



Sources of the PM₁₀ aerosol in Flanders, Belgium, and re-assessment of the contribution from wood burning



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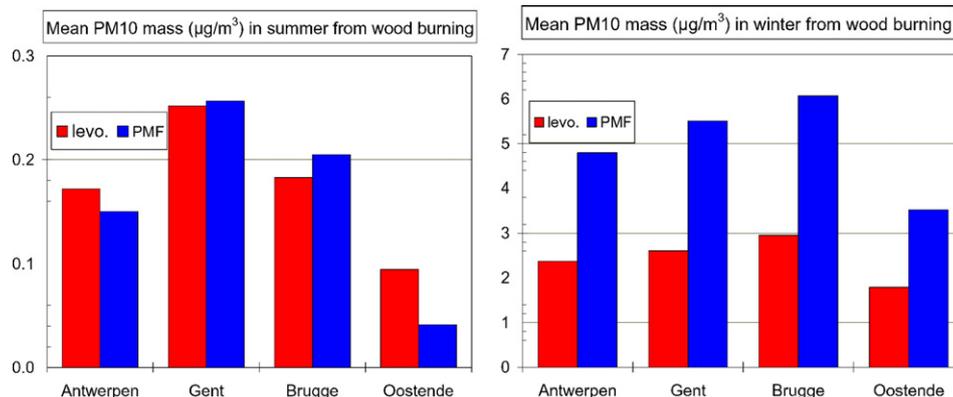
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HIGHLIGHTS

- A one-year study with 4 urban background sites and a total of 372 daily PM₁₀ samples
- First application of Positive Matrix Factorisation on aerosol data sets from Belgium
- Comparison of two approaches for estimating the contribution from wood burning
- Wood burning appears to be a notable source of As, Cd, and Pb
- The impact from wood burning in Flanders is larger than previously thought

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 18 February 2016

Received in revised form 5 April 2016

Accepted 11 April 2016

Available online xxxx

Editor: D. Barcelo

Keywords:

PM₁₀ mass

Organic carbon

Source apportionment

PMF

Wood burning

Levoglucosan

ABSTRACT

From 30 June 2011 to 2 July 2012 PM₁₀ aerosol samples were simultaneously taken every 4th day at four urban background sites in Flanders, Belgium. The sites were in Antwerpen, Gent, Brugge, and Oostende. The PM₁₀ mass concentration was determined by weighing; organic and elemental carbon (OC and EC) were measured by thermal-optical analysis, the wood burning tracers levoglucosan, mannosan and galactosan were determined by gas chromatography/mass spectrometry, 8 water-soluble ions were measured by ion chromatography, and 15 elements were determined by a combination of inductively coupled plasma atomic emission spectrometry and mass spectrometry. The multi-species dataset was subjected to receptor modeling by PMF. The 10 retained factors (with their overall average percentage contributions to the experimental PM₁₀ mass) were wood burning (9.5%), secondary nitrate (24%), secondary sulfate (12.6%), sea salt (10.0%), aged sea salt (19.2%), crustal matter (9.7%), non-ferrous metals (1.81%), traffic (10.3%), non-exhaust traffic (0.52%), and heavy oil burning (3.0%). The average contributions of wood smoke for the four sites were quite substantial in winter and ranged from 12.5 to 20% for the PM₁₀ mass and from 47 to 64% for PM₁₀ OC. Wood burning appeared to be also a notable source of As, Cd, and Pb. The contribution from wood burning to the PM₁₀ mass and OC was also assessed by making use of levoglucosan as single marker compound and the conversion factors of Schmidl et al. (2008), as done in our previous study on wood burning in Flanders (Maenhaut et al., 2012). However, the apportionments were much lower than those deduced from PMF. It seems that the conversion factors of Schmidl et al. (2008) may not be applicable to wood

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burning in Flanders. From scatter plots of the PMF-derived wood smoke OC and PM versus levoglucosan, we arrived at conversion factors of 9.7 and 22.6, respectively.

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1. Introduction

Source identification and apportionment are important topics in atmospheric aerosol research in Europe (e.g., Viana et al., 2008), especially in countries and regions where there are difficulties complying with the European Union (EU) limit values for particulate matter (Council Directive 1999/30/EC). The source apportionment results allow one to make appropriate suggestions to policy makers to reduce the PM10 mass concentrations.

Although increased attention has been given in recent years to PM10 characterization studies in Flanders, source apportionment has very rarely been carried out. In a number of studies (e.g., Putaud et al., 2010; Vercauteren et al., 2011), use was made of aerosol chemical mass closure, which provides a quantitative measure of some important source categories, but does not identify all sources and, for example, fails to differentiate between natural mineral dust and road dust for the crustal component and does not resolve the different contributors to the organic aerosol component. Kubátová et al. (2002) measured about 100 individual organic compounds in PM10 aerosols for a site in Gent and assessed the contribution from 7 compound classes to the organic matter and the PM10 mass, but this does not really qualify as a source apportionment study. A real source apportionment for the separate coarse (PM10-2) and fine (PM2) size fractions at two locations in Flanders was carried out by Maenhaut and Cafmeyer (1998). No data for water-soluble or organic species or for carbonaceous components were available in this study, though. Vercauteren et al. (2011) used factor analysis to identify the PM10 sources in their one-year study at six sites in Flanders. In the latter study, the following 5 sources were identified: (1) crustal matter, (2) sea salt, (3) secondary aerosol, (4) traffic and metals, and (5) heavy oil burning. Up to 7 sources were identified and quantified for the fine (PM2) size fraction in the study of Maenhaut and Cafmeyer (1998). Besides the three sources (i.e., crustal matter, sea salt, and heavy oil burning), which were identified by Vercauteren et al. (2011), a general pollution source and 3 sources with mainly single elements (i.e., Mn, Br, In) were quantified. The general pollution source and the oil component together accounted for about 50% of the fine PM mass and the crustal component made up for about 35%. Each of the other 4 components accounted for at most 5%. Besides this single source apportionment study by Maenhaut and Cafmeyer (1998), also the study by Maenhaut et al. (2012) is noteworthy. The latter study involved the assessment of the contribution from wood burning to the PM10 mass (and to the organic carbon) for 7 sites in Flanders, thereby making use of the single marker compound levoglucosan and emission factors for Austrian wood stoves (Schmidl et al., 2008). It was found that the annual average contributions from wood burning to the PM10 mass were in the range of 5–6% (from 0.9 to 1.8 $\mu\text{g}/\text{m}^3$) for 6 of the 7 sites, whereas the contribution from wood burning organic carbon (OC) to the PM10 OC was in the range of 20–25% for 6 of the 7 sites.

In the current study, which involved a one-year PM10 characterization at four urban background sites in Flanders, the sources and source contributions were determined by making use of receptor modeling by Positive Matrix Factorisation (PMF) (Paatero and Tapper, 1994). In addition, the contribution from wood burning was independently assessed through making use of levoglucosan as single marker compound, as was done by Maenhaut et al. (2012).

2. Methods

2.1. Sampling sites and aerosol collections

The 4 sampling sites were located at urban background sites in Antwerpen (i.e., at 51°11'55" N, 4°24'24" E, 8 m a.s.l.), Gent (i.e., at 51°03'

30" N, 3°43'46" E, 11 m a.s.l.), Brugge (i.e., at 51°12'48" N, 3°14'04" E, 2 m a.s.l.), and Oostende (i.e., at 51°13'19" N, 2°55'08" E, 5 m a.s.l.). All sites were located in park type areas with a minimum of local influence but still within the inner city. 24-h PM10 aerosol samples (with filter changing at 23:55 UT) were simultaneously taken every 4th day at the 4 sites from 30 June 2011 to 2 July 2012. Leckel SEQ 47/50 low-volume samplers operating at 2.3 m^3/h and 47-mm diameter Pallflex® Tissuquartz™ 2500 QAT-UP (prefired in the factory) quartz fibre filters were used. The exposed area of the filter during sampling (aerosol deposit area) was 11.64 cm^2 and the air volume for each sample was 55.1 m^3 . Consequently, the sampling face velocity was 54.8 cm/s . A total of 93 actual samples were taken at each site; also 9 field blanks were taken at each site; no air was drawn through the field blanks.

