Fossil fuel combustion, biomass burning and biogenic sources of fine carbonaceous aerosol in the Carpathian Basin

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Abstract. Fine-fraction aerosol samples were collected, air pollutants and meteorological properties were measured in-situ in regional background environment of the Carpathian Basin, a suburban area and central part of its largest city, Budapest in each season for 1-year-long time interval. The samples were analysed for $PM_{2.5}$ mass, organic carbon (OC), elemental carbon (EC), water-soluble OC (WSOC), radiocarbon, levoglucosan (LVG) and its stereoisomers, and some chemical elements. Carbonaceous aerosol species made up 36% of the $PM_{2.5}$ mass with a modest seasonal variation and with a slightly increasing tendency from the regional background to the city centre (from 32 to 39%). Coupled radiocarbon-LVG marker method was applied to apportion the total carbon (TC=OC+EC) into contributions of EC and OC from fossil fuel (FF) combustion (EC_{FF} and OC_{FF}, respectively), EC and OC from biomass burning (BB) (EC_{BB} and OC_{BB}, respectively) and OC from biogenic sources (OC_{BIO}). Fossil fuel combustion showed rather constant daily or monthly mean contributions (of 35%) to the TC in the whole year in all atmospheric environments, while the daily contributions of BB and biogenic sources changed radically (from <2 up to 70–85%) at all locations and over the years. In October, the three major sources contributed equally to the TC in all environments. In January, it was the BB that was the major source with a share of 70% at all sites. The contributions from biogenic sources in January were the smallest. In April, FF combustion and biogenic sources were the largest two contributors at all locations with typical shares of 45–50% each. In July, biogenic sources became the major source type with a monotonically increasing tendency (from 56 to 72%) from the city centre to the regional background. The share of BB was hardly quantifiable in July. The EC_{FF} made up more than 90% of EC in April and July, while in October and January, the contributions of EC_{BB} were considerable. Biomass burning in winter and autumn offers the largest and considerable potentials for improving the air quality in cities as well as in rural areas of the Carpathian Basin.

1 Introduction and objectives

Carbonaceous aerosol constituents make up a major part (e.g. 20–60% in the continental mid-latitudes and up to 90% in tropical forests) of the $PM_{2.5}$ mass (Kanakidou et al., 2005; Fuzzi et al., 2015). Their largest emission or production source types are fossil fuel (FF) combustion, biomass burning (BB) and biogenic sources (Le Quéré et al., 2018). These processes also represent the highest source of certain important aerosol species such as soot (species mainly containing C with imperfect/fragmented graphitic structure; Andreae and Gelencsér, 2006) and of some pollutant or greenhouse gases such as CO, NO_x, CO_2 and volatile organic compounds (VOCs) on global spatial scale (Wiedinmyer et al., 2011; Tian et al., 2016). The sources produce both primary and secondary particles, and they are linked to a variety of anthropogenic activities directly or indirectly and in many ways (Hallquist et al., 2009). The perturbations in atmospheric concentrations and chemical, physical and meteorological properties caused by these sources have important consequences on the Earth system. The related processes include the radiation balance (Lohmann et al., 2000), cloud formation/properties, water cycling and other biogeochemical cycles (Andreae and Rosenfeld, 2008; Cecchini et al., 2017), atmospheric chemistry and nucleation (Fuzzi et al., 2015; Nozière et al., 2015; Kirkby et al., 2016), atmospheric transport/mixing (Rosenfeld et al., 2019), forest growth and agriculture production (Artaxo et al., 2009; Rap et al., 2015), ecosystems (Cirino et al., 2014), built environment and cultural heritage (Bonazza et al., 2005), and human health/wellbeing (Lelieveld and Pöschl, 2017; Burnett et al., 2018). Some of the sources, e.g. fuel wood or agricultural residue burnings are expected to be increased in both domestic and industrial sectors due to their role in decentralised and substitute energy production (Vicente and Alves, 2018). At the same time, the potential disadvantages and risk of these major source types – including BB in particular – have been less recognised (Hays et al., 2003; Chen et al., 2017). It is, therefore, highly relevant to estimate the relative contribution of FF combustion, BB and biogenic sources to major carbonaceous aerosol species, namely to organic carbon (OC) and elemental carbon (EC).

Huge number, composite character, spatial and temporal variability of the sources together with atmospheric transformation of their products make the quantification of the source types or their inventory-based source assessment challenging (Nozière et al., 2015). There are several methods to apportion the particulate matter (PM) mass or carbonaceous species among some or all their major sources. They include 1) source-specific marker methods (Fraser et al., 2000;

Szidat et al., 2006, 2009; Minguillón et al., 2011; Zhang et al., 2012; Bernardoni et al., 2013), 2) multi-wavelength optical methods (Sandradewi et al., 2008a, 2008b; Zotter et al., 2017) and 3) various multivariate statistical methods based on online or offline data (Hopke, 2016; Maenhaut et al., 2016). Recently, the latter, 3rd approach takes also advantage of dedicated molecular tracers/fragments from mass spectrometry or advanced optical techniques (Forello et al., 2019; Stefenelli et al., 2019). The marker methods are beneficial from the point of view that they do not demand many samples or very extensive data sets and that the required analytical data are ordinarily available in related studies. Among the most frequently and successfully adopted markers are radiocarbon (^{14}C, $T_{1/2}$=5730 y), which is used for quantifying FF combustion and levoglucosan (LVG, monosaccharide anhydride $C_6H_{10}O_5$), which is utilised for assessing BB. The monosaccharide anhydride analysis often involves the stereoisomers of LVG, namely mannosan (MAN) and galactosan (GAN) as well in addition to LVG since their concentration ratios can be connected to biomass/wood type (e.g. hardwood or softwood; Fine et al., 2004; Schmidl et al., 2008). Formation, modelling utilisation, atmospheric processes and analytical determinations of these two markers together with their advantages and limitations were described, evaluated and discussed in detail earlier (e.g. Simoneit et al., 1999, 2004; Fraser and Lakshmanan, 2000; Nolte et al., 2001; Pashynska et al., 2002; Zdráhal et al., 2002; Puxbaum et al., 2007; Saarikoski et al., 2008; Caseiro et al., 2009; Fabbri et al., 2009; Szidat et al., 2006, 2009; Favez et al., 2010; Hennigan et al., 2010; Hoffmann et al., 2010; Kourtchev et al., 2011; Piazzalunga et al., 2011; Maenhaut et al., 2012, 2016; Yttri et al., 2014). The coupled radiocarbon-LVG marker method, introduced recently (Salma et al., 2017), is a combination of the two marker methods. It allows to apportion the TC (TC=OC+EC) among all major source types, thus among the contributions of EC and OC from FF combustion (EC_{FF} and OC_{FF}, respectively), EC and OC from BB (EC_{BB} and OC_{BB}, respectively), and OC from biogenic sources (OC_{BIO}).

Water-soluble OC (WSOC) is also an important carbonaceous aerosol species because it is considered as an indicator of secondary organic aerosol (SOA) or carbonaceous particles after atmospheric chemical aging (Claeys et al., 2010). It is related to more oxygenated chemical species than freshly emitted or formed organic constituents, and this class of molecules is expected to contribute substantially to cloud condensation nuclei (CCN) activity of particles and represent potentially larger negative health effects of particulate mass deposited in the human respiratory system due to its solubility (Hallquist et al., 2009; Fuzzi et al., 2015; Nozière et al., 2015).

Despite their overall role together with the health, climate and environmental effects, there are serious gaps in our knowledge on FF combustion, BB and biogenic sources – particularly on biogenic sources in more polluted or urban areas. Information on the properties of the major apportioned or secondary carbonaceous aerosol species and on their relationships with other atmospheric quantities have been missing internationally on extended spatial scales as well as on larger cities. The Carpathian Basin (also known as the Pannonian Basin) is the largest, topographically well separated, orogenic basin in Europe (Salma et al., 2016b). Its land is mostly used for intensive agriculture and farming, while larger forested areas with deciduous, coniferous or mixed wood occur in the inner and bounding mountains. Weather situations within the basin are generally uniform, which makes it advantageous for studying atmospheric phenomena and processes. Budapest with 2.3 million inhabitants in the metropolitan area and with its central geographical location is the largest and principal city in the basin. The mean green space intensity – which indicates the healthy green coverage – for Budapest in 2015 was estimated from Landsat satellite images to be approximately 50% with spatial variations from 19% in the city centre to 55% in the suburban zone (Tatai et al., 2017).

