

Response to Referee number 1

1st March 2020

The authors would like to thank Referee no. 1 sincerely for his/her very detailed, expertise and valuable comments to further improve and clarify the MS. We have considered all recommendations and made the appropriate alterations. We would like to touch upon that the Referee most likely evaluated the acp-2019-792-manuscript-version1.pdf and not the latest version acp-2019-792-manuscript-version2.pdf, which was created and resubmitted after the access review. In the latter version, some technical corrections and other smaller updates were adopted on the request of the co-editor, and a geographical map was included as well. This means that some issues raised by the Referee had been already handled and improved. Our specific responses are as follows, while the textual modifications can be followed in the marked-up version of the MS, which is attached.

General comments

English language should be thoroughly checked by a native speaker. Especially in the introduction some sentences are a bit hard to understand.

1. Several sentences of the MS, particularly in the Introduction, were shortened, simplified and the MS was also checked by a native English speaker. We also expect that the MS will receive English language copy-editing during the typesetting.

Chapter 2.1. It would help reader if you could name the stations in this chapter and clearly state stations called hereafter xx, zz and yy. Now there is many kind of variations of the names in the text. Maybe also include a more in-depth description of the area where stations are located (what kind of area, how many inhabitants, how much traffic/biomass burning/industry etc is in area, any prior knowledge about the expected sources?). Maybe the distance between stations or a map would help reader also. Please, add for all measured parameters the instrument, model and manufacturer. Also, it is bit hard to understand where e.g. DMPS was measuring and how long. Check and explain the used acronyms and terms. E.g. for elemental carbon both EC and soot are used, which can be very confusing to some of the readers. Also, terms carbonaceous aerosol, total carbon should be explained. More literature references should be added to the text to further discuss the results and their significance. Also more discussion about where these results could be utilized would be useful. The novelty value of results should be highlighted more!

2. The naming of the three measurement sites was unified all over the MS as regional background, suburban area and city centre. The suburban and centre sites are called together as urban locations. We also described the locations in more detail and added a geographical

map showing the three sampling sites and the distances between them. The description of the instruments was extended somewhat and clarified. The difference between soot and EC was made more exact (see also Responses no. 4 and 5). The total carbon was already defined as $TC=OC+EC$ in the line 85 of the MS ver. 1. We put more emphasize on the novelty of the approach and results, and on their utilization and potentials in Sect. 5, Conclusions.

Tables 2,3: The sampling periods for all stations are different. At the Central station the sampling period is much shorter. Are the mean/meadian values and ratios calculated for all the samples or only for the seven simultaneously collected samples? If sampling times are not exactly same, is it fair to compare the results of stations? e.g. some episode could change the concentrations and affect the observed mean values significantly.. if this episode is only included in longer timeseries measured in Background/suburban areas, this could affect the comparison when the results of different stations are compared.

3. The average atmospheric concentrations in Tables 2 and S2 were calculated by considering all sampling days. The major reason for doing this is to characterize the environments and months as representatively as possible. These overall mean values were compared to previous results. This is mentioned explicitly in the text now. We also added a brief note on constrains imposed by not completely overlapping sampling intervals at the city centre with respect to the other two sites. Of the three locations, we have the most extensive information for the city centre, where the present median concentrations were perfectly in line with the earlier results. Atmospheric concentrations in the Carpathian basin are often governed by local meteorology (Salma et al.: Elemental and organic carbon in urban canyon and background environments in Budapest, Hungary, *Atmos. Environ.*, 38, 27, 2004). Since the meteorological data during the sampling intervals were in line with monthly characteristics and without any extremes, it can be expected that the aerosol samples collected represent the months correctly. This confirms the comparison. We also modified the naming of the sampling intervals in the entire MS to express that an interval is more related to a month than to a whole season. A note was also added on the representativity of the sampling intervals into Sect. 3. The radiocarbon analysis was performed on seven selected samples which were collected in parallel on fully overlapping days at each sampling location and in each month. The comparison of the apportioned concentrations, their contributions and their interpretations (which are the principal objectives of this paper) are fully justified. All these arguments were now included briefly into the text at several places.

Minor comments

Line 35: define carbonaceous

4. The term “carbonaceous aerosol constituents/species” refers to the carbonaceous components of the particles. In the fine size fraction, they ordinarily represent organic compounds and soot. This naming is quite straightforward, it is explained in basic textbooks (e.g. Gelencsér: Carbonaceous aerosol, Springer, 2004, p. 2) and is generally accepted. The carbonaceous species usually occur together with inorganic compounds, and, therefore, the term “carbonaceous particles” is – strictly speaking – already a fiction in most cases because it expresses the carbonaceous constituents of the aerosol particles only. We prefer to use the former and well-defined term in all our papers. We added a reference on this issue (see also answer no. 5).

Line 39; define soot (as there is also EC)

5. The meaning of soot was explained as requested. In addition, soot was also related to EC at the first occurrence of the EC. We also added a reference dealing with this complex terminological issue in detail.

Line s43-51: Sentences are bit long and hard to read. Clarify this and maybe specify if these consequences in the list are positive or negative in the nature.

6. The sentences were shortened and simplified. Discussions of the effects listed, however, would be rather complex and long, and more importantly, they are not in line with the purpose of the MS.

Line 52: "Fuel wood"? does this refer to biomass combustion in residential scale?

7. Fuel wood is made from trunk and larger branches of trees and are burnt in both residential and industrial appliances. The sentence was extended.

Line 59-61: “Huge number, composite character, spatial and temporal variability of the sources together with the complex mixture and atmospheric transformation of their products make the quantification of these source types or their inventory-based source assessment challenging” Clarify this sentence, it is bit hard to understand.

8. We tried to reformulate the sentence without leaving out any important process.

Line 62-63: “There are several methods to apportion the particulate matter (PM) mass or carbonaceous species among some or all major source types.” please clarify this sentence

9. The sentence was clarified.

Lines 63-69. Sentence is really long, maybe split to 2-3 shorter sentences?

10. The sentence was split into two separate parts. The first sentence of them may still seem somewhat longer but – as a matter of fact – it is a list of available methods, which we would like to keep as one item.

Line 74: “The latter molecule is often applied together with its stereoisomers mannosan (MAN) and galactosan (GAN) since..” maybe change to “ Monosaccharide anhydride analysis often contains stereoisomers mannosan (MAN) and galactosan (GAN) in addition to levoglucosan since..

11. Adopted.

Line 100: what is the “latter type” referring to? please clarify

12. The expression “latter type” referred to biogenic sources. The sentence was modified.

Table 1. Why there is extra space between date and month as well as between the month and year in all time periods? please check the journal instructions how to give the dates..

13. The writing of the dates and times was adjusted the journal instructions in the whole MS.

Lines 152-167: Please add the model and manufacturer for all the instruments, provide the instrument information for the meteorological data as well information in which stations these instruments were used. E.g. was DMPS run in all stations constantly, or was one dmeps moved between the stations? Maybe a table with station, instruments, models and measured components would help reader to understand the situation.

14. The parts on particle number concentrations and the related reference were removed on the request of the Referee no. 2. We included the types and manufacturers of the meteorological instruments which make it feasible to obtain further information on them. We consider listing all details for measuring auxiliary variables, which are part of a quality-controlled national meteorological network not completely justifiable since these long and detailed lists would make the article over-descriptive and would detract the attention from the aimed messages.