2.2. PM mass measurement and chemical analyses

After sampling, the PM10 mass concentration was determined by dual weighing of the filters according to the more stringent PM2.5 European reference method EN 14907 (2005). Both the actual samples and the field blanks were weighed and the PM10 mass data for the actual samples were corrected for the net masses obtained with the field blanks. The median field blank value for the PM10 mass was 0.93 $\mu\text{g}/\text{m}^3$. Filter punches (of 1.5 cm^2) and the remainder of the filter were taken for chemical analysis. Between weighing and analysis, the punches for carbonaceous species and wood burning tracers were stored below $-18\text{ }^\circ\text{C}$ while the punches for elemental analysis and the remaining filter parts for ion analysis were stored below $5\text{ }^\circ\text{C}$. One punch was used for the determination of organic, elemental and total carbon (OC, EC and TC) by thermal-optical analysis using the NIOSH temperature protocol (Birch and Cary, 1996; Maenhaut et al., 2009) and light transmission (TOT) and reflectance (TOR) were simultaneously monitored. The median field blank values were 0.89 $\mu\text{g}/\text{m}^3$ for OC and TC and 0.002 $\mu\text{g}/\text{m}^3$ for EC. The TOT data were retained for further analysis.

A 1.5 cm^2 filter punch of each actual sample and field blank was subjected to analysis for levoglucosan, mannosan, and galactosan (L, M, and G). The analyses were performed by gas chromatography/mass spectrometry (GC/MS) after extraction and trimethylsilylation, as described in Maenhaut et al. (2012), but instead of total ion chromatograms, extracted ion chromatograms (at m/z 217) were used in the current study. The median field blank value was 0.64 ng/m^3 for levoglucosan and below the detection limit of 0.03 and 0.013 ng/m^3 for mannosan and galactosan, respectively.

Another 1.5 cm^2 filter punch of each actual sample and field blank was used for measuring elements. The punch was subjected to microwave assisted acid digestion in nitric acid and hydrogen peroxide based on NEN-EN 14902 (2005). Two elements (i.e., Fe and Zn) were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) with Perkin Elmer Optima 3300 DV and Optima 7300DV instruments according to EN ISO 11885 (2007). For the remaining 13 elements (i.e., Al, As, Ba, Cd, Cr, Cu, Mn, Mo, Ni, Pb, Sb, Ti, V) use was made of inductively coupled plasma atomic mass spectrometry (ICP-MS) with a Perkin Elmer Elan DRC II quadrupole instrument according to EN ISO 17294-2 (2003).

The remainder of the filter was used for the measurement of water-soluble ions. It was subjected to a 15 min ultrasonic extraction with 10 mL Milli-Q water, the mixture was centrifuged and the supernatant was analyzed for 8 species by suppressed ion chromatography with conductometric detection. For measuring the 5 cationic species (NH_4^+ , Na^+ , Mg^{2+} , K^+ and Ca^{2+}) use was made of Dionex CS12A and CG12A analytical and guard columns, with 20 mM methane sulfonic acid as

eluent; for the 3 anionic species (Cl^- , NO_3^- and SO_4^{2-}), Dionex AS11-HC and AG11-HC columns were used with 30 mM NaOH as eluent.

All species and elemental data for the actual samples were corrected for the data obtained for the field blanks.

2.3. Source apportionment

The source profiles were identified and quantified by PMF. Use was made of EPA PMF 5 (Norris et al., 2014) and of its capability of handling multiple site data. Thus, the data sets of the 4 sites were analyzed together. A total of 29 variables (i.e., PM mass, OC, EC, levoglucosan, mannosan, galactosan, 8 water-soluble ions and 15 elements) were included. The input data were prepared using the procedure suggested by Polissar et al. (1998). PM mass concentration was given a very high (400%) uncertainty and was specified as total (and strong) variable. The base model PMF results for a varying number of factors (from 8 to 13) were examined; a number of analyses with constraints were also made and the uncertainties of the results were assessed by classical bootstrap calculations (with 100 bootstraps and using a minimum correlation R-value of 0.6).

Besides the PMF-based source apportionment, a separate quantitative assessment of the contribution from wood burning to the PM10 mass and OC was made by using levoglucosan as single marker compound and emission factors for Austrian wood stoves (Schmidl et al., 2008), as was done by Maenhaut et al. (2012). In particular, PM10 OC and PM10 mass from wood burning were determined by multiplying the levoglucosan concentration by factors of 5.59 and 10.7, respectively.

3. Results and discussion

3.1. Median concentrations and interquartile ranges

The medians and interquartile ranges for the PM10 mass, OC, and EC, the water-soluble ions and the elements at each of the 4 sites are given in Table 1. As there was a very clear seasonal cycle for the monosaccharide anhydrides, seasonal medians and interquartile ranges are given for these species in Table 2 in addition to the overall (annual) data.

For the monosaccharide anhydrides (Table 2), the highest levels at each site are noted in winter, followed by autumn, spring and summer, thus indicating that wood burning is most common in the coldest season of the year. A similar seasonal trend was observed in our previous study (Maenhaut et al., 2012).

The Gent site in the current study was the same as that used in the previous study. The data for the PM10 mass, carbonaceous species and monosaccharide anhydrides of the two studies for this site are fairly similar; the overall medians for OC are the same; for the PM10 mass, the overall median is about 10% lower in the current study and for levoglucosan it is now about 20% higher.

3.2. Time series and correlations between the concentration data from the 4 sites

Fig. 1 shows the time series for levoglucosan in PM10 at the 4 sites. The levoglucosan concentrations of the 4 sites are well correlated with each other, but there are two days for Brugge (i.e., 3 Dec. 2011 and 9 Feb. 2012) with deviating high concentrations. The site-by-site correlation coefficients between the levoglucosan data of Antwerpen, Gent and Oostende were larger than 0.9, whereas those between the data of Brugge and the other 3 sites were around 0.8. When excluding the two deviating high concentrations from Brugge, all site-by-site correlation coefficients were larger than 0.9. The high correlations can be explained by a combination of similar seasonality and meteorology, as well as regional or long-range transport.

Also for the PM10 mass, OC, nitrate and sulfate, very high site-by-site correlation coefficients were obtained. For the PM10 mass the correlation coefficients between the data of Gent and the other 3 sites were all larger than 0.9, and the other values were between 0.85 and 0.9. For OC fairly similar values were observed as for levoglucosan, but the correlation coefficients between the data of Brugge and the other 3 sites were somewhat higher (i.e., around 0.85). For nitrate all site-by-site correlation coefficients were larger than 0.9, with the exception of that between the data of Oostende and Antwerpen, which was 0.87. For sulfate somewhat lower values were observed, but they were all still between 0.81 and 0.97. These high correlations suggest that also

Table 1
Median concentrations and interquartile ranges over the full year (30 June 2011–2 July 2012) for the PM10 mass, OC, EC, 8 water-soluble species (in $\mu\text{g}/\text{m}^3$), and 15 elements (in ng/m^3) at 4 sites in Flanders, Belgium.