As part of a research project, we collected aerosol samples in the regional background atmospheric environment of the Carpathian Basin, suburban area and city centre of Budapest in each season for 1-year-long time interval and analysed them for various aerosol constituents, which are required for source apportionment. The analytical results were complemented by supporting air pollutant and meteorological data as well. The major objectives of the present paper are to report the main findings of this research, to discuss the properties and contributions of FF combustion, BB and biogenic sources and related atmospheric processes, to interpret the relationships among various variables for different months and environmental types, and to formulate some general conclusions on air quality of the region and the city.

2 Methods

2.1 Collection of aerosol samples and in-situ measurements

The aerosol samples were collected at three sites in Hungary, in a rural background area and at two urban sites in Budapest (Fig. 1). The samplings at the rural location were realised at K-puszta station (N 46° 57' 56", E 19° 32' 42", 125 m above mean sea level, a.s.l.), which is situated on the Great Hungarian Plain in a clearing within a mixed forest of coniferous (60%)

and deciduous trees (30%) and some grassland (Salma et al., 2016b). The nearest city of Kecskemét (with 110 thousand inhabitants) is situated ca. 15 km to the SE of K-puszta. The station is part of the European monitoring and evaluation of the long-range transmission of air pollutants programme (EMEP network) and represents the largest part (regional background) of the Carpathian Basin. One of the urban sites was at an open suburban area of residential Budapest at the Marczell György Main Observatory (N 47° 25' 46", E 19°10' 54", 138 m a.s.l.) of the Hungarian Meteorological Service. The collections at the other urban location were performed at the Budapest platform for Aerosol Research and Training (BpART) Laboratory (N 47° 28' 30", E 19° 03' 45", 115 m a.s.l.) of the Eötvös University. The latter site is situated on the bank of the Danube and represents a well-mixed average atmosphere of the city centre (Salma et al., 2016a). Some further details of the sampling campaign are summarised in Table 1.

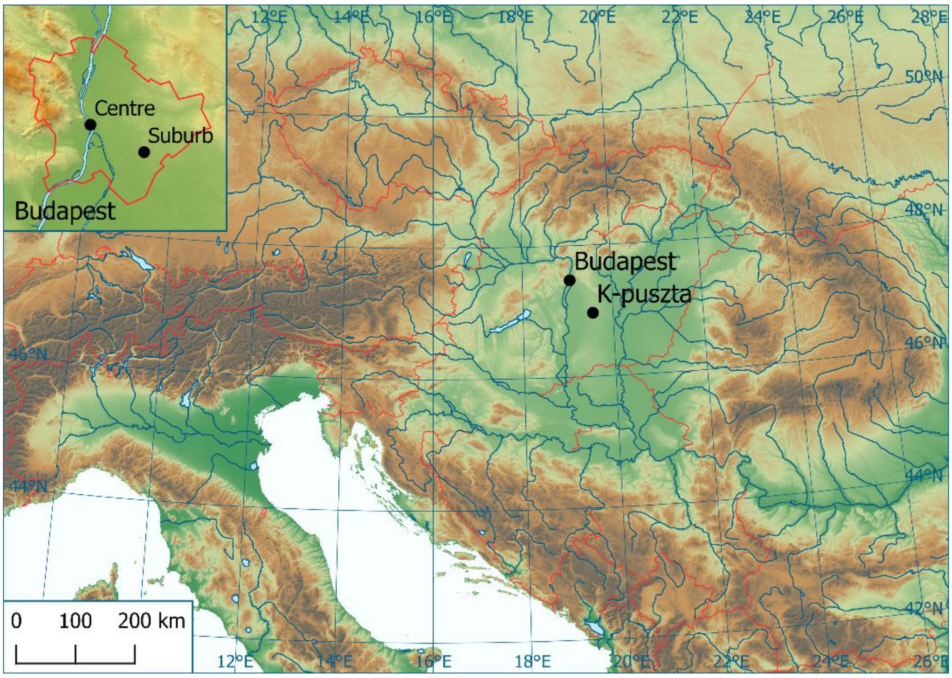

Figure 1. The Carpathian Basin with location of the sampling sites in Budapest (city centre and suburban area) and at K-puszta station (regional background).

The aerosol sampling was realised by three identical high-volume DHA-80 devices equipped with $PM_{2.5}$ inlets (Digitel, Switzerland). The collection substrates were quartz fibre filters with a diameter of 150 mm (QR-100, Advantec, Japan). Daily aerosol samples were taken starting at 00:00 LT (LT=UTC+1 or daylight-saving time UTC+2). The sampled air volumes were ca.

720 m^3. One field blank sample was also taken at each site and month. All filters were pre-heated at 500 °C for 24 h before the exposure and were stored in a freezer after the collections.

Table 1. Start and end dates of the sampling periods and number of aerosol samples collected in regional background of the Carpathian Basin, suburban area and city centre of Budapest.

Site type	Time interval: month year	Autumn month: October 2017	Winter month: January 2018	Spring month: April 2018	Summer month: July 2018
Region	Days	18–31	09–22	17–30	17–30
	Samples	14	14	14	14
Suburb	Days	18–31	06–22	17–30	17–01*
	Samples	14	17	14	14
Centre	Days	18–27	10–16	17–23	17–23
	Samples	7	7	7	7

* 01 August 2018.

Concentrations of criteria air pollutants, i.e. SO_2, NO/NO_x, CO, O_3 and PM_{10} mass were obtained from regular stations of the National Air Quality Network. For the regional background and suburban area, they were measured directly at the sampling sites, while for the city centre, the pollutants were recorded in a distance of 4.5 km in the upwind prevailing direction from the sampling site. The concentrations are measured by UV fluorescence (Ysselbach 43C), chemiluminescence (Thermo 42C), IR absorption (Thermo 48i), UV absorption (Ysselbach 49C) and beta-ray attenuation methods (Thermo FH62-I-R), respectively with a time resolution of 1 h. Local meteorological data including air temperature (*T*) and relative humidity (RH), wind speed (WS) and global solar electromagnetic radiation (GRad) were acquired by standardised meteorological methods (Vaisala HMP45D humidity and temperature probe, Vaisala WAV15A anemometer, both Finland and CMP3 pyranometer, Kipp & Zonen, The Netherlands) near the sampling sites with a time resolution of 10 min. According to our knowledge, there were no extensive agricultural burns or wildfires in the basin during to the actual sampling time intervals, and the BB in the basin is expected to be dominated by biofuel utilisation.

2.2 Analysis of aerosol samples

The PM mass was determined by weighing each filter before and after the sampling on a microbalance with a sensitivity of 10 μg (Cubis MSA225S-000-DA, Sartorius, Germany). The exposed and blank filters were pre-equilibrated before weighing at a *T* of 19–21°C and RH of 45–50% for at least 48 hours. The measured mass data for the exposed filters were corrected for the field blank values considering the uncertainties in weighing, sampled air volume, RH and some other environmental conditions during the weighing as well. The PM mass data were above the limit of quantitation (LOQ), which was approximately 1 μg m^{-3}.

One or two punches with an area of 1.5 cm^2 each of the filters were directly analysed by thermal-optical transmission (TOT) method (Birch and Cary, 1996) using a laboratory OC/EC analyser (Sunset Laboratory, USA) adopting the EUSAAR2 thermal protocol. The measured OC data for the exposed filters were corrected for the field blank values, while the EC on the blanks was negligible. All measured OC and EC data were above the LOQ, which was 0.38 and 0.04 μg m^{-3}, respectively.

One or two sections with an area of 2.5 cm^2 each of the filters were extracted in water, the extracts were filtered, and the filtrates were analysed for WSOC by a Vario TOC cube analyser (Elementar, Germany) in three repetitions with an injected volume of 1 ml each. The measured WSOC data for the exposed filters were corrected for the field blank values. All measured WSOC data were above the LOQ, which was ca. 0.08 μg m^{-3}.

A section with an area of 2 cm^2 of each filter was analysed for LVG, MAN and GAN by gas chromatography/mass spectrometry (GC/MS) after trimethylsilylation (Blumberger et al., 2019). The filter sections were extracted repeatedly by dichloromethane-methanol in an ultrasonic bath. The extracts were filtered and spiked with an internal standard (IS) of methyl β-L-arabinopyranoside. The trimethylsilylation was realised by hexamethyldisilazane as silylating agent, pyridine as solvent and trifluoroacetic acid as catalyst at 70 °C. The prepared samples were analysed by a Varian 4000 GC-MS/MS system (USA) with a GC/MS column of SGE forte BPX-5 capillary (length×inner diameter 15 m×0.25 mm; film thickness 0.25 µm, SGE, Australia). The quantification was carried out in the selected ion monitoring mode by quantifier ions with mass-to-charge ratios of m/z=204 for LVG and of 217 for MAN, GAN and IS. The LVG data for the exposed filters were corrected for blank values; while MAN and

GAN were not detected in the blanks. The LVG amount in the blank filters can be related to the sampling itself, to chemicals used, to various chemical and sample preparation procedures performed and to the variation of the baseline of the measurement (Maenhaut et al., 2012). The blanks were the largest with respect to the corrected values in the summer samples, in which the measured LVG amounts were approximately ten times larger than in the blank filters. In all the other samples, the relative contributions of the blanks were even smaller than this. The LOQ for LVG and MAN was approximately 1.2 ng m^{-3}, while it was approximately 0.5 ng m^{-3} for GAN. All LVG data were above the LOQ, while the MAN and GAN could not be quantified in the summer samples.