Line 169: Add balance model and manufacturer

15. The requested information was added.

Line 173: where does this LOQ value for PM mass comes from?

16. The specified limit was determined from evaluating measured data for several box blank filters identical to our substrate within a separate exercise. The procedure included for instance uncertainties of weighing, sampled air volume determination and environmental conditions according to EN12341:2014. We added a brief note on this into the text.

Line 198: what is origin of the LVG observed in the blank filters? please add how much levoglucosan was observed in the blanks.. has this kind of blank values seen in other studies?

17. 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 variations of the baseline of the measurement. We added a reference on them (Maenhaut et al.: Assessment of the contribution from wood burning to the PM₁₀ aerosol in Flanders, Belgium, Sci. Total Environ., 437, 226, 2012) , which showed similar blank values. The blanks were the largest in comparison 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 substantially smaller than this. We added a related brief comment to the text.

Line 203: which days?

18. The overlapping days were given in an explicit way.

Line 236-240: “Whenever it was possible, the comparisons of atmospheric concentration, other variables or their ratios with respect to sites or seasons 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”. Please explain what variables/ratios this refers to?

19. A sentence was modified to be more specific.

Line 275: Maybe add some values for average temperature, wind etc meteorological parameters to article also (not only supplement) as people not living in Budapest may not know the normal local conditions mean.

20. The average values for the complete list of meteorological variables are given in Table S5 in the Supplement. We would like to avoid showing only some selected variables separately, and instead, we further emphasized that they are fully available in the Supplement.

Line 284-286: “The former variable represents the bulk fine PM; EC is a typical primary aerosol constituent, while WSOC is expresses the SOA.” Sentence is hard to understand, please clarify what this means.

21. A typo unfortunately remained in the sentence which made it difficult to understand. It was corrected and in addition, the sentence was further clarified.

Chapters 3.2-3.6 please add some numerical values to text also. Would be also useful to compare more to literature whether the values were as expected or may be lower/higher..

22. We compared our present results better to existing partial or overlapping information and discussed the consequences of the inter-comparison. We also added some further references.

Line 391-395: maybe this information should be in experimental chapter?

23. The suggestion could indeed be a plausible option. Our specific intention by keeping this short but important discussion among the results was to attract more attention to possible contamination by anthropogenic ^{14}C in large cities. This risk is not very often mentioned in urban radiocarbon studies.

Line 359-365; “These can be explained by larger intensity of soot emissions from incomplete burning (road vehicles, residential heating and cooking by solid fuel), which is a typical anthropogenic source, and which is associated with seasonal variation (e.g. due to residential heating) as well as with constant sources (e.g. due to traffic or cooking) over a year.” what is the seasonal variation observed for road vehicles? how they differ from traffic that does not have seasonal variation?

24. The seasonal variability was related to residential heating, while the road traffic is expected to be a source with more-or-less constant daily average intensity of soot particles (see Salma et al., Elemental and organic carbon in urban canyon and background environments in Budapest, Hungary, Atmos. Environ., 38, 27, 2004). The related sentence was reformulated to avoid the possible misunderstanding.

Line 501-502: Did temperature have similar trend as OC_{BIO}? I would have expected to see highest OC_{BIO} concentrations in summer.. or is there a reason why autumn OC_{BIO} was higher? how does this observation compare to other studies?

25. The tendency in the air temperature can be seen in Table S5 in the Supplement. It did not change in line with the OC_{BIO} (in contrast to species apportioned to BB). Their coefficients of correlations at all sampling sites are summarized in Table S6, which indicate a modest

(linear) relationship between T and OC_{BIO} in the regional background and insignificant dependencies at the urban sites. The links between them are rather complex and not fully uncovered. The OC_{BIO} depends on many other atmospheric properties and parameters than T in a multifactorial manner. This set including for instance the changes of the SOA yield for mixed air masses of biogenic and urban origin. Our conclusion is in line with a recent and very important article dealing with the SOA formation (McFiggans et al.: Secondary organic aerosol reduced by mixture of atmospheric vapours, Nature, 565, 587, 2019). It is needed to continue the studies on the SOA yield in these specific atmospheric environments by dedicated experiments and methods to get better insights into and explanations for our apportioned results. The related text was reformulated to further emphasize these aspects, and a brief comparison was also added.

Chapter 3.6 and conclusions: the impact of results is now discussed from the air quality point of view. Maybe add something about climate and health point of view also? assumably the anthropogenic emissions and BC have different, likely more negative health impacts. BC has also strong climate impact..

26. The consequences of the study on the air quality in the Carpathian Basin are dealt with in a separate section (3.6). These are related to health effects as well. We amended several modifications into the Conclusions section to indicate their climate implications as well.

Imre Salma
corresponding author

Response to Referee number 2

1st March 2020

The authors would like to thank Referee no. 2 very much for his/her very detailed, expertise and valuable comments to further improve and clarify the MS. We have considered all recommendations and made the appropriate alterations. Our specific responses are as follows, while the textual modifications were amended and can be followed in the marked-up version of the MS, which is attached.

General comments

First, the naming of sampling periods (winter, spring, summer and autumn) suggests that the data are representative for these seasons. This is not true as sampling was performed during 14 (or even 7) consecutive days during each season and only 7 over-lapping days were fully analysed for 14C. These sampling periods are too short to be representative for a season and therefore, months, when data were taken, are more proper for naming of the sampling periods. For the same reason, the authors should more concentrate on differences between the sites and less on “seasonal” characterisation and differences. More detailed weather characteristics for each sampling period can explain more various type of events that change differences among the sites.

1. We modified the naming of the sampling periods in the entire MS to express that they are related more to a month than to a whole season. A note was also added on the representativity of the sampling intervals into Sect. 3 to clarify the situation more carefully and considerately. We included the aspect raised by the Referee in the second part of this comment into the interpretations of the data, and performed several modifications of the text accordingly.

Second, median values presented e.g. in the Table 2 or 4 can be representative for only part of the data especially if two types of atmospheric mixing were present during short sampling period. Therefore, either medians with high and low percentiles or averages with standard deviations should be presented together. In addition, particle number concentrations paragraph (lines 437-442) is completely out of topic of the paper, it should be omitted together with related references.

2. We added new tables into the Supplement (Tables S2 and S4), which contain the means and SDs of atmospheric concentrations of aerosol constituents and gases, and explained its motivation in the MS. We originally included the particle number concentrations to demonstrate the decoupling between PM mass and particle number. As requested, we removed the related paragraphs and reference.

Finally, the combination of OM/OC conversion factors used by authors is not logical and is not based on current scientific knowledge and must be corrected. Therefore, most of the calculations must be corrected.

Line 251-253 - The authors use conversion factor for city centre 1.6 and for suburban and rural sites 1.4. This is taken opposite way than it is usual. While both lower values 1.4 and used value 1.6 can be accepted for places with fresh traffic aerosols – city centre the value 1.4 used for urban and rural background is unacceptably low. Some seasonal dependence of this factor can be also expected. Actually, cited work of Turpin and Lim 2001 says in its abstract: "This investigation suggests that 1.4 is the lowest reasonable estimate for the organic molecular weight per carbon weight for an urban aerosol and that 1.4 does not accurately represent the average organic molecular weight per carbon weight for a non-urban aerosol. Based on the current evaluation, ratios of 1.6 ± 0.2 for urban aerosols and 2.1 ± 0.2 for non-urban aerosols appear to be more accurate" Therefore, the calculation for suburban and rural sites have to be recalculated with higher conversion ratio OM/OC (at least also 1.6).

