated according to Puxbaum et al. (2007) and Fuller et al. (2014). A factor of 7.35 is suggested to convert mean LG concentration to organic carbon and additional factor of 1.4 for biomass organic matter. Caseiro and Oliveira (2012) show the similar relationship with biomass smoke PM was equal to mean LG concentration multiplied by the factor of 10.7. The PM estimated concentration from wood burning for Rokitno was 3.76 μ g/m³ and for Zabrze was 6.24 μ g/m³. Those results are higher than for Kensington, England, which was 1.8 μ g/m³ (Fullet et al., 2014) as well as for roadside sampling Fig. 10 The levoglucosan

against the mannosan con-

concentration plotted

centration at Zabrze



Fig. 11 The levoglucosan concentration plotted against the galactosan concentration at Zabrze



point in Copenhagen and Oporto of 1.12 and 1.8 µg/m³, respectively (Caseiro and Oliveira, 2012).

The coefficient of divergence (CD) allows for quantifying the similarities between two sampling points (emission sources) (Jiang et al., 2018; Kong et al., 2011; Li et al., 2020; Tan et al., 2014).

$$CD_{jk} = \sqrt{\frac{1}{\rho} \sum_{i=1}^{\rho} \left(\frac{X_{ij} - X_{ik}}{X_{ij} + X_{ik}}\right)^2}$$

where *j* and *k* are sampling points, X_{ij} and X_{ik} are average concentration of biomass burning tracer for

a given sampling site, and ρ is three because there are 3 biomass burning tracers: levoglucosan, mannosan, and galactosan. If the coefficient of divergence approaches zero, the sources of emission are similar and if the CD goes to one, the sources at the sampling sites are relevantly different. It is assumed that when the CD is higher than 0.31, there is a considerable difference between the profiles at the two sampling sites. Li et al. (2020) used the phthalate concentration to determine the CD parameter and Tan et al. (2014) used CO, NO_x, and SO₂ concentrations. Fig. 12 The mannosan

against the galactosan con-

concentration plotted

centration at Zabrze



Table 5 Average concentration of total tracers sum to ambient aerosol (TC/PM₁₀) ratio, levoglucosan concentration to ambient aerosol (LG/PM₁₀) ratio, mannosan concentration to ambi-

ent aerosol (MN/PM₁₀) ratio, and galactosan concentration to ambient aerosol (GA/PM₁₀) at both sites for measurement campaign, standard deviation in bracket

Ratio	Rokitno	Zabrze
Tracers sum/PM ₁₀ (SD)	$1.8 \times 10^{-2} (1.6 \times 10^{-2})$	$1.2110^{-2} (6.9 \times 10^{-3})$
LG/PM ₁₀ (SD)	$1.4 \times 10^{-2} (1.3 \times 10^{-2})$	$1.0 \times 10^{-2} (5.8 \times 10^{-3})$
MN/PM ₁₀ (SD)	$2.6 \times 10^{-3} (2.3 \times 10^{-3})$	$1.7 \times 10^{-3} (1.3 \times 10^{-3})$
GA/PM ₁₀ (SD)	$1.3 \times 10^{-3} (1.4 \times 10^{-3})$	$5.8 \times 10^{-4} (4.1 \times 10^{-4})$

Taking into consideration the results described elsewhere (Janoszka et al., 2020) from Krynica (the local environment of health resort) in the same time period as this study with concentration of the LG, MN, and GA equals to 139, 14.2, and 4.5, respectively, the CD between Krynica and Rokitno as well as Krynica and Zabrze was equal to 0.633 and 0.714, respectively. The CD between Rokitno and Zabrze was equal to 0.178. From this, it follows that assuming 0.3 as the limit value, Zabrze and Rokitno have a similar source of pollution. However, Krynica has a different emission source than Zabrze and Rokitno, as the coefficient of divergence is greater than 0.3. Moreover, the PM concentration from wood burning for Krynica according to the levoglucosan method (biomass smoke PM = LG \times 7.35 \times 1.4) which was 1.43 μ g/m³ and the restrictions introduced by the administrative authorities on the domestic heating (heating of residential buildings with gas) show different emission sources than for Rokitno and Zabrze.

4 Conclusions

In Zabrze, the PM_{10} concentration is about two times higher than in Rokitno. In December, January, and March, the average biomass burning concentration was higher in the industrial region Zabrze and in February the higher biomass tracer concentration was determined in the rural area Rokitno. Among the determined biomass burning tracers, levoglucosan is the most abundant compound; its mean percentage in total sum of tracers varies from 78.1 to 88.8%. While galactosan was the least frequent, its mean percentage in total tracers sum varies from 4.4 to 6.4%. The obtained results for the mean LG/MN and LG/ (MN+GA) concentration ratios indicate that in the case of industrial region the monosaccharides are most likely emitted from softwoods burning and in the rural area there was co-combustion of softwood with hardwood. Moreover, taking into consideration the average monthly temperatures and respective average monthly LG/MN and LG/(MN+GA) ratios, the material used for domestic heating is determined by temperature and localization. In colder months, the co-firing with hardwood dominated and in months with higher temperature the use of softwood was dominant. The agreement between biomass burning tracers is strong, with correlation coefficient from 0.60 to 0.96. The biomass burning tracer concentration sum to PM₁₀ concentration ratio was 1.77×10^{-2} for Rokitno and 1.21×10^{-2} for Zabrze. Considering the CD parameter, Zabrze and Rokitno have a similar source of pollution and Krynica has a different emission source.

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Data Availability As the results of this research were done under an internal grant, they do not require to be available. The datasets generated/analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of Interest The authors declare no competing interests.

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