	Antwerpen	Gent	Brugge	Oostende
	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)
PM	22 (16.7–32)	25 (17.7–34)	23 (16.0–31)	24 (17.0–30)
OC	2.5 (1.59–4.4)	3.0 (2.0–4.8)	2.1 (1.26–3.9)	1.90 (1.07–3.4)
EC	0.76 (0.60–1.04)	0.71 (0.56–1.03)	0.55 (0.38–0.81)	0.62 (0.45–0.86)
NO_3^-	3.2 (1.88–8.0)	3.7 (1.88–7.5)	3.4 (1.78–7.4)	3.6 (1.73–7.3)
Cl^-	0.38 (0.129–0.93)	0.56 (0.177–1.07)	0.63 (0.26–1.32)	0.86 (0.34–2.4)
SO_4^{2-}	2.2 (1.56–3.3)	2.1 (1.51–3.7)	2.1 (1.38–3.2)	1.99 (1.53–3.1)
Na^+	0.52 (0.23–0.95)	0.57 (0.26–0.96)	0.76 (0.33–1.26)	1.06 (0.46–1.86)
NH_4^+	1.23 (0.52–3.2)	1.18 (0.51–2.9)	1.34 (0.47–2.9)	1.29 (0.27–2.7)
K^+	0.095 (0.072–0.149)	0.109 (0.076–0.155)	0.101 (0.080–0.148)	0.107 (0.084–0.141)
Mg^{2+}	0.101 (0.056–0.143)	0.114 (0.062–0.171)	0.135 (0.070–0.20)	0.170 (0.084–0.28)
Ca^{2+}	0.29 (0.197–0.46)	0.40 (0.23–0.62)	0.26 (0.174–0.44)	0.27 (0.188–0.42)
Al	101 (70–164)	121 (61–210)	60 (26–100)	94 (50–157)
As	1.23 (0.65–1.77)	0.59 (0.36–0.89)	0.52 (0.35–0.98)	0.49 (0.32–0.74)
Ba	6.0 (4.3–8.6)	5.9 (3.0–10.3)	3.9 (1.36–8.0)	4.4 (1.82–8.2)
Cd	0.34 (0.162–0.56)	0.189 (0.102–0.35)	0.163 (0.074–0.49)	0.114 (0.052–0.20)
Cr	2.5 (1.40–4.4)	1.60 (0.56–3.5)	0.98 (0.35–1.89)	1.30 (0.50–2.9)
Cu	15.9 (11.2–24)	9.2 (6.6–14.8)	7.2 (4.9–11.6)	7.9 (4.7–10.8)
Fe	370 (250–540)	330 (175–550)	210 (154–320)	220 (128–350)
Mn	8.8 (6.5–12.0)	8.9 (4.7–16.1)	6.7 (4.0–9.3)	5.7 (2.9–10.2)
Mo	0.82 (0.55–1.41)	0.86 (0.51–1.80)	0.52 (0.34–0.94)	0.55 (0.34–0.89)
Ni	2.3 (1.49–3.6)	2.9 (1.57–4.7)	1.99 (1.06–3.4)	2.7 (1.33–4.8)
Pb	15.0 (8.3–22)	8.9 (6.2–14.4)	5.9 (3.5–12.0)	5.3 (2.8–10.2)
Sb	2.3 (1.48–3.8)	1.45 (1.04–2.4)	1.15 (0.70–1.85)	1.09 (0.66–1.50)
Ti	5.8 (3.57–9.2)	6.7 (3.7–13.1)	3.8 (2.3–6.6)	4.9 (2.7–8.3)
V	1.71 (1.13–2.8)	2.1 (1.08–3.6)	1.69 (1.15–3.6)	2.4 (1.17–4.6)
Zn	35 (23–63)	32 (18.1–53)	22 (11.2–43)	23 (10.4–45)

Table 2

Overall (annual) and seasonal median concentrations and interquartiles ranges for the PM and OC (in $\mu\text{g}/\text{m}^3$) and for the monosaccharide anhydrides (in ng/m^3) at 4 sites in Flanders, Belgium.

	Overall	Spring	Summer	Fall	Winter
	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)
<i>Antwerpen</i>					
PM	22 (16.7–32)	27 (18.4–37)	17.9 (15.5–21)	27 (19.0–34)	24 (16.9–35)
OC	2.5 (1.59–4.4)	2.4 (1.69–4.1)	2.0 (1.59–2.7)	4.7 (2.0–7.0)	3.1 (1.69–4.3)
Levoglucoosan	61 (22–185)	50 (31–93)	17.9 (8.3–28)	142 (42–340)	185 (96–310)
Mannosan	6.9 (2.6–20)	6.2 (3.3–10.3)	1.59 (0.91–3.2)	19.1 (4.5–35)	20 (11.7–32)
Galactosan	2.6 (1.03–7.4)	2.4 (1.34–3.5)	0.80 (0.49–1.22)	6.9 (1.36–10.0)	7.4 (3.8–12.5)
<i>Gent</i>					
PM	25 (17.7–34)	30 (20–42)	20 (15.5–24)	26 (20–35)	25 (17.0–33)
OC	3.0 (2.0–4.8)	2.7 (1.94–4.8)	2.3 (1.91–3.5)	4.7 (2.8–7.3)	3.0 (2.1–4.7)
Levoglucoosan	82 (32–200)	63 (50–150)	23 (14.3–36)	169 (56–350)	191 (121–320)
Mannosan	8.3 (3.1–21)	7.5 (4.7–14.8)	2.2 (1.19–3.8)	13.4 (5.1–35)	21 (12.0–31)
Galactosan	3.4 (1.20–8.3)	2.8 (1.90–5.5)	0.90 (0.63–1.64)	7.3 (1.91–13.7)	8.3 (4.7–14.3)
<i>Brugge</i>					
PM	23 (16.0–31)	27 (18.2–35)	16.9 (15.2–21)	23 (17.3–41)	24 (18.2–32)
OC	2.1 (1.26–3.9)	2.2 (1.47–3.7)	1.53 (1.23–1.99)	3.3 (1.34–7.3)	2.4 (1.26–4.9)
Levoglucoosan	62 (18.8–173)	57 (26–127)	11.5 (8.2–31)	131 (45–350)	142 (81–310)
Mannosan	6.3 (2.1–18.2)	5.4 (2.8–12.8)	1.12 (0.79–3.0)	13.1 (4.2–37)	15.3 (9.0–33)
Galactosan	2.6 (0.97–6.9)	2.1 (1.23–4.5)	0.70 (0.47–1.36)	6.8 (1.56–12.5)	5.7 (3.3–13.6)
<i>Oostende</i>					
PM	24 (17.0–30)	28 (20–40)	17.7 (14.5–22)	25 (19.6–38)	24 (19.1–30)
OC	1.90 (1.07–3.4)	1.93 (1.38–3.2)	1.34 (1.10–1.98)	3.3 (1.20–5.6)	1.90 (0.93–3.6)
Levoglucoosan	39 (12.3–123)	39 (15.4–75)	8.6 (4.9–16.1)	79 (23–220)	122 (53–250)
Mannosan	4.3 (1.26–14.1)	4.0 (2.1–8.4)	1.02 (0.71–1.75)	7.7 (2.3–22)	12.5 (5.9–22)
Galactosan	1.92 (0.73–5.0)	1.74 (0.75–2.5)	0.68 (0.34–0.95)	3.6 (1.01–8.1)	4.9 (2.3–10.0)

most of the PM₁₀ mass, OC and nitrate and sulfate, whereby the latter two species are essentially secondary in Flanders, have a regional character. For EC, which is in Flanders mainly derived from traffic (Lefebvre et al., 2011), lower site-by-site correlation coefficients were noted, with values ranging from 0.61 to 0.82, and thus demonstrating its local character.

3.3. Correlations among and concentration ratios between the monosaccharide anhydrides; relative importance of softwood and hardwood burning

At each of our 4 sites, the 3 monosaccharide anhydrides levoglucosan, mannosan and galactosan were very highly correlated with each other. All correlation coefficients were higher than 0.95. Inspection of the various scatter plots indicated that the excellent correlations were not due to a few exceptional high concentration data. As a consequence of those

excellent correlations, the concentration ratios among the anhydrosugar species showed relatively little variability over the entire year.

The ratio of levoglucosan to mannosan (L/M) has been proposed to distinguish between softwood and hardwood burning (e.g., Schmidl et al., 2008). Based on data that were obtained for the combustion of common hardwoods (beech and oak) and softwoods (spruce and larch) in wood stoves in Austria, Schmidl et al. (2008) derived the following equation for obtaining the % of spruce burned (relative to the % of hardwood):

$$\% \text{ spruce} = (14.8 - R_{L/M}) / 0.112 \quad (1)$$

where $R_{L/M}$ is the ratio of the levoglucosan to mannosan concentrations in the ambient air.

By applying Eq. (1) to our individual levoglucosan/mannosan ratios for the 4 sites, individual % spruce data were obtained for each sample.

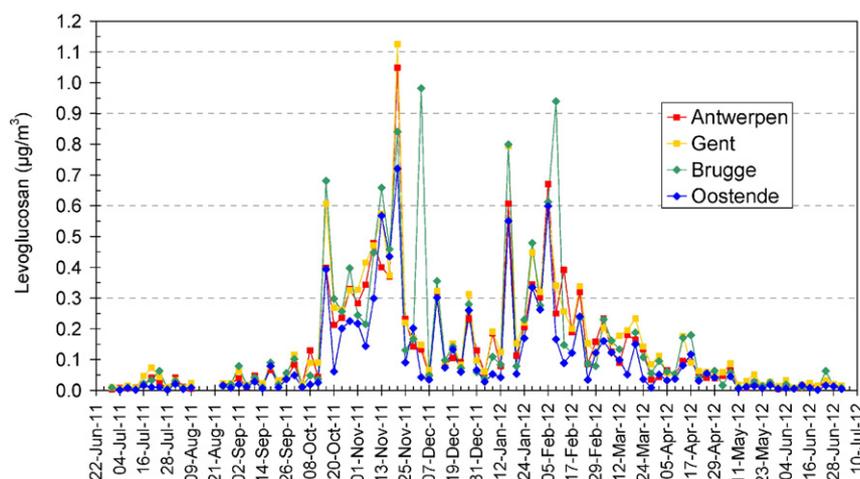


Fig. 1. Time series for PM₁₀ levoglucosan at 4 sites in Flanders, Belgium.