Filters collected in parallel on seven overlapping days, i.e. on 18–21, 25 and 26 October, on 10–16 January, on 17–23 April and on 17–23 July were subjected to C isotope analysis of the TC content by accelerator mass spectrometry (AMS) with an off-line combustion system (Molnár et al., 2013; Janovics et al., 2018). Carbonaceous aerosol species on eighth section of each filter were oxidised quantitatively to CO_2 gas (Major et al., 2018). This was later introduced into an IonPlus Enviro Mini Carbon Dating System spectrometer (Switzerland) via its dedicated gas ion source interface. The measured results for the exposed filters were corrected for the blank values. The $^{14}C/^{12}C$ ratios were also corrected for isotopic fractionation by using the $^{13}C/^{12}C$ ratios (Wacker et al., 2010) that were obtained simultaneously in the actual AMS measurements. The $^{14}C/^{12}C$ isotope ratios derived were also normalised to that of the oxalic acid II 4990C standard reference material (NIST, USA), and the measurement results were expressed as fraction of modern carbon (f_m), which denotes the $^{14}C/^{12}C$ ratio of the samples relative to that of the unperturbed atmosphere in the reference year of 1950 (Burr and Jull, 2009). Since majority of currently combusted fuel wood was growing after the atmospheric nuclear fusion bomb tests in the late 1950s and early 1960s, the samples were also corrected by a mean factor of 1.08 derived for the Northern Hemisphere (Szidat et al., 2009; Heal et al., 2011). Thus, the fraction of contemporary carbon (f_c) was calculated as $f_c=f_m/1.08$. The same correction factor was also adopted for the TC from biogenic sources, although it is expected to show a somewhat smaller ^{14}C abundance. The differences in the f_c caused by the refined correction factor are ordinarily small when compared to the method uncertainties (Minguillón et al., 2011) and, therefore, this effect was neglected.

A quarter section of each filter was utilized to determine the K (as a possible inorganic tracer for BB), Ni (as a possible tracer for residual oil combustion) and Pb (as a former tracer for vehicles with gasoline engine) content of the aerosol samples by inductively coupled plasma optical emission spectrometry using an iCAP7400 DUO instrument (Thermo Fischer Scientific, Germany). The filter sections were extracted by microwave-assisted HNO_3–H_2O_2 digestion. The analytical results for the exposed filters were corrected for the blank values. The LOQ values of the elements listed were approximately 0.02 μg m^{-3}, 0.4 and 0.5 ng m^{-3}, respectively, and most atmospheric concentration were above them.

2.3 Data evaluation and modelling

Concentrations of organic matter (OM) were derived from the OC data by multiplying them with an organic aerosol-to-organic carbon mass conversion factor. This factor is an estimate of the average molecular mass per C atom for OM in general. It is site-dependent and can have seasonal and diurnal variations as well. It is usually derived by indirect considerations (Russell, 2003). Mass conversion factors between 1.2 and 1.4 were estimated for fine atmospheric aerosol in mildly oxidizing atmospheric environments (Turpin et al., 2000). Some further studies suggest that a factor of 1.6±0.2 describes better the oxidizing urban environments or chemically aged (long-range transported) aerosol (Turpin and Lim, 2001). It should also be noted that the conversion factor is one of the most substantial sources of uncertainty in aerosol chemical mass closure calculations. It was estimated that the relative uncertainty associated with the conversion is approximately 30% (Maenhaut et al., 2012). In the present study, a factor of 1.4 was adopted for the regional and suburban environments (considering that local or regional sources are mostly substantial/dominating in the Carpathian Basin) and a factor of 1.6 was utilised for the city centre.

The comparisons of atmospheric concentrations, other variables or their ratios with respect to the months or sites were accomplished by calculating first the ratios on a sample-by-sample or day-by-day basis and then by averaging these individual ratios for the subset under consideration.

The coupled radiocarbon-LVG marker method was utilised to apportion the TC among the EC_{FF}, OC_{FF}, EC_{BB}, OC_{BB} and OC_{BIO} (Salma et al., 2017). The method consists of pragmatic attribution steps, which are realised by multiplications with apportionment factors. The factors are calculated for each sample from measured TC, f_c, EC, OC and LVG concentrations as primary input data and from general, a priori known EC/OC ratio for BB [$(EC/OC)_{BB}$] and OC/LVG ratio for BB [$(OC/LVG)_{BB}$]. Their combined adaptation is related to subsequent and step wise subtraction of contemporary TC, EC_{BB} and OC_{BB} from TC on the one hand, and of EC_{FF} from fossil TC on the other hand. The apportionment factors are expressed as: $f_1=f_c$, $f_2=(OC/LVG)_{BB}\times LVG\times(EC/OC)_{BB}/f_1/TC$, $f_3=(OC/LVG)_{BB}\times LVG/f_1/(1-f_2)/TC$ and $f_4=(EC/TC-f_1\times f_2)/(1-f_1)$ (Salma et al., 2017). For the $(EC/OC)_{BB}$ ratio, we implemented a mean of 17% derived from a critically evaluated ratio and standard deviation (SD) of (16±5)% (Szidat et al., 2006) and from a ratio and SD of (18±4)% (Bernardoni et al., 2011, 2013) obtained specifically for wood burning. As far as the $(OC/LVG)_{BB}$ ratio is concerned, its actual value depends predominantly on the wood type and burning conditions (Puxbaum et al., 2007). We adopted an $(OC/LVG)_{BB}$ ratio of 5.59 (Schmidl et al., 2008). The mean apportionment factors separately for the different months and site types are summarised in Table S1 in the Supplement. It is the OC_{BIO} and OC_{BB} which are the most sensitively influenced by the input uncertainties. Their relative uncertainty for some individual low concentrations could be up to 40–50%, while it is expected to be approximately 30% or smaller for the other carbonaceous species.

3 Results and discussion

The results of the study are interpreted in a conservative manner thus with regard to the months, but they are most likely representative with respect to the seasons as well. This expectation is based on the favourable meteorological conditions during the sample collections and the basin character of the region. General relationships that can exist among the months and atmospheric environments including coupled meteorological and chemical processes need to be overviewed before evaluating the temporal and spatial variability and tendencies in aerosol properties.

3.1 Differences and similarities among months and atmospheric environments

The median concentrations of SO_2, NO, NO_2 and PM_{10} mass over the sampling time intervals were larger in the city centre than in the suburban area in all months (Table S2 in the Supplement). This can mainly be explained by their anthropogenic sources in the city centre, mostly due to the increased intensity and density of road traffic. In contrast, the O_3 level was substantially higher in the suburban area than in the city centre and considerably larger in the regional background that in the suburban area. It tended to show a maximum in July. Such a behaviour is typical for large-scale O_3 formation mechanism. This all suggests that there could be substantial differences in photochemical activity in general as indicated by O_3 between the regional background and the urban sites except for July. To access the extent of atmospheric dynamics during the sampling campaigns, the mean values and SDs of the same variables are shown in Table S3 as auxiliary data. The variability of the concentrations partially supports the conclusions on the importance of regional meteorology within the basin (Figs. 2–4).

The meteorological data over the sampling time intervals are in accordance with ordinary monthly mean values and denote weather situations without extremes (Table S4 in the Supplement). The *T* data indicate an urban heat island in central Budapest, particularly in winter and autumn. At the regional site, there was snow cover with a thickness from 2 to 4 cm during the sample collections in January for approximately 4 days, while in the Budapest area, there was snow in spots with a thickness of 1–2 cm for 2–3 days. The data suggest that there was somewhat milder weather over the sample collections in January than usually present.