3. The organic aerosol-to-organic carbon (OC) mass conversion factor is an estimate of the average molecular mass per C atom for organic matter (OM) in general. It is site-dependent and can have seasonal and diurnal variations as well. Therefore, the factor cannot be considered as a conclusive or constant/generally valid value. It is usually derived by indirect considerations (Russell, Aerosol organic-mass-to-organic-carbon ratio measurements, *Environ. Sci. Technol.*, 37, 2982, 2003). Mass conversion factors between 1.2 and 1.4 were estimated for fine atmospheric aerosol in mildly oxidizing atmospheric environments (Turpin et al.: Measuring and simulating particulate organics in the atmosphere: problems and prospects, *Atmos. Environ.*, 34, 2983, 2000). Some further studies suggest that a factor of 1.6 ± 0.2 describes better the oxidizing urban environments (Turpin and Lim: Species contributions to $PM_{2.5}$ mass concentrations: revisiting common assumptions for estimating organic mass, *Aerosol Sci. Technol.*, 35, 602, 2001). Identical partial mass conversion factors of 1.81 were obtained for HULIS both at a rural site of the Carpathian Basin and in Budapest (Kiss et al.: Characterization of water-soluble organic matter isolated from atmospheric fine aerosol, *J. Geophys. Res.*, 107(D21), 8339, 2002; Salma et al., Sampling artefacts, concentration and chemical composition of fine water-soluble organic carbon and humic-like substances in a continental urban atmospheric environment, *Atmos. Environ.*, 41, 4106, 2007, respectively). HULIS are comprised primarily of a complex multi-component mixture of compounds that bear aliphatic chains with carboxyl, hydroxyl, carbonyl or phenol terminal groups. Thus, they contain relatively rather large number of heteroatoms to C but exhibit an OM/OC ratio of "only" 1.81, while they mass contribution to OC could be 20–30%. It should also be noted that the conversion factor is one of the most substantial sources of uncertainty in aerosol chemical mass closure calculations involving OM. It was estimated that the

relative uncertainty associated with the conversion is approximately 30% (Maenhaut et al., Assessment of the contribution from wood burning to the PM₁₀ aerosol in Flanders, Belgium, Sci. Total Environ., 437, 226, 2012). In the present study, we adopted the factor of 1.4 for the regional and suburban environments and the factor of 1.6 for the city centre. We would like to keep our selection because of several reasons. 1) We think that the larger factor mentioned and quoted by the Referee for rural and suburban environments is primarily valid for chemically aged aerosol, which was not the typical case at our sampling sites in the Carpathian Basin. Most aerosol particles are generated by local or regional sources here. 2) The two factors of 1.4 and 1.6 under discussion have uncertainties which are identical to or even larger than the differences between the factors. Moreover, the factor does not affect at all the major objectives of the MS, namely the apportionment of the basic classes of OC and EC from FF combustion, BB and biogenic sources, and their contributions to TC. 3) Our previous studies in this field and geographical area justify our selection since we obtain consistency in the results in general for various organic aerosol types and environmental types within the Carpathian Basin. 4) We utilized the present ratios in our several earlier publications (including ACP articles as well) and keeping the present conversion factors also facilitates the comparison among the present and previous results. As a compromise, we extended the related parts of the MS with these discussions and explanations, and further emphasised the role of methodological uncertainties or limitations in the text whenever it was relevant.

Specific comments

Line 265 – It should stay “Their” instead of “They”

4. Corrected.

Line 291-293 – Measure of photochemical activity is not ozone concentration itself

5. The sentence was reformulated.

Line 305 – WSOC vs SOA relation can be biased by biomass burning emissions. Therefore, the sentence needs correction. Compare also with lines 385 and 388.

6. The sentence was extended by this aspect as well.

Line 378 - “minimum in summer” can be omitted as it is mentioned again in the next sentence.

7. The expression was removed.

Lines 437-442 – It is out of topic, remove the paragraph

8. We originally included the information on the particle number concentrations to demonstrate the decoupling between PM mass and particle number. Nevertheless, we removed the paragraph as requested.

Lines 456 - 457 – The sentence should be corrected, the results do not justify fully such sentence.

9. The related part of the sentence was deleted.

Line 488 – OC/EC ratios can be influenced also by other effects, therefore less strong opinion would be more proper

10. The sentence was reformulated in the requested manner.

Line 547 – correlation coefficients are significant or insignificant based on given statistical criteria. Correct the sentence.

11. The sentence was extended by the significance limit.

Line 548 – “linear” relationship of OC_FF with NO was seen for suburban site only (corr. coef. 0.93) while for city centre was only 0.39. Therefore, the sentence needs correction or clarification.

12. The sentence was corrected and extended into a more precise and clearer formulation.

Line 551 – the last sentence should be removed or corrected. The correlations can support results but not approve them.

13. The sentence was removed.

Line 569-570 – the differences in share of OC_BIO are negligible in comparison with their uncertainty, therefore, no tendency can be retrieved from the data. Correct the sentence accordingly.

14. The sentence was changed to include this limitation.

Lines 662-664 - again OM/OC conversion factors – correct as mentioned above.

15. Section 3.6 deals with the potentials of the apportioned chemical species on the air quality as it is explicitly expressed in the text, and some rough assumptions, which are also outlined, were utilized. From this aspect, the differences caused by the two possible OM/OC conversion factors of 1.4 or 1.6 seem unimportant. The limits of the approach were further explained and discussed in Sect. 3.6. See also the answer no. 3.

Graph 8 – if authors want to show differences in OC shares during their sampling periods they should stop call them seasonal differences, as their sampling periods cannot fully represent seasons. Moreover, the lines in graphs are not representative for the data giving sometimes unrealistic impression about the data. Redo the graph.

16. The naming of the sampling periods was modified as requested and the name of the corresponding months were adopted instead. The line with a time tendency in question was removed from the plot.

Imre Salma
corresponding author

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

Imre SALMA¹, Anikó VASANITS-ZSIGRAI¹, Attila MACHON², Tamás VARGA³,
István MAJOR³, Virág GERGELY³, Mihály MOLNÁR³

¹ Institute of Chemistry, Eötvös University, Budapest, Hungary

² Air Quality Reference Center, Hungarian Meteorological Service, Budapest, Hungary

³ Isotope Climatology and Environmental Research Centre, Institute for Nuclear Research, Debrecen, Hungary

Correspondence to: Imre Salma (salma@chem.elte.hu)

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 seasonal-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 seasonsyears. In Octoberautumn, the three major sources contributed equally to the TC in all environments. In winterJanuary, it was the BB that was the major source with a share of 70% at all sites. The contributions from biogenic sources in Januarywinter were the smallest; ~~although they were still non-negligible with an increasing share (from 5 to 8%) from the regional background to the urban sites.~~ In Aprilspring, FF combustion and biogenic sources were the largest two contributors at all locations with typical shares of 45–50% each. In summerJuly, 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 Julysummer. The EC_{FF} made up more than 90% of EC in springApril and summerJuly, while in autumn-October and winterJanuary, 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, CO₂ 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 ~~the complex mixture and~~ 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 $\text{C}_6\text{H}_{10}\text{O}_5$), 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 ~~The latter molecule is often applied together with its stereoisomers mannosan (MAN) and galactosan (GAN)~~ 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 ($\text{TC}=\text{OC}+\text{EC}$) 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 ~~the latter type 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.