The averages and associated standard deviations for the % spruce data were $52 \pm 13\%$ for Antwerpen, $45 \pm 15\%$ for Gent, $44 \pm 23\%$ for Brugge, and $49 \pm 24\%$ for Oostende. These percentages are lower than the percentages of 70%, which were observed for our previous study in 2010–2011 (Maenhaut et al., 2012). It is estimated that the uncertainty in our % spruce is of the order of 10% points and that the uncertainty is largely of a systematic nature (i.e., resulting from the calibration curves for levoglucosan and mannosan in the current and previous studies). The difference between the around 50% in the current study and the 70% in the previous one may therefore not be significant.

3.4. Source identification and apportionment by PMF

A summary of the base model PMF results for a varying number of factors (from 8 to 13) is presented in Table 3. The factors in the 8-factor solution (with in parentheses the overall average percentage contribution to the observed PM10 mass) are named: wood burning (11%), secondary nitrate (25%), secondary sulfate (15%), sea salt (12%), aged sea salt (17%), crustal matter (10%), non-ferrous metals (1.9%), and traffic (7%). In the 9-, 10-, and 11-factor solutions, the following factors are sequentially added: non-exhaust traffic, heavy oil burning, and an organic factor. When comparing the percentages of the 11- and 10-factor solutions, it appears that the organic factor (with 13%) is added at the expense of secondary sulfate (down from 13% to 8%) and traffic (down from 10% to 5%). The only main species in the organic factor was OC, with a fractional contribution of 0.41. Taking into account that conversion factors for converting OC into organic matter (OM) range from 1.4 to over 2.0, with ratios of 1.6 ± 0.2 for urban aerosols appearing to be more accurate (Turpin and Lim, 2001), this means that the fraction of OM in the organic factor may be up to 0.66 or even higher. It is not easy to come up with a specific source or source process for such organic factor; it might represent or include secondary organic aerosol (SOA). Adding even one or two factors beyond 11 resulted in additional factors, which were mainly characterized by elements, which are even harder to explain.

Considering the above, it was decided to retain the 10-factor solution. A few constrained runs were also made for 10 factors, e.g., with the Cl/Na ratio in the sea-salt factor forced to the ratio of 1.8 in average bulk sea water (Riley and Chester, 1971) and with the monosaccharide anhydrides pulled down maximally (or set to zero) in the crustal matter and non-exhaust traffic factors but this led only to minor changes in the results, so that the base model 10-factor solution was retained. Incidentally, the Cl/Na ratio in this solution was 1.99, which is only 10% larger than that in sea water, and the Mg/Na ratio in this solution was the same (i.e., 0.12) as in sea water.

The source profiles for the base model 10-factor solution and the average percentage apportionments of the various species and elements to the factors are shown in Figs. 2 and 3. The key species in the various profiles (that is those with typically at least one third of their mass, on average, attributed to the profile) are shown in Table 4. Considering the key species, the source association of the 10 factors seems overall quite logical. It appears, though, that the secondary nitrate and secondary sulfate are not well separated from each other, as the former also accounts for a substantial amount of sulfate. The association of a lot of OC with the secondary sulfate factor is also somewhat puzzling; this OC is like the sulfate also likely mostly secondary and thus SOA, whereas the OC in the wood burning factor is mostly primary. The secondary sulfate factor could thus also be termed as a mixed secondary sulfate - SOA factor. It is further evident that the traffic and non-exhaust traffic factors are not well separated; the elements Cu, Fe, Mo and Sb, which are key elements in the traffic factor, are essentially associated with non-exhaust traffic emissions (e.g., Weckwerth, 2001; Wählin et al., 2006; Dall'Osto et al., 2013). It may therefore be concluded that the non-exhaust traffic factor is actually an additional non-exhaust traffic factor, since it only represents a minor fraction of the non-exhaust traffic emissions and most of those emissions are accounted for by the traffic factor.

The overall average percentage apportionments of the experimentally measured PM10 mass were as follows: wood burning 9.5%, secondary nitrate 24%, secondary sulfate 12.6%, sea salt 10.0%, aged sea salt 19.2%, crustal matter 9.7%, non-ferrous metals 1.81%, traffic 10.3%, non-exhaust traffic 0.52%, and heavy oil burning 3.0%. Our percentages for the sum of secondary nitrate and secondary sulfate (which is 37%), crustal matter and sea salt agree well with the data that were obtained by aerosol chemical mass closure calculations in the one-year aerosol characterization study at 6 sites in Flanders of Vercauteren et al. (2011). These authors found that secondary inorganic ions (nitrate, sulfate and ammonium), crustal matter and sea salt accounted, on average, for respectively 41%, 14% and 8% of the experimental PM10 mass. Our average apportionments for each of the 4 sites separately are presented in Figs. 4 and 5. The annual mean concentrations for both sea salt and aged sea salt (Fig. 4) are clearly largest for Oostende, followed by Brugge, and then come Gent and Antwerpen with similar data for these two sites. As Oostende is located on the North Sea coast and the sampling sites of Brugge, Gent and Antwerpen were at respectively 25, 90 and 110 km from the North Sea, the observed tendency is fully logical. Heavy oil combustion exhibits a fairly similar trend as the two sea-salt components with largest level and percentage at Oostende ($1.19 \mu\text{g}/\text{m}^3$; 4.4%) followed by Brugge ($0.77 \mu\text{g}/\text{m}^3$; 2.9%) and then Gent ($0.78 \mu\text{g}/\text{m}^3$; 2.5%) and Antwerpen ($0.64 \mu\text{g}/\text{m}^3$; 2.4%); this is likely caused by the larger impact from ship emissions at the sites closer to the coast. It is well-known that ships are an important source of heavy

Table 3
Summary of the PMF base results as a function of the number of factors: $Q_{\text{true}}/Q_{\text{expected}}$ ratio, factor names with in parentheses the overall average percentage contribution of the factor to the experimental PM10 mass; the second percentage for the wood burning factor is the overall average percentage contribution of wood burning to the experimental OC mass.

#Factors	$Q_{\text{true}}/Q_{\text{exp}}$	Factors (%)
8	2.96	WoodBurning (11, 29), SecNitrate (25), SecSulfate (15), SeaSalt (12), AgedSeaSalt (17), Crustal (10), NonFerrous (1.9), Traffic (7)
9	2.65	WoodBurning (9, 27), SecNitrate (26), SecSulfate (14), SeaSalt (12), AgedSeaSalt (15), Crustal (9), NonFerrous (1.6), Traffic (12), NETraffic (1.3)
10	2.39	WoodBurning (9, 29), SecNitrate (24), SecSulfate (13), SeaSalt (10), AgedSeaSalt (19), Crustal (10), NonFerrous (1.8), Traffic (10), NETraffic (0.5), HeavyOil (3)
11	2.17	WoodBurning (9, 26), SecNitrate (20), SecSulfate (8), SeaSalt (10), AgedSeaSalt (20), Crustal (8), NonFerrous (1.5), Traffic (5), NETraffic (0.3), HeavyOil (5), Organic (13)
12	2.02	WoodBurning (7, 22), SecNitrate (16), SecSulfate (13), SeaSalt (9), AgedSeaSalt (20), Crustal (9), NonFerrous (0.7), Traffic (5), NETraffic (0), HeavyOil (3), Organic (13), AlCrTiZn (3)
13	1.86	WoodBurning (8, 23), SecNitrate (18), SecSulfate (8), SeaSalt (9), AgedSeaSalt (18), Crustal (10), NonFerrous (0.3), Traffic (6), NETraffic (0), HeavyOil (3), Organic (14), MnZn (3), CrMoTi (4)