Time series of $PM_{2.5}$ mass, EC and WSOC over the sampling time intervals separately in the different months and environments are shown in Figs. 2, 3 and 4, respectively. The $PM_{2.5}$ mass represents the bulk fine PM; EC is a typical primary aerosol constituent, while WSOC expresses mainly SOA and partially BB products (which also exhibit substantial hygroscopicity and thus, water-solubility; Swietlicki et al., 2008). These species are rather different as far as their sources are concerned. Nevertheless, their atmospheric concentrations often changed coherently at the locations with their strongest link in winter. It can likely be explained by the common effects of regional meteorology on atmospheric concentrations especially under anticyclonic weather situations – particularly of boundary layer mixing height – over the Carpathian Basin. It seems that the daily evolution of regional meteorology often has higher influence on the changes in concentrations than the source intensities if the sources are distributed across a large area (Salma et al., 2001, 2004). The strongest connection is related to cold air masses above the Carpathian Basin which generate a lasting *T* inversion layer (the so-called cold pillow) and which restricts the vertical mixing and results in poor air quality over extended areas of the basin in larger and smaller cities as well as in rural areas.

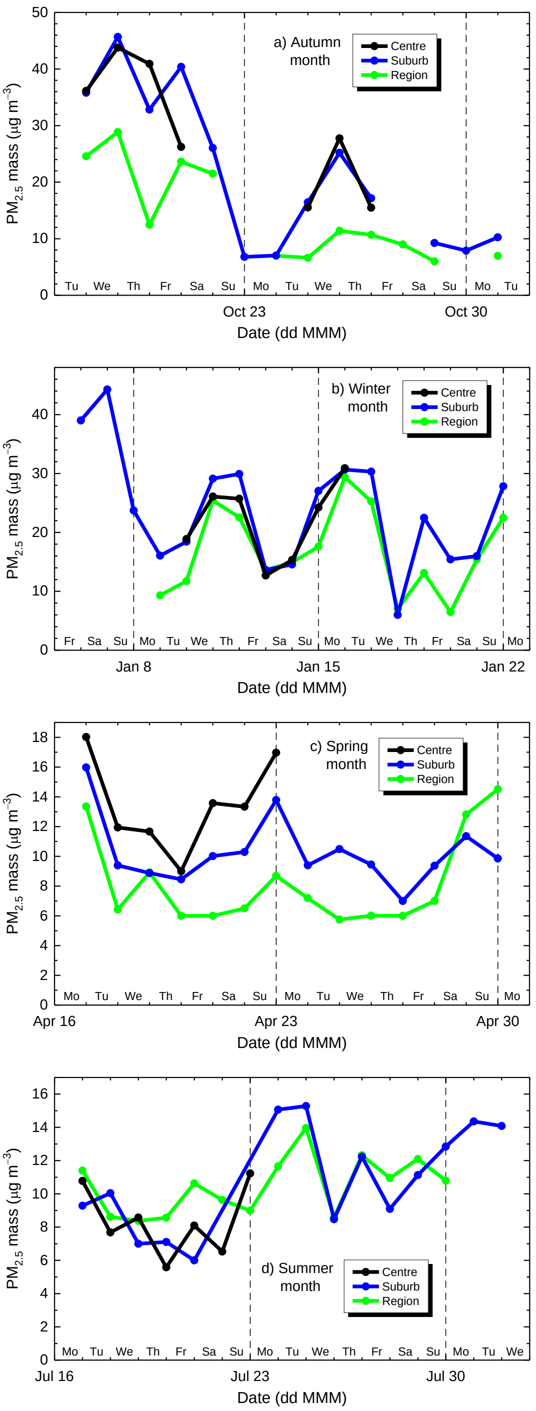

Figure 2. Time variation of $PM_{2.5}$ mass for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

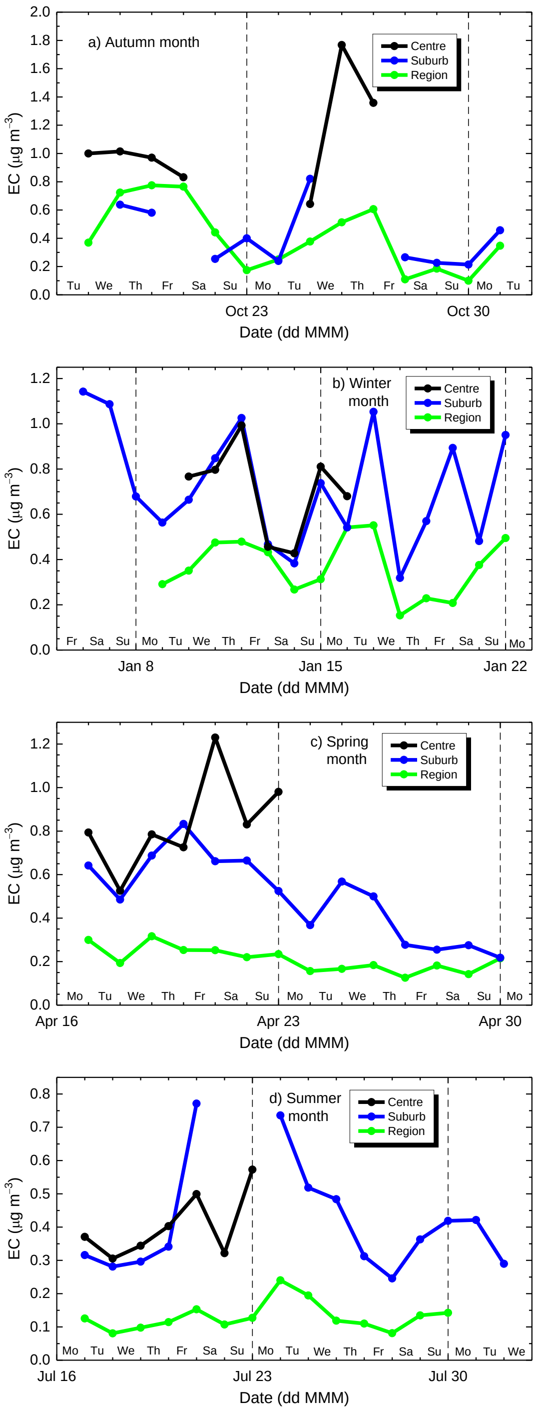

Figure 3. Time variation of EC for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

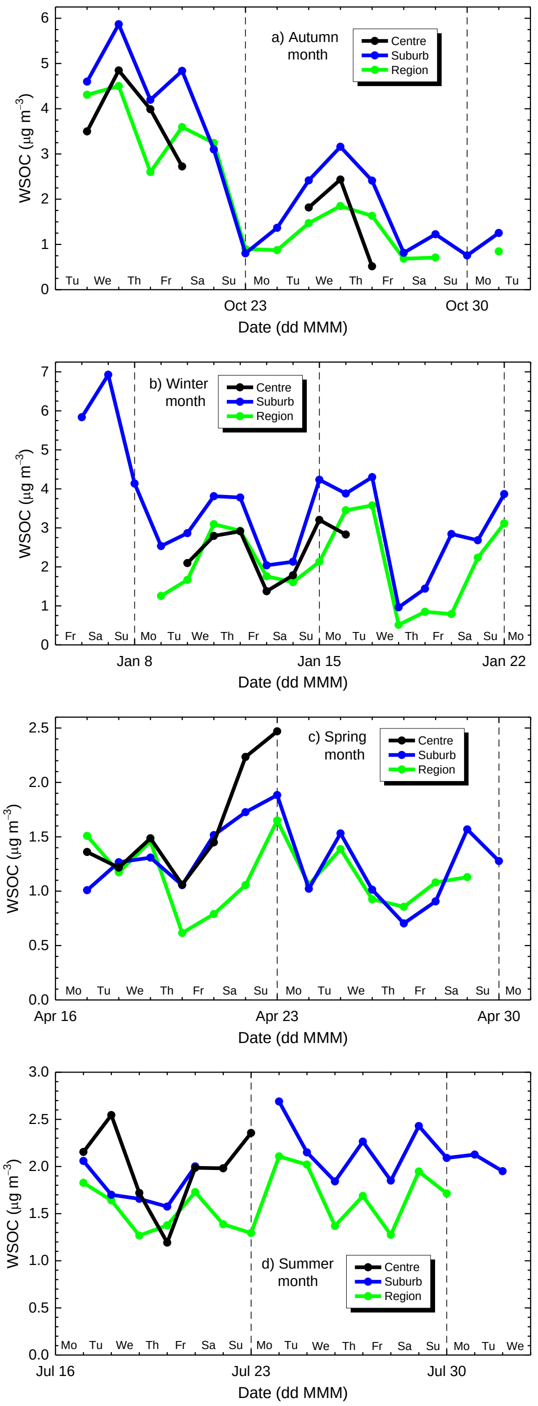

Figure 4. Time variation of WSOC for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

3.2 Tendencies in aerosol concentrations

Median atmospheric concentrations of the measured aerosol constituents separately for the different months and environmental types are presented in Table 2, while their means and SDs are shown in Table S5 in the Supplement. The concentrations are in line with or somewhat smaller than the corresponding results obtained in earlier studies at the same or similar locations usually for shorter time intervals (Salma and Maenhaut, 2006; Kiss et al., 2002; Salma et al., 2004, 2007, 2013, 2017; Ion et al., 2005; Maenhaut et al., 2005, 2008; Puxbaum et al., 2007; Blumberger et al., 2019). The $PM_{2.5}$ mass and OC concentrations in the city centre were larger by a mean factor of 1.6–1.7 than in the regional background, while they were similar to the suburban data. Their values in January were usually the largest, and they reached the minimum in July or April.