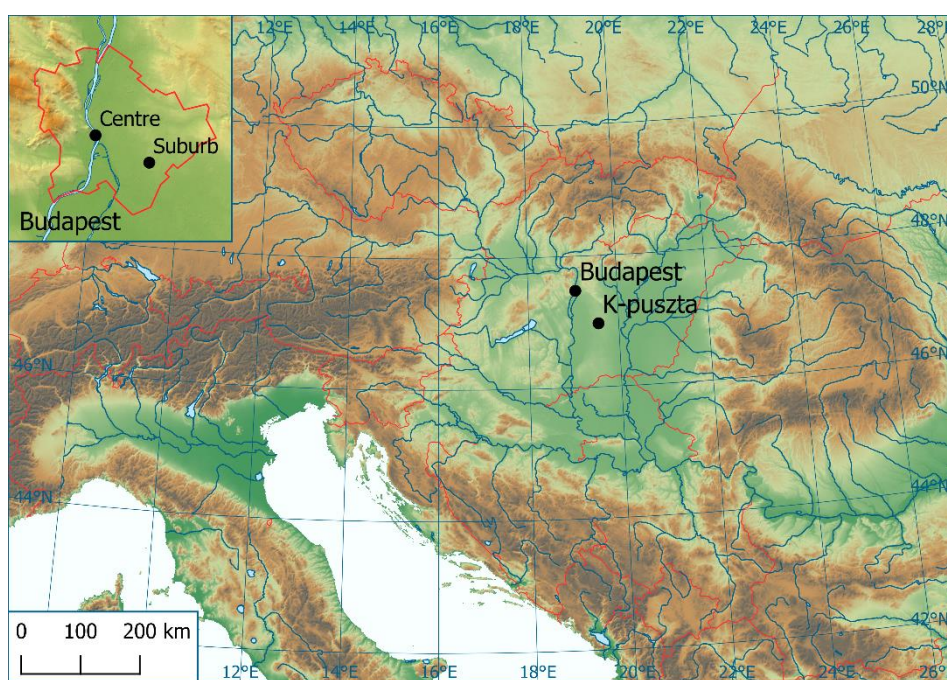


Figure 1. The Carpathian Basin with location of the sampling sites in Budapest (city centre and suburban area) and at K-puszta station (regional background).

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 m³. One field blank sample was also taken at each site and month. All filters were preheated 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. ~~Initials of the consecutive months allocated to the seasons are indicated in brackets.~~

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

* 01 August 2018.

~~Total particle number concentrations (N_{6-1000}) were derived by two differential mobility particle sizer (DMPS) systems of very similar construction with a time resolution of 8 min (Salma et al., 2016b, 2019). The DMPS measurements have been performed both in the city centre and regional background continuously and according to the recommendations of international technical standards.~~ Concentrations of criteria air pollutants, i.e. SO₂, NO/NO_x, CO, O₃ and PM₁₀ 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 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 cm² 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 cm² 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 cm² 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 ~~in each season (, 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 $^{14}\text{C}/^{12}\text{C}$ ratios were also corrected for isotopic fractionation by using the $^{13}\text{C}/^{12}\text{C}$ ratios (Wacker et al., 2010) that were obtained simultaneously in the actual AMS measurements. The $^{14}\text{C}/^{12}\text{C}$ 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 $^{14}\text{C}/^{12}\text{C}$ 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 HNO₃–H₂O₂ 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.

~~Whenever it was possible, the~~ The comparisons of atmospheric concentrations, other variables or their ratios with respect to the ~~seasons-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 $[(OC/LVG)_{BB}]$. ~~They~~ 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 $(16 \pm 5)\%$ (Szidat et al., 2006) and from a ratio and SD of $(18 \pm 4)\%$ (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 ~~seasons-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 ~~seasons-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 ~~seasons~~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 ~~seasons-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 than in the suburban area. It tended to show a maximum in summer. Such

a behaviour is typical for large-scale O₃ formation mechanism. This all suggests that there ~~were~~ could be substantial differences in photochemical activity in general as indicated by O₃ between the regional background and the urban sites except for summer. 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 winter 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 ~~seasons-months~~ and environments are shown in Figs. 2, 3 and 4, respectively. The PM_{2.5} mass former variable represents the bulk fine PM; EC is a typical primary aerosol constituent, while WSOC expresses mainly the-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.

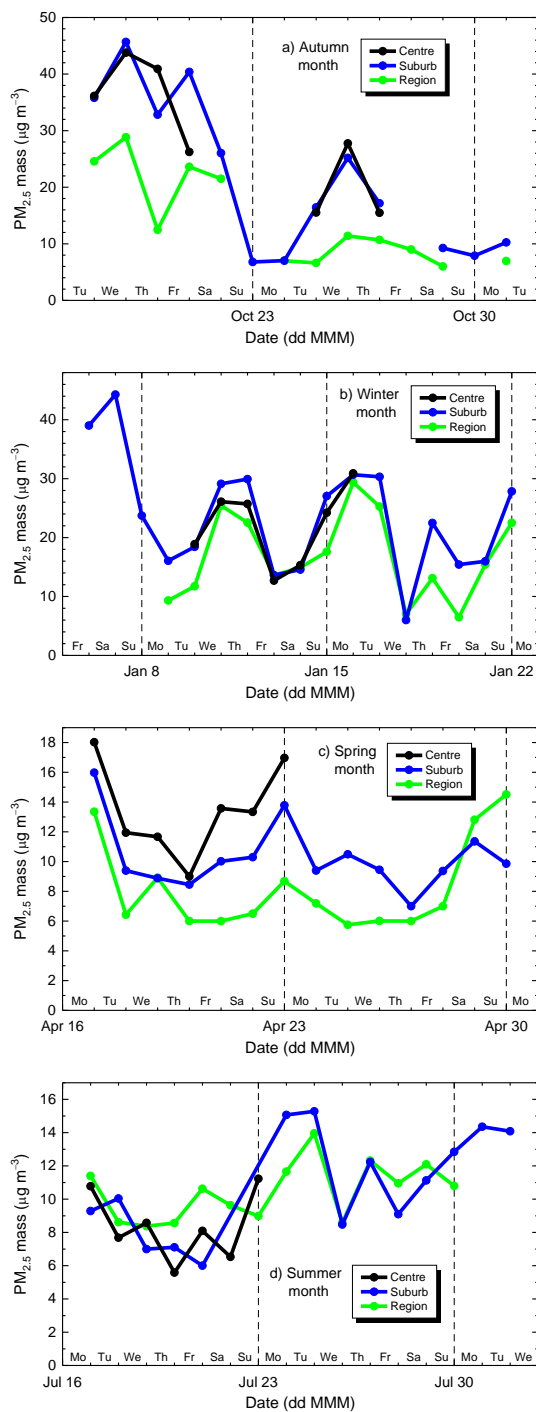


Figure 2. Time variation of $PM_{2.5}$ mass for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