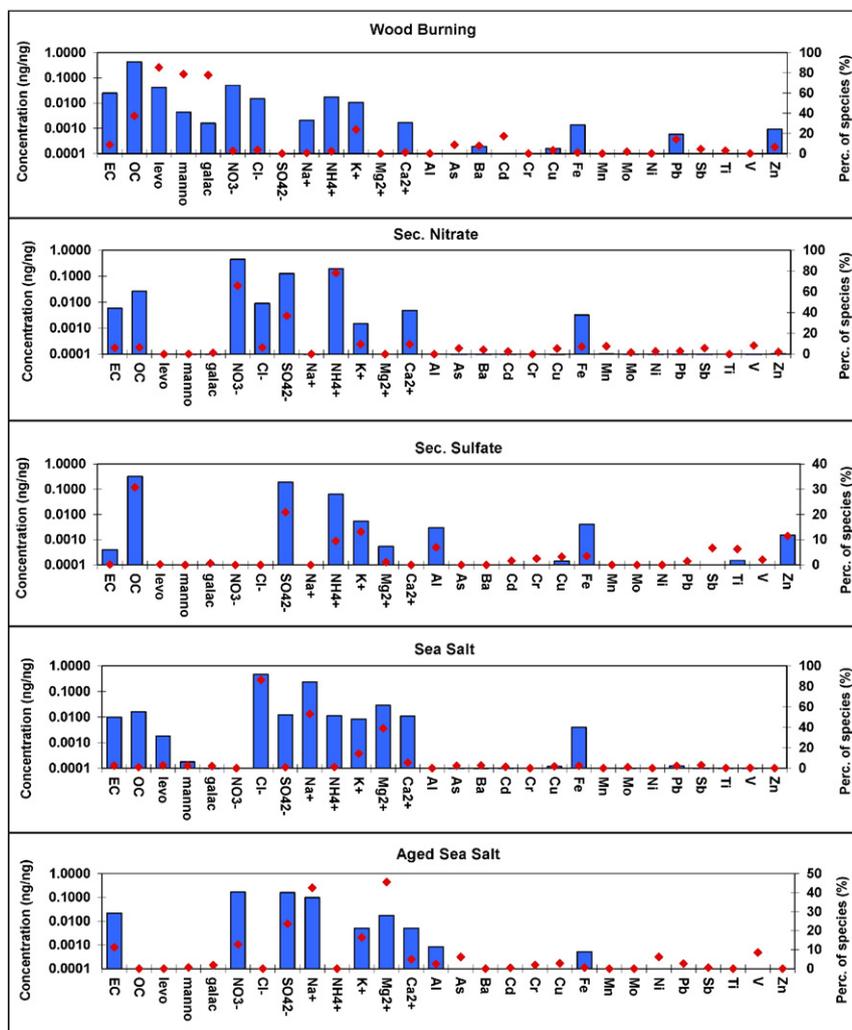


Fig. 2. Source profiles (blue bars) and average percentage attributions (red diamonds) of the various species to the factors for the base model 10-factor solution: Results for 5 of the 10 factors.

oil combustion particles (Viana et al., 2014). It is further noteworthy that the non-ferrous metals factor exhibits by far its highest concentration and percentage at Antwerpen. There is a major non-ferrous metals plant in Hoboken, which is located within greater Antwerpen, and this plant is still a large source of airborne As, Cd, and Pb (VMM, 2014). This plant is undoubtedly the major source of our non-ferrous metals component at Antwerpen and possibly also at our other 3 study sites. Finally, the impact of crustal matter is clearly more pronounced at Gent than at the other 3 sites. This is likely caused by the fact that there is a lot of exposed soil at the urban background site of Gent, which is not the case at the urban background sites in the other 3 cities.

The uncertainties of the average apportionments and source profiles, as derived from the bootstrap calculations, were overall quite reasonable. The base bootstrap box plot with the variability in the attributed percentage of each species to each of the 10 factors is shown in Fig. 6. For the key species mentioned above, the median bootstrap value was typically within 10% of the base run value and the interquartile range of the bootstrap run values was mostly within $\pm 10\%$ of the bootstrap run median; one clear exception was noted for the key species in the secondary nitrate profile, where the 25th percentiles of the bootstrap values were only 38% and 58% of the medians for nitrate and ammonium, respectively. As far as the variability in the concentration of the key species in the source profiles is concerned, similar results were obtained. The median bootstrap value was again typically within 10% of the base run value and the interquartile range of the bootstrap run

values was mostly within $\pm 10\%$ of the bootstrap run median; one clear exception again was noted for the key species in the secondary nitrate profile, where the 25th percentiles of the bootstrap values were only 40% and 57% of the medians for nitrate and ammonium, respectively. Furthermore, for both the attributed percentages and the source profiles, the base run value was for all key species situated within the interquartile range of the bootstrap run values.

3.5. Contribution from wood burning to the PM10 OC and the PM10 mass and to selected species

The overall average contributions from wood burning to the experimentally measured PM10 mass as derived from PMF and also the annual average contributions for each of our 4 sites were presented in the previous section. Here, the contributions to both the PM10 mass and PM10 OC are examined as a function of season for each of the sites and the data as derived from PMF are compared with those obtained from using levoglucosan as single marker compound and the same conversion factors as employed in our previous study on wood burning in Flanders (Maenhaut et al., 2012). The averages and associated standard deviations as obtained by the two different approaches are presented in Table 5. The data derived from levoglucosan compare very well with those that were obtained in our previous study using the same approach. The annual average levoglucosan-derived contributions from wood burning OC to the PM10 OC are in the range of 19–23% for our 4

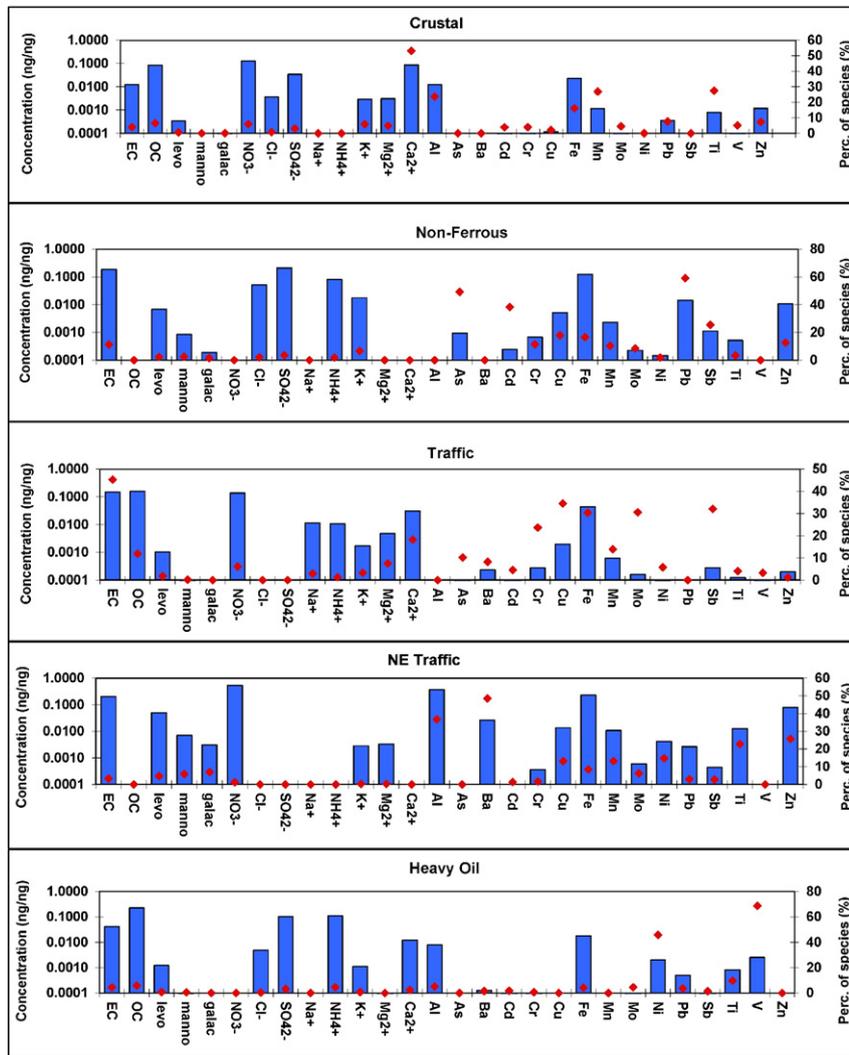


Fig. 3. Source profiles (blue bars) and average percentage attributions (red diamonds) of the various species to the factors for the base model 10-factor solution: Results for the additional 5 of the 10 factors.