The concentrations of EC increased monotonically in the order of the environments: regional background, suburban area and city centre location by typical factors of 2 and 3, respectively. In the regional background, the EC data for October and January were similar to each other and they were the largest. In the suburban area, the EC data showed a maximum in January and a minimum in July. In the city centre, the EC levels in October, January and April were similar to each other, and they all showed a minimum in July. These can be explained by larger intensity of soot emissions from incomplete burning (road vehicles, residential heating and cooking by solid fuel), which is typically of anthropogenic origin. These sources can have either seasonal variability (e.g. residential heating) or constant intensity (traffic or cooking) over a yearly time span.

The WSOC showed maximum medians in January at all sites. In October and July, the urban locations had similar concentrations to each other, while it was somewhat smaller in the regional background. In January, the suburban site exhibited the maximum median concentration. This is explained by larger influence of BB in this environment and month and by higher water solubility of its products (see Sects. 3.3 and 3.5). In April, the medians had a monotonically increasing tendency from the regional background to the city centre.

Table 2. Median atmospheric concentrations of $PM_{2.5}$ mass, elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC), levoglucosan (LVG), mannosan (MAN), galactosan (GAN), fraction of contemporary total carbon (f_c), median concentrations of K, Ni and Pb for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Constituent	Site type	October	January	April	July
$PM_{2.5}$ mass	Region	12.5	16.5	8.6	10.7
(µg m^{-3})	Suburb	25	26	9.7	11.7
	Centre	28	24	13.3	8.2
EC	Region	0.37	0.36	0.20	0.122
(µg m^{-3})	Suburb	0.45	0.68	0.51	0.35
	Centre	0.99	0.77	0.79	0.37
OC	Region	2.3	3.2	2.0	2.2
(µg m^{-3})	Suburb	4.5	5.4	2.4	2.7
	Centre	6.6	4.6	2.8	2.6
WSOC	Region	1.63	2.0	1.08	1.66
(µg m^{-3})	Suburb	2.4	3.8	1.27	2.0
	Centre	2.7	2.8	1.45	2.0
LVG	Region	0.172	0.40	0.0180	0.0081
(µg m^{-3})	Suburb	0.44	0.71	0.040	0.0124
	Centre	0.38	0.48	0.036	0.0103
MAN	Region	19.2	18.5	2.6	<1.2
(ng m^{-3})	Suburb	37	39	2.9	<1.2
	Centre	26	21	4.1	<1.2
GAN	Region	n.a.	10.6	1.20	<0.5
(ng m^{-3})	Suburb	16.4	20	0.61	<0.5
	Centre	11.7	14.1	1.21	<0.5
f_c	Region	69	75	61	74
(%)	Suburb	66	74	48	60
	Centre	76	74	48	60
K	Region	0.182	0.23	0.088	0.081
(µg m^{-3})	Suburb	0.22	0.25	0.097	0.075
	Centre	0.26	0.27	0.106	0.057
Ni	Region	0.75	0.68	1.21	1.12
(ng m^{-3})	Suburb	0.88	0.78	1.24	1.09
	Centre	1.10	0.63	1.51	1.08
Pb	Region	3.8	3.0	3.2	2.8
(ng m^{-3})	Suburb	5.8	6.8	4.1	3.5
	Centre	7.5	5.2	4.4	2.3

n.a.: not available

The mean atmospheric concentrations of the monosaccharide anhydrides were decreasing in the order of LVG, MAN and GAN. The concentrations of LVG were larger by ca. 1 order of magnitude than for the joint concentrations of MAN and GAN. Their mean ratio was the largest in January and the smallest in October. This could be affected by the share of hardwood burnt in different months (Fine et al., 2004; Schmidl et al., 2008; Maenhaut et al., 2012). The LVG concentration did not vary monotonically with respect to the sites; it was larger in the city centre by a factor of 1.7 than in the regional background and was smaller by approximately 20% than in the suburban area. This could be related to the spatial distribution of biofuel utilisation mainly for residential heating and to atmospheric dispersion of their emission products in the different environments.

As far as the contemporary C is concerned, there were three individual consecutive samples collected in the city centre in October with significantly larger values than any other data in the set. There are several applications of nuclide ^{14}C mostly in pharmaceutical/medical and biological academy field in Budapest. They could release radiocarbon of anthropogenic origin into the ambient air (in particular from labelled inorganic compounds such as $NaHCO_3$). These three data were regarded to be outliers and were excluded from the further evaluation. The centre/suburb f_c ratio in October, however, remained still somewhat higher (1.15) with respect to the other months (for which the ratios were uniformly 1.00). This indicates that the anthropogenic ^{14}C contamination could slightly affect the remaining analytical results as well. Its consequences on the source apportionment are discussed in Sect. 3.5. In October and January, the mean centre/suburb, centre/region and suburb/region ratios were similar to each other (with an overall mean and SD of 1.02±0.10) at all sites, while in April and July; they decreased in the order of the ratios above with means and SDs of 1.04±0.20, 0.82±0.13 and 0.80±0.15, respectively. These tendencies are governed by carbonaceous matter of different origin.

The concentrations of K in October, January and April increased monotonically for the regional background, suburb area and city centre. They showed a maximum in January. Its concentrations were the smallest in July and exhibited an opposite tendency as far as the location types are concerned, thus they decreased monotonically for the sites listed above. The concentrations of Ni were similar to each other without any evident tendency. Except for its concentrations in April, which seemed to be the largest. The concentration of Pb showed an increasing tendency from the regional background to the urban sites. The present data are smaller than the median levels of 16 ng m^{-3} in the city centre and of 9 ng m^{-3} in the near-city background measured in spring 2002 after the phase out of leaded gasoline in Hungary in April 1999, and are in line with its overall decreasing trend (Salma and Maenhaut, 2006; Salma et al., 2000).

3.3 Tendencies in concentration ratios

Mean values and SDs of some important concentration ratios separately for the months and different environments are shown in Table 3. The $PM_{2.5}/PM_{10}$ mass ratio exhibited strong time dependency. In April and July, the PM_{10}–$PM_{2.5}$ fraction particles (coarse mode) made up approximately 2/3 of the particulate mass, while in October and perhaps also in January, the $PM_{2.5}$ mass prevailed with a similar ratio. These imply and confirm that in spring and summer, the suspension or resuspension of soil, crustal rock, mineral and roadside dust is substantial in Budapest, while in autumn and winter, the aerosol mass levels are more influenced by residential heating, cooking and road traffic (Salma and Maenhaut, 2006).