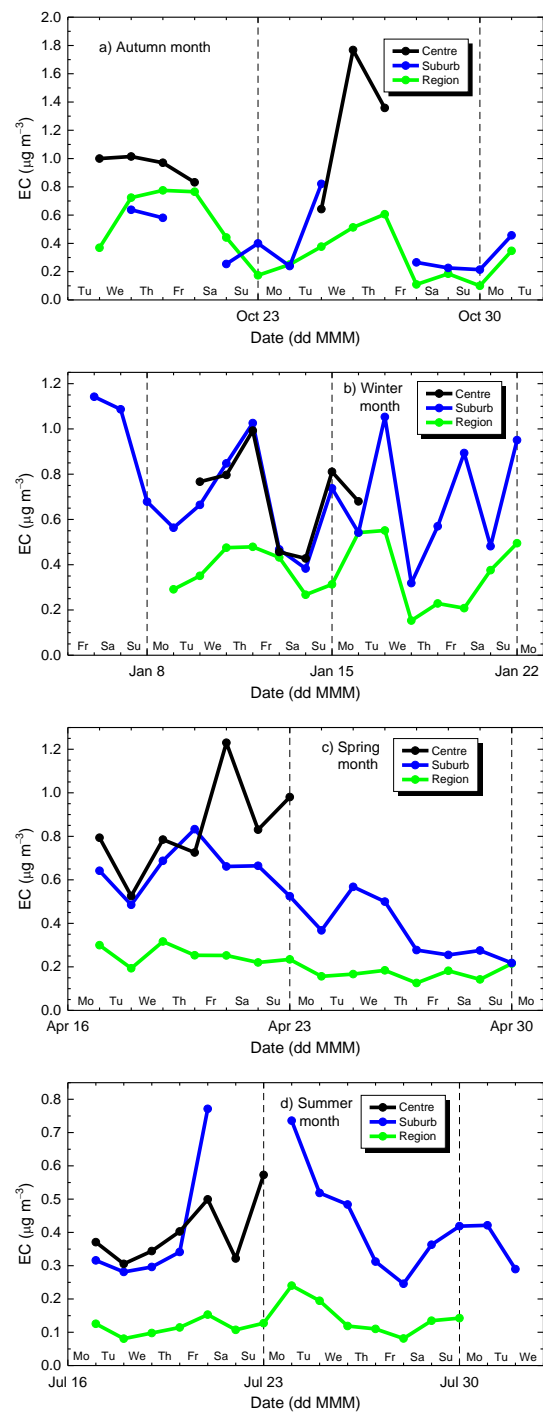


Figure 3. Time variation of EC for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

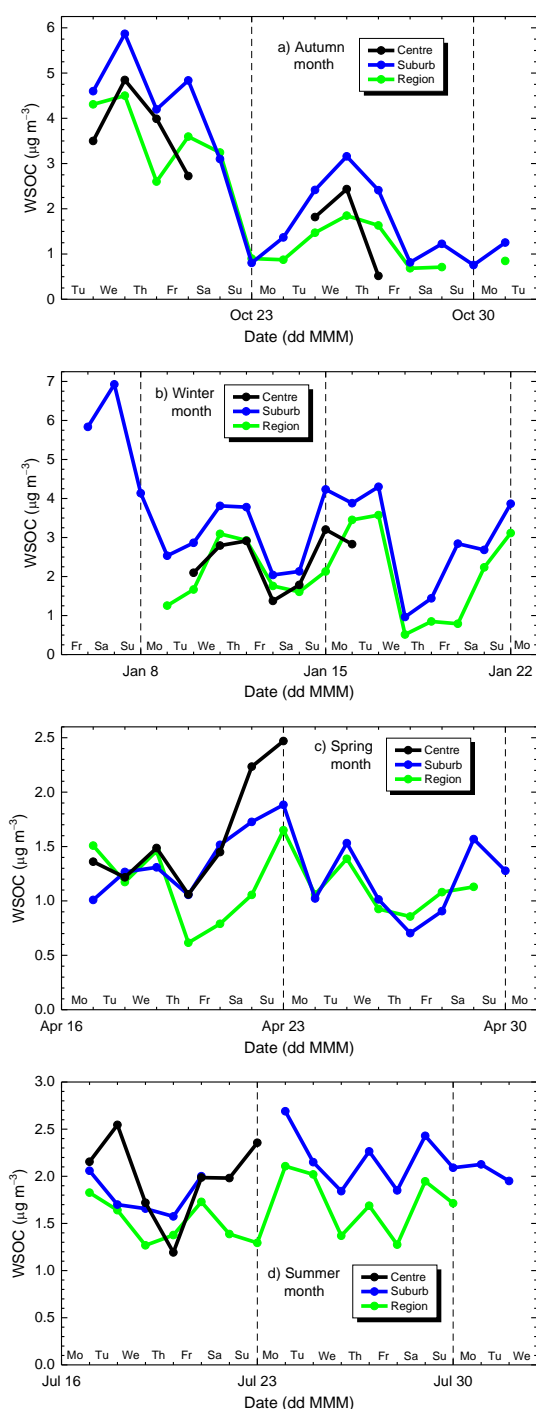


Figure 4. Time variation of WSOC for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling time intervals for different months (a–d).

3.2 Tendencies in aerosol concentrations

Median atmospheric concentrations of the measured aerosol constituents separately for the different ~~seasons-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 ~~winter~~-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 ~~autumn-October~~ and ~~winter-January~~ were similar to each other and they were the largest. In the suburban area, the EC data showed a maximum in ~~winter-January~~ and a minimum in ~~summer-July~~. In the city centre, the EC levels in ~~autumn-October~~, ~~winter-January~~ and ~~spring-April~~ were similar to each other, and they all showed a minimum in ~~summer-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 ~~a-typically of anthropogenic sourceorigin. ,and-which~~ These sources can have either is associated with seasonal variation-variability (e.g. ~~due to~~ residential heating) ~~as well as with or~~ constant intensitysources (e.g. ~~due to~~ traffic or cooking) over a yearly time span.

The WSOC showed maximum medians in ~~winter-January~~ at all sites. In ~~autumn-October~~ and ~~summer-July~~, the urban locations had similar concentrations to each other, while it was somewhat smaller in the regional background. In ~~winter-January~~, the suburban site exhibited the maximum median concentration. This is explained by larger influence of BB in this environment and ~~season-month~~ and by higher water solubility of its products (see ~~later and~~ Sects. 3.3 and 3.5). In spring, 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 ($\mu\text{g m}^{-3}$)	Region	12.5	16.5	8.6	10.7
	Suburb	25	26	9.7	11.7
	Centre	28	24	13.3	8.2
EC ($\mu\text{g m}^{-3}$)	Region	0.37	0.36	0.20	0.122
	Suburb	0.45	0.68	0.51	0.35
	Centre	0.99	0.77	0.79	0.37
OC ($\mu\text{g m}^{-3}$)	Region	2.3	3.2	2.0	2.2
	Suburb	4.5	5.4	2.4	2.7
	Centre	6.6	4.6	2.8	2.6
WSOC ($\mu\text{g m}^{-3}$)	Region	1.63	2.0	1.08	1.66
	Suburb	2.4	3.8	1.27	2.0
	Centre	2.7	2.8	1.45	2.0
LVG ($\mu\text{g m}^{-3}$)	Region	0.172	0.40	0.0180	0.0081
	Suburb	0.44	0.71	0.040	0.0124
	Centre	0.38	0.48	0.036	0.0103
MAN (ng m^{-3})	Region	19.2	18.5	2.6	<1.2
	Suburb	37	39	2.9	<1.2
	Centre	26	21	4.1	<1.2
GAN (ng m^{-3})	Region	n.a.	10.6	1.20	<0.5
	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 ($\mu\text{g m}^{-3}$)	Region	0.182	0.23	0.088	0.081
	Suburb	0.22	0.25	0.097	0.075
	Centre	0.26	0.27	0.106	0.057
Ni (ng m^{-3})	Region	0.75	0.68	1.21	1.12
	Suburb	0.88	0.78	1.24	1.09
	Centre	1.10	0.63	1.51	1.08
Pb (ng m^{-3})	Region	3.8	3.0	3.2	2.8
	Suburb	5.8	6.8	4.1	3.5
	Centre	7.5	5.2	4.4	2.3
N_{6-1000} (10^3-cm^{-3})	Region	3.9 [‡]	3.7 [‡]	4.8 [‡]	n.a.
	Suburb	n.a.	n.a.	n.a.	n.a.
	Centre	14.6	10.9	13.9	6.4