sites; the averages for summer are 3.6–5.8% and those for winter 34–41%. In the earlier study (Maenhaut et al., 2012), the annual average contributions from wood burning OC to the PM10 OC were in the range 20–25% for 6 of our 7 sites; the averages for summer were 2.0–3.9% for the 6 sites and the corresponding data for winter were 36–43%. As to the contribution from wood burning to the PM10 mass, the levoglucosan-derived annual average percentages in the current study are in the range of 3.9–5.8% for our 4 sites; the averages for summer are 0.55–1.31% and those for winter 6.5–9.8%. In the earlier study

(Maenhaut et al., 2012), the annual average percentages were in the range 4.8–6.3% for 6 of our 7 sites; the averages for summer were 0.51–1.14% and the corresponding data for winter were 8.6–11.3%. In both the earlier and the current study, the levoglucosan-derived average percentage contributions from wood burning to the OC and PM

Table 4

Key species in the various factor profiles (key species are defined as those species with typically at least one third of their mass, on average, attributed to the profile).

Factor profile	Key species
Wood burning	OC, levoglucosan, mannosan, galactosan
Secondary nitrate	Nitrate, sulfate, ammonium
Secondary sulfate	OC, sulfate
Sea salt	Cl ⁻ , Na ⁺ , Mg ²⁺
Aged sea salt	Sulfate, Na ⁺ , Mg ²⁺
Crustal matter	Ca ²⁺ , Al, Mn, Ti
Non-ferrous metals	As, Cd, Pb
Traffic	EC, Cu, Fe, Mo, Sb
Non-exhaust traffic	Al, Ba, Zn
Heavy oil burning	Ni, V

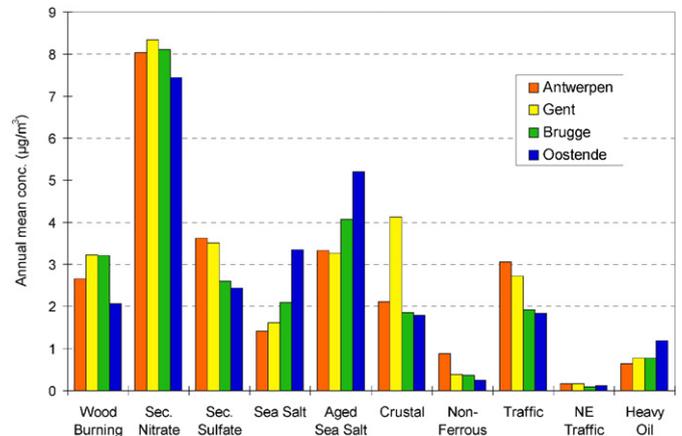


Fig. 4. Annual mean concentrations of the 10 factor profiles for each of the 4 study sites.

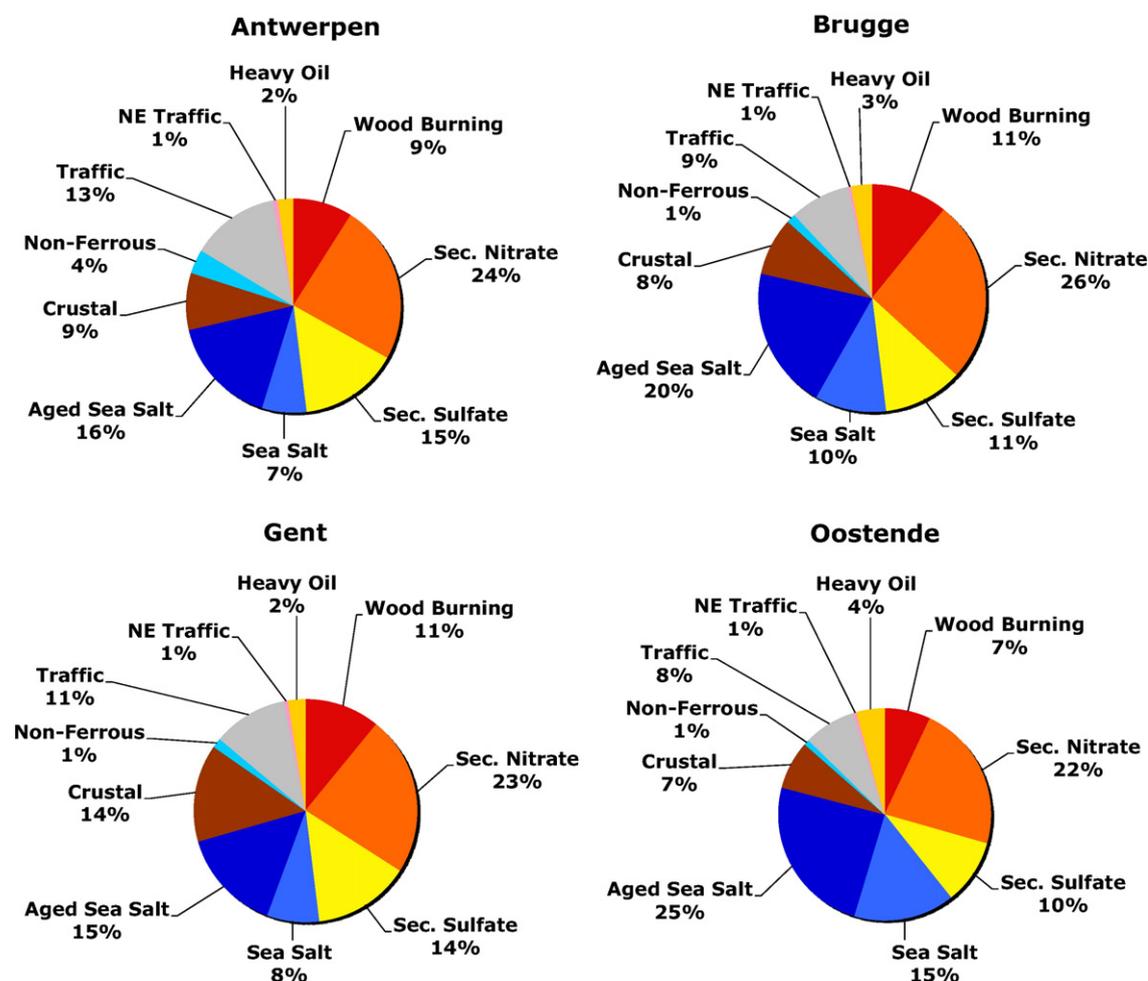


Fig. 5. Average percentage apportionments of the experimentally measured PM10 mass for each of the 4 study sites.

mass were largest for winter, followed by fall, spring, and summer. This was also the case for the corresponding PMF-derived average percentage contributions.

However, for both OC and the PM mass the PMF data are substantially larger than those derived from levoglucosan. This could partly be caused by the fact that the PMF wood smoke factor may include some SOA, whereas this is not the case for the wood smoke OC and PM mass derived with the conversion factors of Schmidl et al. (2008). One could also question the reliability of the PMF approach. However, for the key species (OC, levoglucosan, mannosan and galactosan) of the wood burning factor, the interquartile ranges of the bootstrap run values for both the attributed percentages and concentrations in the source profile were within less than 10% (typically only a few percent) of the median of the bootstrap run values and those medians deviated by less than 10% (typically only a few percent) from the base run values. Furthermore, when considering the PMF solutions for the various numbers of factors (from 8 to 13; see Table 3), the apportionments to wood burning for both the PM mass and the OC appear quite robust. All percentages are within 20% relative of those for the 10-factor PMF solution. Considering all this, it is estimated that the uncertainty in the PMF-derived wood smoke OC and PM mass attributions is at most 20% relative. As to the uncertainty of the levoglucosan-derived contributions, in our earlier study (Maenhaut et al., 2012) we estimated that it is around 30%. The latter uncertainty was likely underestimated. In any case, we have to conclude that the conversion factors from levoglucosan to wood smoke OC and PM of 5.59 and 10.7, respectively, as obtained for Austrian wood stoves by Schmidl et al. (2008) are too low for wood burning in Flanders. From the scatter plots of PMF-derived wood