Table 3. Mean values and SDs for the $PM_{2.5}/PM_{10}$ mass, $OM/PM_{2.5}$ mass, $EC/PM_{2.5}$ mass, $TC/PM_{2.5}$ mass, WSOC/OC and OC/EC ratios for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Ratio	Site type	October	January	April	July
$PM_{2.5}/PM_{10}$ mass (%)	Region	64±10	n.a.	n.a.	n.a.
	Suburb	64±4	67±11	30±8	48±8
	Centre	67±9	56±14	32±4	33±7
$OM/PM_{2.5}$ mass (%)	Region	33±6	29±5	32±5	33±4
	Suburb	32±9	31±5	32±6	32±5
	Centre	36±5	30±3	30±4	36±5
$EC/PM_{2.5}$ mass (%)	Region	3.1±1.4	2.3±0.5	2.3±0.7	1.2±0.3
	Suburb	3.2±1.0	3.1±0.9	4.9±2.2	3.5±1.1
	Centre	4.3±2.4	3.3±0.6	6.4±1.7	4.6±0.9
$TC/PM_{2.5}$ mass (%)	Region	28±5	22±4	25±4	23±3
	Suburb	24±7	26±4	27±5	26±4
	Centre	27±5	22±3	28±5	29±10
WSOC/OC (%)	Region	64±11	58±7	54±9	72±5
	Suburb	55±16	64±6	53±7	76±6
	Centre	42±16	59±4	56±9	76±11
OC/EC	Region	8.8±3.2	9.0±1.4	11±3	18±4
	Suburb	8.4±3.5	7.3±1.5	5.4±2.0	7.3±1.6
	Centre	6.3±2.2	5.9±0.7	3.5±0.7	6.7±1.5

Contribution of the OM to the $PM_{2.5}$ mass for the regional background, suburban area and city centre showed little time variation with annual means and SDs of (31±5)%, (32±6)% and (35±7)%, respectively in particular if we consider the uncertainty related to the OM/OC conversion (Sect. 2.3). These balanced contributions are in line with other European results (Puxbaum et al., 2007; Putaud et al., 2010). The mean contributions of EC to the $PM_{2.5}$ mass were between 1 and 6%, with a minimum in the regional background in July. The contributions can change substantially in different microenvironments within a city (e.g. 14% for a street canyon in central Budapest in spring; Salma et al., 2004; Maenhaut et al., 2005). The carbonaceous particles (OM+EC) in the regional background, suburban area and city centre made up (32±5)%, (36±7)% and (39±7)%, respectively of the $PM_{2.5}$ mass as annual means and SDs. Their means revealed limited variability (except for the city centre, where it changed from 33% in January to 48% in July). The TC/$PM_{2.5}$ mass ratios are given as auxiliary information to allow the recalculation of the contributions to the TC shown in Fig. 5 to that to the $PM_{2.5}$ mass.

The mean WSOC/OC ratios in October showed a monotonically increasing tendency from the city centre to the regional background. This is just opposite to the atmospheric WSOC concentration (which decreased monotonically). In January, the suburban area exhibited the maximum concentration. This can be explained by intensive BB in the area with respect to the other environments (Sect. 3.5) and with the fact that BB particles possess relatively high hygroscopicity (Swietlicki et al., 2008) and water solubility. In the remaining two months, the shares of the WSOC were similar to each other and varied without an obvious tendency. This can be linked to comparable and large photochemical activity in all environments in April and July (Sect. 3.1). The present ratios are in line with the values reported earlier for the corresponding locations (Kiss et al., 2002; Ion et al., 2005; Maenhaut et al., 2005, 2008; Viana et al., 2006; Puxbaum et al., 2007; Salma et al., 2007). It is noted that the determined OC (and WSOC) concentrations are somewhat method dependent; their ratios can change sensitively e.g. with the thermal protocol used in the OC/EC TOT analyser for samples containing large amounts of refractory C (Kuhlbusch et al., 2009; Pantheliadis et al., 2015).

The highest OC/EC ratios are often linked to atmospheric conditions under which the SOA formation is large. The ratio had a maximum in the regional background in July, which can be associated with large photochemical activity and strong GRad. The ratios for the urban locations did not indicate obvious time tendencies. Formation, composition and properties of

SOA and atmospheric humic-like substances (HULIS) together with modelling the air mass transport within the Carpathian Basin are to be dealt with in a separate paper after additional investigations are completed.

Finally, it is noted for completeness that the annual mean LVG/MAN ratios and SDs for the regional background, suburban area and city centre were 13.9±5.9, 14.3±6.2 and 14.7±5.8, respectively, and that ca. 40% of all available individual ratios were larger than the limit of 14.8 derived by Schmidl et al. (2008). The latter value was obtained for the combustion of common hardwood (beech and oak) and softwood species (spruce and larch) in domestic wood stoves in Austria. This means for our samples and conditions, the relationship between the softwood and hardwood burnt mentioned is not applicable because of several reasons, e.g. the likely differences in fireplaces and fuel wood in Hungary and mid-European Alpine regions.

3.4 Apportioned carbonaceous species

Median atmospheric concentrations of the apportioned EC_{FF}, EC_{BB}, OC_{FF}, OC_{BB} and OC_{BIO} aerosol constituents derived by the coupled radiocarbon-LVG model separately for the different months and environments are summarised in Table 4. The present values are coherent with the earlier median concentration from late winter/early spring of 2014 at the BpART Laboratory (Salma et al., 2017) and comparable to results for the regional background (Gelencsér et al., 2007; Puxbaum et al., 2007). A more sensible evaluation is to compare the contributions of the apportioned species to TC with other similar studies, which is completed in Sect. 3.5. The uncertainty of the individual apportioned data is larger than for the experimental results (e.g. TC) and, therefore, the substantial differences among their means and their obvious tendencies are only interpreted.

The median concentrations of EC_{FF} were similar to each other in October, April and perhaps in July as well and exhibited a minimum in January. In all months, its concentrations in the urban environments tended to be larger by a factor of 2–3 than in the regional background. The OC_{FF} concentrations at the urban locations were similar to each other in all months, while they tended to be larger than the regional values by a factor of 2–3 in October and July. The EC_{BB} and OC_{BB} concentrations showed a maximum in January and a minimum in July. The concentrations of OC_{BB} in the city centre seemed to be somewhat smaller than in the suburban area, while the latter was larger by a factor of 2–3 than in the regional background in October and April. The concentrations of OC_{BIO} showed a monotonically decreasing tendency from

October to July, April and January in all environments. The fluxes of biogenic VOCs (BVOCs) from plants strongly depend on environmental conditions, age of leaves and vegetation, water and nutrient availability, and it is also affected by the presence of some anthropogenic emissions. Photochemical oxidation reactions of BVOCs, interactions among biogenic and anthropogenic precursors and products, and aerosol formation yield considerations play a rather important role in the process (McFiggans et al., 2019). The tendencies are further discussed after deriving the contributions of the apportioned species to various quantities in Sect. 3.5.

Table 4. Median atmospheric concentration of apportioned elemental carbon from fossil fuel combustion (EC_{FF}) and from biomass burning (EC_{BB}), of apportioned organic carbon from fossil fuel combustion (OC_{FF}), from biomass burning (OC_{BB}) and from biogenic sources (OC_{BIO}) in µg m^{-3} for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Constituent	Site type	October	January	April	July
EC_{FF}	Region	0.35	0.057	0.23	0.12
	Suburb	0.35	0.10	0.57	0.32
	Centre	0.60	0.24	0.74	0.36
EC_{BB}	Region	0.19	0.34	0.020	0.0076
	Suburb	0.40	0.62	0.050	0.0083
	Centre	0.36	0.46	0.047	0.0095
OC_{FF}	Region	0.85	1.0	0.71	0.53
	Suburb	2.1	1.1	1.0	0.83
	Centre	1.5	1.2	1.0	0.81
OC_{BB}	Region	1.1	2.0	0.12	0.045
	Suburb	2.4	3.6	0.29	0.049
	Centre	2.1	2.7	0.27	0.056
OC_{BIO}	Region	2.0	0.22	1.3	1.8
	Suburb	2.3	0.36	1.2	1.8
	Centre	3.1	0.31	1.3	1.6

Pearson’s coefficients of correlation between the variables were calculated to examine their possible paired relationships. The results should be interpreted with caution since many data sets are not (fully) independent from each other and can be biased by meteorological processes (Sect. 3.1), can be coupled by their potential common sources or can be influenced jointly by further factors/causes for them. Moreover, interactions among biogenic and anthropogenic VOCs or among organic precursors with rather different SOA yields can significantly enhance or suppress, respectively the SOA production (Hoyle et al., 2011; McFiggans et al., 2019). Selected coefficients are shown in Table S6 in the Supplement. Potassium correlated with both carbonaceous species of BB origin at all locations, while its coefficients with the other variables seemed insignificant (at a level of p>0.8). There was a linear relationship between NO (which is emitted in 60–70% by road vehicles in Budapest) and OC_{FF} only in the suburban area. The relationships between T and the apportioned constituents indicated that BB was more intensive under cold weather conditions, while the utilisation of FFs was more constant over the year (campaign). No obvious consistent pattern was observed for FF carbonaceous species (and their contribution to the TC; see section 3.5), which can suggest that domestic heating is a minor source of OC_{FF} compared for instance to vehicular road traffic.

3.5 Contributions of source types

Fossil fuel combustion showed the most balanced and constant daily or monthly mean contributions to the TC at all sites and over the whole year. Its annual means and SDs for the regional background, suburban area and city centre were (31±7)%, (36±12)% and (36±13)%, respectively. In contrast, the daily mean contributions of BB and biogenic sources changed radically over the year at all locations. For BB, the individual contributions for the atmospheric environments listed above ranged from <2 to 73% (with a median of 10%), from <2 to 73% (24%) and from <2 to 72% (19%), respectively. The analogous daily data for biogenic sources spanned from <2 up to 88% (52%), from <2 to 70% (35%) and from <2 to 67% (39%), respectively.