n.a.: not available; [‡]: for N_{6-800}

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 ~~winter~~ January and the smallest in ~~autumn~~ October. This could be affected by the share of hardwood burnt in different ~~seasons~~ 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 ~~autumn~~ 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, which~~ 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 ~~autumn~~ October, however, remained still somewhat higher (1.15) with respect to the other ~~seasons~~ 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 ~~autumn~~ October and ~~winter~~ 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 ~~spring~~ April and ~~summer~~ 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 ~~autumn~~ October, ~~winter~~ January and ~~spring~~ April increased monotonically for the regional background, suburb area and city centre. They showed a maximum in ~~winter~~ January. Its concentrations were the smallest in ~~summer~~ 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 ~~spring~~ 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).

~~The particle number concentrations in the city centre were 1 order of magnitude larger than in the regional background. This is explained by the spatial distribution of their main sources and by the relatively short atmospheric residence time of ultrafine particles (Salma et al., 2011). The latter property also causes that there are larger variations in their atmospheric concentrations, which implies that the present measurement (sampling) time intervals are not long enough for reliable conclusions on tendencies.~~

3.3 Tendencies in concentration ratios

Mean values and SDs of some important concentration ratios separately for the ~~seasons-months~~ and different environments are shown in Table 3. The $\text{PM}_{2.5}/\text{PM}_{10}$ mass ratio exhibited strong ~~seasonal-time~~ dependency. In ~~spring-April~~ and ~~summer-July~~, the $\text{PM}_{10}-\text{PM}_{2.5}$ fraction particles (coarse mode) made up approximately 2/3 of the particulate mass, while in ~~autumn-October~~ and perhaps also in ~~winter-January~~, the $\text{PM}_{2.5}$ 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).

Contribution of the OM to the $\text{PM}_{2.5}$ mass for the regional background, suburban area and city centre showed little ~~seasonal-time~~ variation with annual means and SDs of $(31 \pm 5)\%$, $(32 \pm 6)\%$ and $(35 \pm 7)\%$, 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), ~~and indicate a huge number, big variety and spatially more or less equally distributed sources of OC in the Carpathian Basin~~. The mean contributions of EC to the $\text{PM}_{2.5}$ mass were between 1 and 6%, with a minimum in the regional background in ~~summer-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 ~~seasonal~~ means revealed limited variability (except for the city centre, where it changed from 33% in ~~winter~~ January to 48% in ~~summer~~ 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.

Table 3. Mean values and SDs for the PM_{2.5}/PM₁₀ 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

The mean WSOC/OC ratios in ~~autumn~~ 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 ~~winter~~ 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 ~~seasons~~ 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 spring-April and summer-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~~ ~~indicate the~~ to atmospheric conditions under which the SOA formation is large~~the largest~~. The ratio had a maximum in the regional background in ~~summer~~July, which can be associated with large photochemical activity and strong GRad. The ratios for the urban locations did not indicate obvious ~~seasonal-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 ~~seasons-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 ~~could be~~ 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.

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 $\mu\text{g m}^{-3}$ 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

The median concentrations of EC_{FF} were similar to each other in ~~autumn~~October, ~~spring~~April and ~~perhaps in summer~~July as well and exhibited a minimum in January. In all ~~seasons~~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 ~~seasons~~months, while they tended to be larger than the regional values by a factor of 2–3 in ~~autumn~~October and ~~summer~~July. The EC_{BB} and OC_{BB} concentrations showed a maximum in ~~winter~~January and a minimum in ~~summer~~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 ~~autumn~~October and ~~spring~~April. The concentrations of OC_{BIO} showed a monotonically decreasing tendency from ~~autumn~~October to ~~summer~~July, ~~spring~~April and ~~winter~~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.

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. ~~The relationships approve the correct attribution of the apportioned species.~~

3.5 Contributions of source types

Fossil fuel combustion showed the most balanced and constant daily or ~~seasonal-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 $(31 \pm 7)\%$, $(36 \pm 12)\%$ and $(36 \pm 13)\%$, 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.