smoke OC and PM versus levoglucosan (see Fig. 7), we arrive at conversion factors of 9.7 and 22.6, respectively. The conversion factor of 22.6 for wood smoke PM10 might seem large. However, similar or even larger factors were found in (or can be derived from) some other recent studies in central and northern Europe. In their winter-time study at a residential site in The Netherlands, Kos and Weijers (2009) derived a conversion factor of 37. Saarnio et al. (2012) used a conversion factor of 24 ± 9 in their study in the Helsinki Metropolitan Area. From the PMF wood smoke and levoglucosan data for 4 sites in Switzerland (Gianini et al., 2013), conversion factors ranging from 15.3 to 29 (median: 24) can be derived. The regression line between the PMF wood smoke and levoglucosan data for a study in a Swiss Alpine valley (Ducret-Stich et al., 2013) indicates a conversion factor of 37. From a similar regression line for a study in the Czech Republic (Hovorka et al., 2015) a conversion factor of 27.9 can be deduced, be it that the regression line pertained to the PM1 aerosol. Our PMF-derived annual average wood smoke percentages are also not unreasonable large. For example, in their winter-time study, Kos and Weijers (2009) attributed between 9 and 27% of the PM10 mass to wood burning. Our average PMF winter-time percentages are in the range 12.5–20% for our 4 sites. Saarnio et al. (2012) found that the average contribution from wood burning to the PM2.5 mass during the cold season ranged from 18 to 29% at urban sites in the Helsinki Metropolitan Area and from 31 to 66% at suburban sites. Gianini et al. (2013) found by PMF annual average percentage wood smoke contributions of 14% for 3 of their Swiss sites and of 31% for the fourth site. Our PMF-derived annual wood smoke average contributions are in the range 7.1–11%. Ducret-Stich et al. (2013) found in their Swiss Alpine valley study, for which the

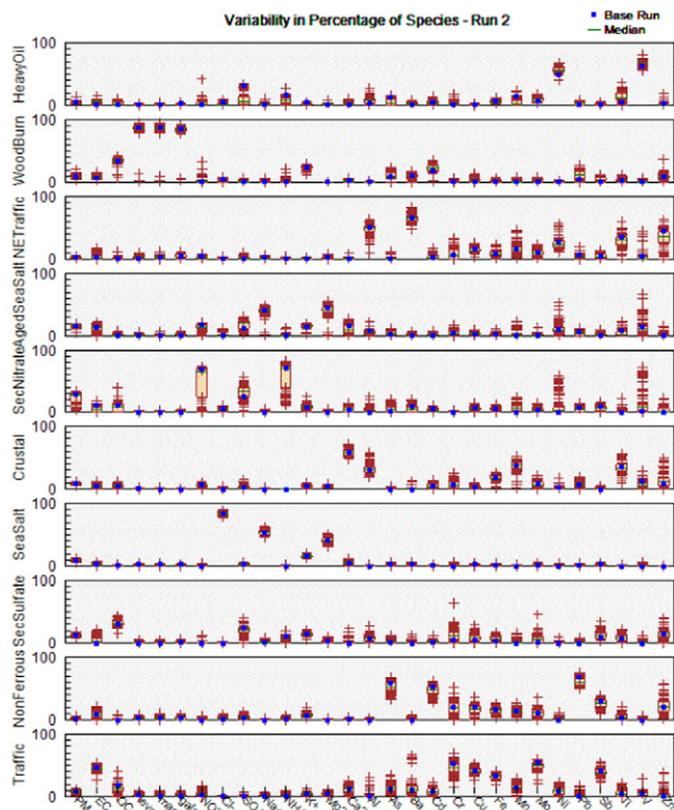


Fig. 6. Base bootstrap box plot with the variability in the attributed percentage of each species to each of the 10 factors. The box represents the interquartile range of the bootstrap run values.

samplings were made from November 2007 to June 2009, that 18% of the PM₁₀ mass was, on average, attributed to wood burning. Much larger annual and winter-time average percentages than ours were obtained by Nava et al. (2015) in their PMF source study for the urban background site of Capannori, Lucca, northern Tuscany. These authors found that biomass burning accounted for 37% of the PM₁₀ mass as annual average, and more than 50% during winter.

While wood burning is the dominant source for its key marker species levoglucosan, mannosan and galactosan (Fig. 2, Table 6), for water-soluble K⁺ and Zn, which are also often used as tracers for wood or biomass burning, medians of less than 20% and less than 6%, respectively, of the experimentally measured concentrations of these two species are obtained for the percentage attributions to wood burning (Table 6). Interestingly, As, Cd and Pb, which were mainly attributed to the non-ferrous metals factor, had similar or higher median percentage apportionments to wood burning as K⁺, in particular for the sites of Gent and Brugge (Table 6). This strongly suggests that the wood burned was contaminated with those elements. Nava et al. (2015) also found that wood burning can be a substantial source of several heavy metals at their urban background site of Capannori in northern Tuscany. They did not measure As and Cd, but found that wood burning explained 18% of the Pb. Table 6 further shows that the non-ferrous metals factor is not only a dominant source of As, Cd and Pb, but also a prominent source of Sb and Zn. The latter two elements most likely also have the non-ferrous metals plant in Hoboken as major source for this factor; their percentage attributions to the non-ferrous metals factor are highest for Antwerpen and show a clear tendency to decrease with distance from Hoboken.

4. Conclusions

The current study, which involved 24-h PM₁₀ samplings in parallel every fourth day at four sites in Flanders over a full year (from 30 June

Table 5

Overall (annual) and seasonal means and associated standard deviations for the % contribution from wood burning (WB) to the experimental PM₁₀ OC and PM₁₀ mass and for the PM₁₀ mass concentration (in µg/m³) from wood burning for 4 sites in Flanders, Belgium. For the percentage contribution from wood burning, both results obtained using levoglucosan as single marker and results derived from PMF are given; for the PM₁₀ mass data from wood burning, the PMF results are given.

	Overall	Spring	Summer	Fall	Winter
	Mean ± s.d.				
Antwerpen					
% of OC levo.	20 ± 15	15.1 ± 9.6	4.6 ± 3.6	22 ± 13	38 ± 10
% of OC PMF	28 ± 27	20 ± 17	2.9 ± 4.5	33 ± 27	58 ± 19
% of PM levo.	5.0 ± 4.7	2.7 ± 1.9	1.02 ± 0.78	7.6 ± 5.6	8.3 ± 3.6
% of PM PMF	9.0 ± 10.2	4.6 ± 4.2	0.83 ± 1.37	14.5 ± 12.7	16.4 ± 8.5
WB PM (µg/m ³) PMF	2.7 ± 3.9	1.27 ± 1.31	0.15 ± 0.26	4.5 ± 5.5	4.8 ± 3.9
Gent					
% of OC levo.	21 ± 14	17.3 ± 9.6	5.5 ± 3.6	22 ± 12	36 ± 7
% of OC PMF	31 ± 26	24 ± 16	4.6 ± 5.9	36 ± 25	59 ± 14
% of PM levo.	5.8 ± 5.3	3.3 ± 2.1	1.31 ± 0.88	9.0 ± 6.8	9.1 ± 3.8
% of PM PMF	11.0 ± 12.1	5.6 ± 4.4	1.35 ± 1.76	18.5 ± 15.9	19.0 ± 8.9
WB PM (µg/m ³) PMF	3.2 ± 4.5	1.70 ± 1.42	0.26 ± 0.38	5.6 ± 6.5	5.5 ± 4.3
Brugge					
% of OC levo.	23 ± 17	19.0 ± 13.1	5.8 ± 6.0	25 ± 14	41 ± 12
% of OC PMF	33 ± 30	25 ± 23	5.2 ± 8.8	38 ± 29	64 ± 22
% of PM levo.	5.7 ± 6.2	3.2 ± 2.5	1.12 ± 1.19	8.2 ± 6.5	9.8 ± 7.3
% of PM PMF	10.8 ± 13.5	5.4 ± 5.5	1.27 ± 2.10	16.6 ± 14.8	20.1 ± 15.7
WB PM (µg/m ³) PMF	3.2 ± 5.0	1.52 ± 1.57	0.20 ± 0.33	5.1 ± 5.8	6.1 ± 6.4
Oostende					
% of OC levo.	19.1 ± 14.7	16.7 ± 12.9	3.6 ± 2.7	20 ± 12	34 ± 9
% of OC PMF	25 ± 26	23 ± 23	0.88 ± 2.04	30 ± 26	47 ± 24
% of PM levo.	3.9 ± 4.5	2.3 ± 2.0	0.55 ± 0.39	5.9 ± 5.2	6.5 ± 4.8
% of PM PMF	7.1 ± 9.9	4.1 ± 4.5	0.26 ± 0.63	11.6 ± 12.0	12.5 ± 11.4
WB PM (µg/m ³) PMF	2.1 ± 3.5	1.00 ± 1.12	0.04 ± 0.09	3.7 ± 4.7	3.5 ± 4.0