The monthly mean contributions of various EC and OC species to the TC separately for the different environmental types are shown in Figure 5 as circle chart diagrams. In October, the three major source types contributed equally to the TC. In January, it was the BB which was the major source with a relative share of approximately 60% at all sites, and its contribution was the largest in this month. The contributions of FF combustion in January were similar to each other for all sites with a typical share around 25%. The contributions of biogenic sources were the smallest in this month, although their firm interpretation is limited by the relatively large uncertainties. Their share might be linked to larger temperatures (urban heat island) and less snow coverage in the city centre (Sect. 3.1) than in its surroundings. In April, FF combustion and biogenic sources were the largest two contributors at all locations with typical shares of 45–50% each. The EC_{FF} showed the largest contributions in April, which were increased monotonically in the order of the location type: region, suburb and centre. In July, biogenic sources became the major contributor with a monotonically increasing share from the centre to the region.

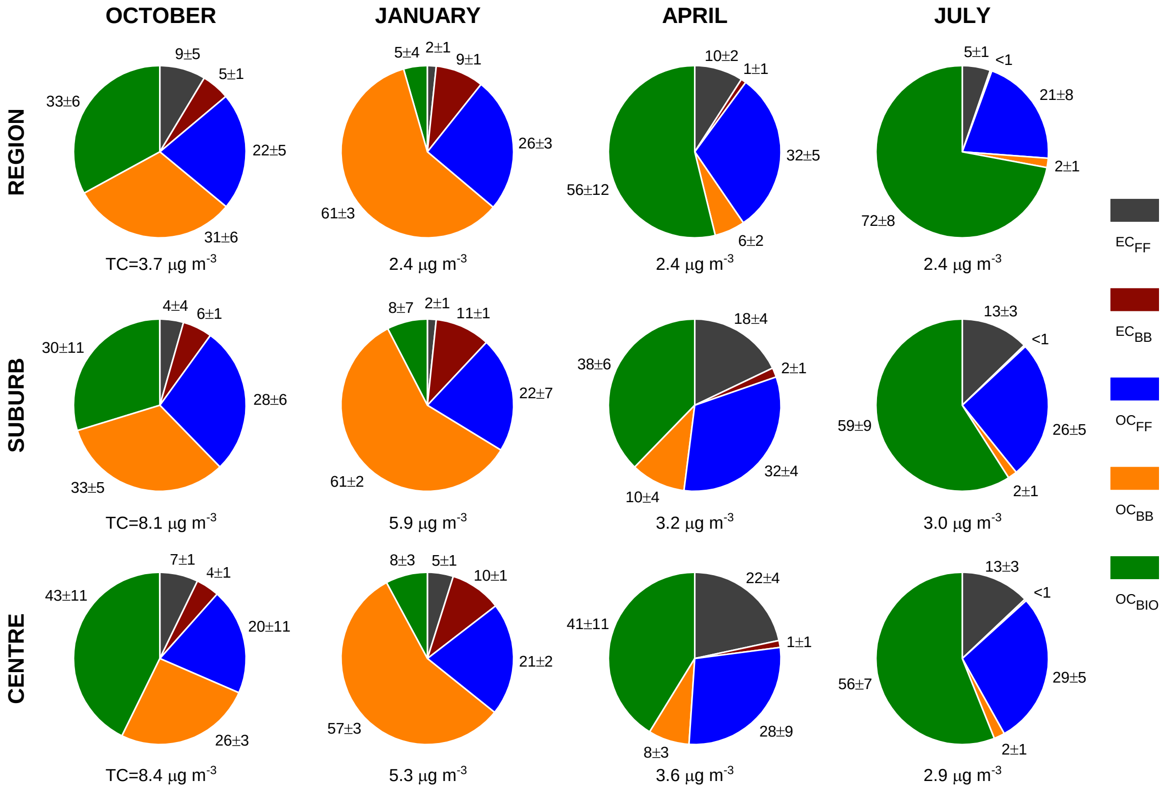

Figure 5. Mean contributions with SDs of elemental carbon from fossil fuel combustion (EC_{FF}) and from biomass burning (EC_{BB}), of organic carbon from fossil fuel combustion (OC_{FF}), from biomass burning (OC_{BB}) and from biogenic sources (OC_{BIO}) to $PM_{2.5}$-fraction total carbon (TC) in % for regional background in the Carpathian Basin, suburban area and city centre of Budapest and for different months of seasons. The median atmospheric concentrations of TC are indicated under individual circle charts, while the corresponding mean TC/$PM_{2.5}$ mass ratios are shown in Table 3.

The overall relative contributions are in good agreement with other similar or accompanying atmospheric studies (Szidat et al., 2006, 2009; Gelencsér et al., 2007; Minguillón et al., 2011; Bernardoni et al., 2013; Bonvalot et al., 2016).

Further conclusions can be derived by focusing on specific contributions of EC_{FF} and EC_{BB} to EC, and of OC_{FF}, OC_{BB} and OC_{BIO} to OC (Figs. 6 and 7, respectively). Elemental carbon is sometimes applied as a marker of automotive emissions mainly from diesel engines in cities of the continental mid-latitude northern hemisphere. The present research indicates that in urban ambient air in Central Europe, this assumption is, however, valid only in April (spring) and July (summer; when the share of the EC_{FF} was indeed larger than 90%). In October, the contributions of EC_{BB} can be considerable (up to 40–60%) at urban sites, so they can be by no means negligible. Furthermore, in January, the relative mass of soot particles from BB can be even larger.

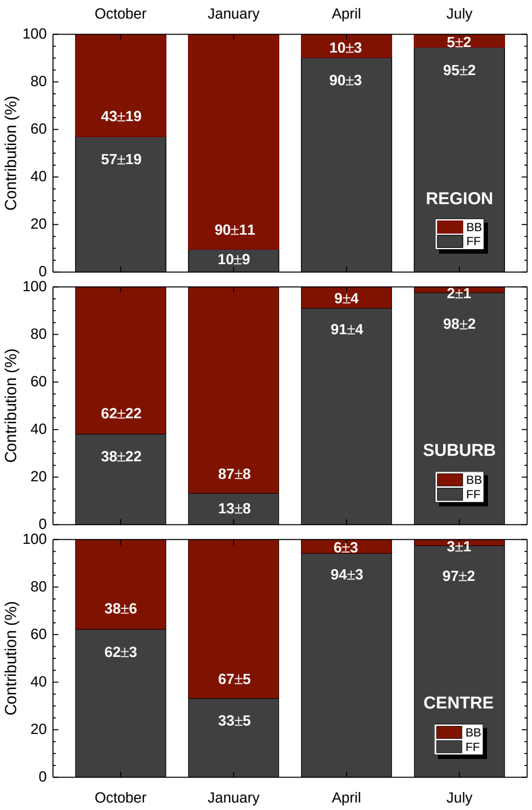

Figure 6. Distribution of mean contributions from FF combustion and BB with SDs to the $PM_{2.5}$-fraction EC for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons. The corresponding median atmospheric concentrations of the EC are given in Table 2.

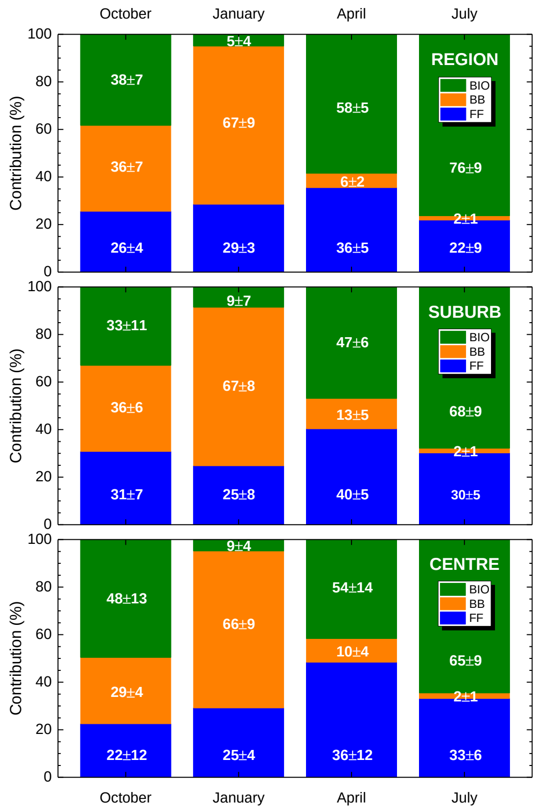

Figure 7. Distribution of mean contributions from FF combustion, BB and biogenic sources (BIO) with SDs to the $PM_{2.5}$-fraction OC for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons. The corresponding median atmospheric concentrations of OC are given in Table 2.