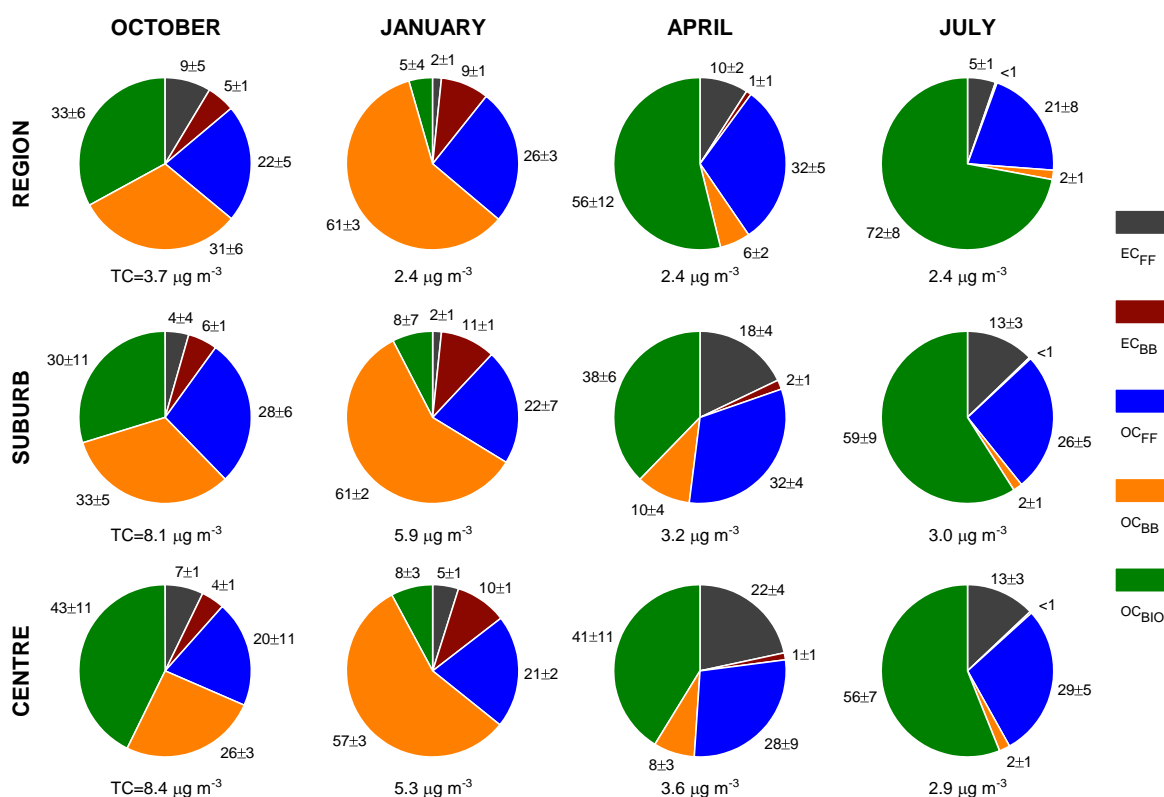


Figure 5. Mean contributions with SDs of elemental carbon from fossil fuel combustion (EC_{FF}) and from biomass burning (EC_{BB}), of organic carbon from fossil fuel combustion (OC_{FF}), from biomass burning (OC_{BB}) and from biogenic sources (OC_{BIO}) to PM_{2.5}-fraction total carbon (TC) in % 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 seasonal-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 autumn October, the three major source types contributed equally to the TC. In January winter, 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 season month. The contributions of FF combustion in January winter were similar to each other for all sites with a typical share around 25%. The contributions of biogenic sources were the smallest in this season month, although their firm interpretation is limited by the relatively large uncertainties ~~they were still non-~~

negligible. Their share ~~showed an increasing tendency from the regional background to the urban sites (from 5 to 8%), which could mightlikely~~ be ~~explained-linked to~~ by larger temperatures (urban heat island) and less snow coverage in the city centre (Sect. 3.1) than in its surroundings. In ~~April~~spring, 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 ~~spring~~April, which were increased monotonically in the order of the location type: region, suburb and centre. In ~~summer~~July, biogenic sources became the major contributor with a monotonically increasing share from the centre to the region.

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~~autumn, 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~~winter, the relative mass of soot particles from BB can be even larger.

It can be seen in Fig. 7 that the contributions from FF combustion to OC were fairly constant with a typical value of 25–35% and with an overall mean and SD of (30±8)% averaged for all atmospheric environments and ~~seasons~~months. In ~~April~~spring, some possible elevation could occur. Biomass burning was the major source of OC with a share of 67% in ~~winter~~January. Biogenic sources prevailed (with a share of 65–75%) in ~~July~~summer and made up about half of the OC in ~~spring~~April. The contributions from BB were hardly quantifiable in ~~summer~~July, while biogenic emissions ~~were-could be~~ still considerable in ~~January~~winter, particularly at the urban sites. In ~~autumn~~October, the three major source types made up balanced (constant and similar) contributions in all environments. Closely looking, there could be a slight overestimation of biogenic sources and a related small underestimation of FF combustion (see the apportionment scheme in Fig. 3 of Salma et al., 2017) in the city centre (left lower column

in Fig. 7) due to possible atmospheric contamination by anthropogenic ^{14}C , which was discussed in Sect. 3.2. It ~~is~~ has to be mentioned that primary organic aerosol is not included in the model, which could influence somewhat the overall contributions of the sources.

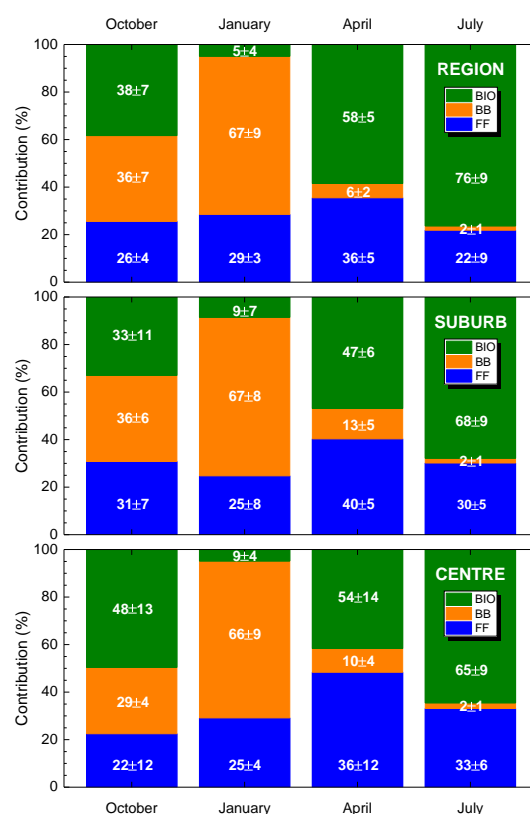
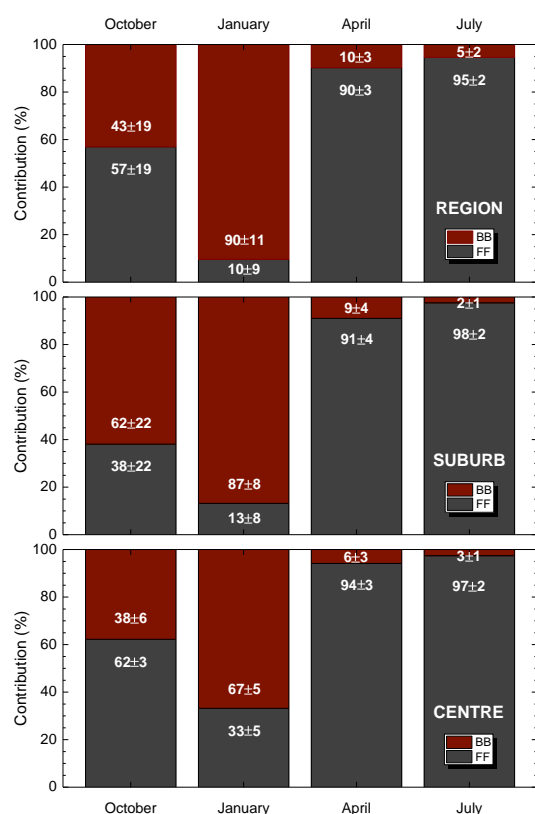


Figure 6. Distribution of mean contributions from FF combustion and BB with SDs to the $\text{PM}_{2.5}$ -fraction EC for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons. The corresponding median atmospheric concentrations of the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

Figure 7. Distribution of mean contributions from FF combustion, BB and biogenic sources (BIO) with SDs to the $\text{PM}_{2.5}$ -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 $\text{PM}_{2.5}$ 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 ~~also~~ 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 ~~seasonal~~ 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*≈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*≈−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.