2011 to 2 July 2012), was the first one in Flanders and in Belgium, in which use was made of PMF for source apportionment. From aerosol chemical closure calculations, which were made in the study of Vercauteren et al. (2011), it was found that secondary inorganic ions (nitrate, sulfate and ammonium), crustal matter and sea salt accounted, on average, for respectively 41%, 14% and 8% of the experimental PM₁₀ mass. The PMF analysis in the current study revealed the presence of 10 source profiles, with in parentheses the following overall average percentage contributions to the experimentally measured PM₁₀ mass: wood burning (9.5%), secondary nitrate (24%), secondary sulfate (12.6%), sea salt (10.0%), aged sea salt (19.2%), crustal matter (9.7%),

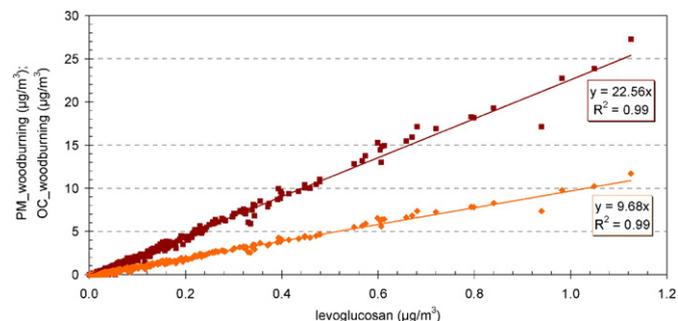


Fig. 7. Scatter plots of PMF-derived wood smoke PM (brown squares) and OC (red diamonds) versus levoglucosan and regression lines, forced through the origin.

Table 6

Medians and interquartile ranges over the full year (30 June 2011–2 July 2012) for the percentage attribution of selected species to wood burning and to the non-ferrous metals factor.

	Antwerpen	Gent	Brugge	Oostende
	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)	Median (25%–75%)
<i>Wood burning</i>				
EC	4.4 (0.3–10.1)	6.4 (1.0–12.1)	6.6 (0.6–12.2)	2.8 (0–7.7)
OC	24 (2.2–47)	30 (4.9–53)	26 (2.5–56)	15.1 (0–46)
Levo.	75 (21–88)	79 (49–92)	81 (40–92)	61 (0–86)
Manno.	67 (24–79)	78 (51–88)	77 (29–93)	57 (0–85)
Galac.	68 (27–82)	74 (46–85)	74 (23–88)	53 (0–84)
K ⁺	11.1 (1.1–34)	18.5 (3.3–36)	11.9 (1.0–33)	5.1 (0–28)
As	3.3 (0.2–7.9)	11.2 (1.4–20)	6.3 (1.2–14.4)	2.6 (0–11.8)
Cd	6.2 (0.5–18.3)	20 (2.3–39)	10.3 (2.2–27)	8.9 (0–31)
Pb	5.6 (0.4–13.7)	12.3 (2.3–26)	11.0 (1.8–24)	6.7 (0–22)
Zn	3.4 (0.2–10.2)	4.7 (0.6–14.0)	5.6 (0.5–14.7)	2.5 (0–10.6)
<i>Non-ferrous metals factor</i>				
As	55 (46–73)	50 (31–72)	34 (18–56)	25 (4.6–42)
Cd	54 (39–73)	35 (26–59)	28 (14.6–46)	24 (6.0–40)
Pb	74 (66–83)	47 (37–57)	48 (29–60)	32 (7.2–55)
Sb	30 (21–44)	19.2 (13.4–31)	18.0 (7.7–29)	12.3 (2.5–25)
Zn	20 (13–31)	9.5 (7.2–16.1)	9.8 (3.5–20)	5.8 (0.7–12.1)

non-ferrous metals (1.81%), traffic (10.3%), non-exhaust traffic (0.52%), and heavy oil burning (3.0%). Thus, the secondary inorganic aerosol component of the study of Vercauteren et al. (2011) was split up into two components in the current study, but together they explained 37%, which agrees well with the 41% for secondary inorganic ions mass in the mass closure calculations of Vercauteren et al. (2011). It should be noted that our aged sea-salt factor also contained a substantial amount of secondary nitrate and sulfate (see Fig. 2). Also our percentages for crustal matter and sea salt are in good agreement with the mass closure data of Vercauteren et al. (2011). The main difference between our PMF source apportionment and the earlier source identification and apportionment studies is that no wood burning source was identified in those earlier studies. This is attributed to the fact that no key indicators for wood burning, such as levoglucosan, were measured in those studies. Wood burning explained, on average, 29% of the PM10 OC and its percentage contributions to both the PM10 mass and OC showed for each of the four sites a clear seasonal variation, with highest percentages in winter, followed by fall, spring, and summer. The average percentage contributions of wood smoke for the four sites were quite substantial in winter and ranged from 12.5 to 20% (from 3.5 to 6.1 $\mu\text{g}/\text{m}^3$) for the PM10 mass and from 47 to 64% for PM10 OC.

Noteworthy in the current study is that the percentage contribution of the heavy oil burning component showed a clear tendency to decrease with distance from the coast; this was likely caused by the larger impact from ship emissions at the sites closer to the coast. It was further noteworthy that the non-ferrous metals factor, which was characterized by As, Cd and Pb, exhibited by far its highest percentage at Antwerpen; there is a major non-ferrous metals plant in Hoboken, which is located within greater Antwerpen, and this plant is still a large source of airborne As, Cd, and Pb (VMM, 2014). This plant is undoubtedly the major source of our non-ferrous metals component at Antwerpen and perhaps also at our other 3 study sites. Wood burning appeared to be also a notable source of As, Cd, and Pb. These three elements had similar or higher median percentage apportionments to wood burning as K⁺, which is typically used as an indicator element for wood smoke.

Besides the PMF-based source apportionment, a separate quantitative assessment of the contribution from wood burning to the PM10 mass and OC was made by using levoglucosan as single marker compound and emission factors for Austrian wood stoves (Schmidl et al., 2008), as was done by Maenhaut et al. (2012) for 2010–2011. In particular, PM10 OC and PM10 mass from wood burning were determined by multiplying the levoglucosan concentration by factors of 5.59 of 10.7,

respectively. Using this approach, very similar apportionments were found as in our 2010–2011 study. However, they were much lower than those deduced from PMF. It seems that the conversion factors of Schmidl et al. (2008) are not applicable to wood burning in Flanders. From scatter plots of the PMF-derived wood smoke PM10 OC and PM versus levoglucosan, we arrive at conversion factors of 9.7 and 22.6, respectively. It is recommended to use these latter factors for deriving the contribution from wood smoke when making use of levoglucosan as single marker in future studies in Flanders. It is estimated that the percentage uncertainty, which is associated with these conversion factors is smaller than 20%. The uncertainty associated with the use of the Schmidl et al. (2008) conversion factors for Flanders was estimated to be of the order of 30% in our 2010–2011 study (Maenhaut et al., 2012).

It is concluded that the impact from wood burning on the PM10 mass and OC levels in Flanders is substantially larger than previously thought. Considering that the contribution from wood burning to the PM10 mass concentrations is largest in winter and that most of the exceedances in Flanders of the EU daily PM10 mass limit of 50 $\mu\text{g}/\text{m}^3$ limit occur in the cold season (Maenhaut et al., 2012), this conclusion has important implications for policy makers.

Acknowledgements

This study received financial support from the Life+ Project ATMOSYS of the European Commission (LIFE09 ENV/BE/000409). The authors are indebted to Franco Lucarelli and Silvia Nava from the I.N.F.N.-Florence and Department of Physics and Astronomy, University of Florence, Italy, for advice on the use of EPA PMF 5.

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