3.6 Potentials for air quality

To examine the potentials of the apportioned carbonaceous species for regulatory and legislation purposes, the contributions of the main source types to the $PM_{2.5}$ mass were roughly estimated. It was assumed that the OM/OC conversion factors for the aerosol particles originating from FF combustion, BB and biogenic sources were equal to the conversion factor for the bulk fine-fraction particles, thus 1.4 for the regional background and suburban area, and 1.6 for the city centre (see Sect. 2.3). We are aware that high emissions of some pyrogenic inorganic species such as K, nitrate or sulphate are neglected in the present apportionment model and that the OM/OC conversion factor can also change for organic species from different source types. The present estimates should, therefore, be considered as the first approximation only, and the contribution of BB to the $PM_{2.5}$ mass is likely underestimated. The results obtained are summarised in Table S7 in the Supplement. The separate contributions typically represent up to 1/5 or 1/4 of the PM mass as lower estimates and are discussed in Sect. 4. The contributions to the PM mass can be especially valuable when inspecting their tendencies.

The contributions were evaluated as function of the $PM_{2.5}$ mass concentration, which is one of the key measures/metrics for air quality considerations. The plots for the monthly mean contributions of OC_{FF}, OC_{BB} and OC_{BIO} to the TC are shown in Fig. 8a–c, respectively. The contributions of FF combustion (Fig. 8a) did not seem to depend substantially on the $PM_{2.5}$ mass level at any of the locations, so FF exhibits a constant and steady-state importance over various air pollution periods. The share of BB showed an increasing tendency with poor air quality (Fig. 8b). The change rate (the slope of the fitted line, b) was larger for the regional background (b=6.7) and smaller but similar to each other for the two urban sites ($b \approx 2.1$). The trends for the biogenic sources were just the opposite (Fig. 8c); their relative importance decreased by poorer air quality. The tendency was similar again for the two urban sites ($b \approx -1.5$) and substantially larger for the regional background (b=–6.9). The tendencies of the EC_{FF} and EC_{BB} were analogous. These results together indicate that BB influences the air quality in the regional background very extensively and it also has substantial effect on the air quality in the Budapest area, mainly in winter and possibly in the autumn months as well. The conclusions have importance in and consequences on the potentials for improving the air quality further interpreted in Sect. 4.

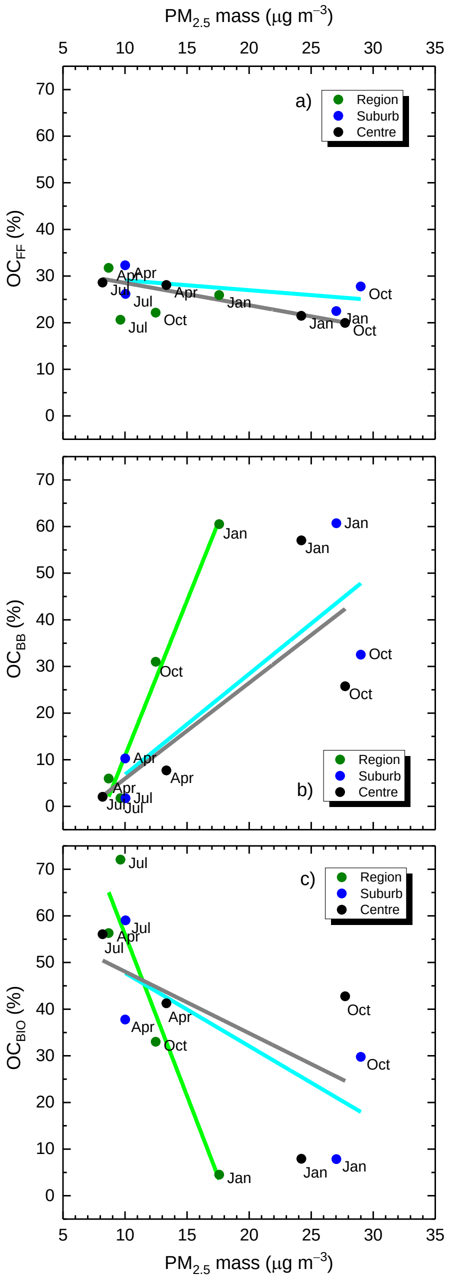

Figure 8. Monthly mean contribution of apportioned OC_{FF} (a), OC_{BB} (b) and OC_{BIO} (c) to TC as function of the monthly median $PM_{2.5}$ mass concentration (surrogate or proximity value for the air quality) for regional background in the Carpathian Basin, suburban area and city centre of Budapest. The months are marked by their starting letters. The fitted linear lines are just to guide the eye.

4 Conclusions

In the present study, the major carbonaceous aerosol species of OC and EC were apportioned among FF combustion, BB and biogenic sources in various types of atmospheric environments of interest in the Carpathian Basin in different months of seasons. The research work is the first extensive adaptation of the coupled ^{14}C-LVG marker method recently developed. In addition, the experimental and derived data and results achieved were obtained from the first systematic complex research project as far as the spatial scale within the basin and time span (of 1 full year) are concerned. The conclusions represent novel and valuable research contributions on a large area in Central Europe.

The carbonaceous particles made up from 30% to 48% of the $PM_{2.5}$ mass (as monthly mean) depending on the environment and months of seasons. It is the BB in winter that represents the largest potential (with a mass share of >20%; Table S7 in the Supplement) for improving the air quality both in cities and on rural areas of the basin. It is worth mentioning that all air pollution (smog) alert episodes in Hungary were announced so far exclusively because of the PM_{10} mass limit exceedances and they all happened in winter. Possibilities in controlling various forms of BB for air quality improvements seem to be, therefore, rather relevant due to this coincidence. In the present case for instance, there were 3, 8 and 8 days, respectively (19 days in total) in the subset of 4×7 days in the regional background, suburban area and city centre which daily mean values exceeded the EU annual $PM_{2.5}$ limit value of 25 µg m^{-3}. They all occurred in winter and autumn. If the BB sources (i.e. OC_{BB} and EC_{BB}) had decreased by half of their actual concentrations then the number of exceedance days would reduce to 2, 6 and 5, respectively (13 days in total, thus by 32%), while a perfect fuel gas aftertreatment of the BB as a sources would result in the number of exceedances of 1, 4 and 5, respectively (10 days in total, thus by 47%). In addition to carbonaceous particles, some adjunct inorganic constituents are also generated and, more importantly, soil or mineral dust and fly ash particles are also mobilised or blown up into the air due to the combustion or burning process itself. These, on the one hand, can further and substantially enhance the overall mass contributions and potentials of the high-temperature sources (including BB), and, on the other hand, may change somewhat their relative contributions. The presented conclusions and overall outcome of the research can directly be utilised as a background in modifying the municipal air quality regulations in Budapest, which is currently under preparation, and for inspiring the users of household heat appliances for societal implications of atmospheric aerosol. The apportioned contributions can also be used as a starting point for climate-related projects as far as the regional or urban climate in Budapest are concerned.

Fossil fuel combustion is an abundant source of PM mass (with a share of >20%; Table S7) only at urban sites and only in April and July. Resuspension or suspension of road and surface dust by moving vehicles can again represent a substantial auxiliary increment for FF contribution. Biogenic sources are normally considered as natural process or to be dominated by natural processes, and, therefore and strictly speaking, they are not associated with the issue of air pollution. It is expected that the unaccounted $PM_{2.5}$ mass contains secondary inorganic aerosol particles mostly sulfates, nitrates and elements, and soil or mineral/crustal rock dust particles as well (Salma et al., 2001). These constituents should definitely be revisited and taken into account in further source apportionment research.

Another challenge in health-related or air-quality-type assessment studies is to refine the apportionment within the major source types with burning of plastics, domestic or agricultural waste (garbage), coal and stained wood in households through identification of their appropriate tracers and via quantification of various emission factors of their specific sources e.g. by advanced hyphenated MS or optical methods combined with powerful statistical data treatment. These additional combustion categories deserve more investigations since many of them seem to be prevalent and of increasing volume in the studied geographical area, and they produce some specific air pollutants or toxics which can present serious risk for human health, wellbeing and the environment.

Data availability. Raw data are available from the corresponding author on reasonable request.

Supplement. The supplement related to this article is available online.

Competing interests. The authors declare that they have no conflict of interest.

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