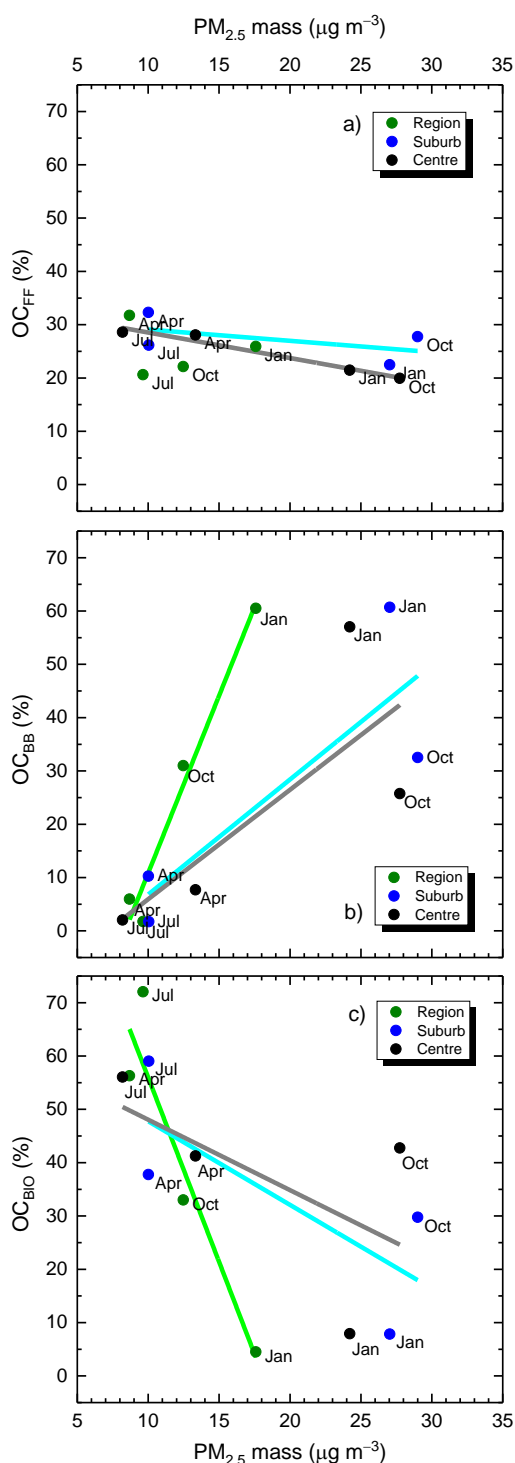


Figure 8. Seasonal-Monthly mean contribution of apportioned OC_{FF} (a), OC_{BB} (b) and OC_{BIO} (c) to TC as function of the seasonal-monthly median PM_{2.5} mass concentration (surrogate or proximity value for the air quality) for regional background in the Carpathian Basin, suburban area and city centre of Budapest. The months are marked by their two-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 in each 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 $\text{PM}_{2.5}$ mass (as seasonal-monthly mean) depending on the environment and months of seasons. It is the BB in January 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 $\text{PM}_{2.5}$ limit value of $25 \mu\text{g m}^{-3}$. 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 spring and summer. 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,~~which~~ 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 fuel~~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.

Acknowledgements. The authors are grateful to Gergő Farkas, Veronika Varga, Debóra Varga and Péter Varga of the Eötvös University for their help in collecting the aerosol samples and their chemical analyses. The map in Fig. 1 was created by Mátyás Gede of the Eötvös University, Department of Cartography and Geoinformatics. Funding by the National Research, Development and Innovation Office, Hungary (K116788 and K132254) is acknowledged. The research was supported by the European Union and the State of Hungary, co-financed by the European Regional Development Fund in the projects of GINOP-2.3.2-15-2016-00028 and GINOP-2.3.2-15-2016-00009 ‘ICER’. The project was realised within the frame of the COST COLOSSAL Action (CA16109).

References

- Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6, 3131–3148, 2006.
- Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud-precipitation interactions. Part 1. The nature and sources of cloud-active aerosols, *Earth-Sci. Rev.*, 89, 13–41, 2008.
- Artaxo, P., Rizzo, L. V., Paixao, M., de Lucca, S., Oliveira, P. H., Lara, L. L., Wiedemann, K. T., Andreae, M. O., Holben, B., Schafer, J., Correia, A. L., and Pauliquevis, T. M.: Aerosol particles in Amazonia: Their composition, role in the radiation balance, cloud formation, and nutrient cycles, in: Amazonia and Global Change, edited by: Keller, M., Bustamante, M., Gash, J., and Dias, P. S., 233–250, Geophysical Monograph Series, AGU, 2009.
- Bernardoni, V., Vecchi, R., Valli, G., Piazzalunga, A., and Fermo, P.: PM₁₀ source apportionment in Milan (Italy) using time-resolved data, *Sci. Total Environ.*, 409, 4788–4795, 2011.
- Bernardoni, V., Calzolari, G., Chiari, M., Fedi, M., Lucarelli, F., Nava, S., Piazzalunga, A., Riccobono, F., Taccetti, F., Valli, G., and Vecchi, R.: Radiocarbon analysis on organic and elemental carbon in aerosol samples and source apportionment at an urban site in Northern Italy, *J. Aerosol Sci.*, 56, 88–99, 2013.
- Birch, M. E. and Cary, R. A.: Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust, *Aerosol Sci. Technol.*, 25, 221–241, 1996.
- Blumberger, Z. I., Vasanits-Zsigrai, A., Farkas, G., and Salma, I.: Mass size distribution of major monosaccharide anhydrides and mass contribution of biomass burning, *Atmos. Res.*, 220, 1–9, 2019.
- Bonazza, A., Sabbioni, C., and Ghedini, N.: Quantitative data on carbon fractions in interpretation of black crusts and soiling on European built heritage, *Atmos. Environ.*, 39, 2607–2618, 2005.
- Bonvalot, L., Tuna, T., Fagault, Y., Jaffrezo, J.-L., Jacob, V., Chevrier, F., and Bard, E.: Estimating contributions from biomass burning, fossil fuel combustion, and biogenic carbon to carbonaceous aerosols in the Valley of Chamonix: a dual approach based on radiocarbon and levoglucosan, *Atmos. Chem. Phys.*, 16, 13753–13772, 2016.
- Burnett, R., Chen, H., Szyszkowicz, M., Fann, N., Hubbell, B., Pope, C. A., Apte, J. S., Brauer, M., Cohen, A., Weichenthal, S., Coggins, J., Di, Q., Brunekreef, B., Frostad, J., Lim, S. S., Kan, H., Walker, K. D., Thurston, G. D., Hayes, R. B., Lim, C. C., Turner, M. C., Jerrett, M., Krewski, D., Gapstur, S. M., Diver, W. R., Ostro, B., Goldberg, D., Crouse, D. L., Martin, R. V., Peters, P., Pinault, L., Tjepkema, M., van Donkelaar, A., Villeneuve, P. J., Miller, A. B., Yin, P., Zhou, M., Wang, L., Janssen, N. A. H., Marra, M., Atkinson, R. W., Tsang, H., Quoc Thach, T., Cannon, J. B., Allen, R. T., Hart, J. E., Laden, F., Cesaroni, G., Forastiere, F., Weinmayr, G., Jaensch, A., Nagel, G., Concin, H., and Spadaro, J. V.: Global estimates of mortality associated with long-term exposure to outdoor fine particulate matter, *P. Natl. Acad. Sci. USA*, 115, 9592–9597, 2018.

Burr, G. S. and Jull, A. J. T.: Accelerator mass spectrometry for radiocarbon research, In:
 Encyclopedia of mass spectrometry, Gross, M. L. and Caprioli, R. (eds.), Elsevier, Amsterdam,
 2009.

Caseiro, A., Bauer, H., Schmidl, C., Pio, C. A., and Puxbaum, H.: Wood burning impact on PM₁₀ in
 three Austrian regions, *Atmos. Environ.*, 43, 2186–2195, 2009.

Cecchini, M. A., Machado, L. A. T., Andreae, M. O., Martin, S. T., Albrecht, R. I., Artaxo, P.,
 Barbosa, H. M. J., Borrmann, S., Fütterer, D., Jurkat, T., Mahnke, C., Minikin, A., Molleker, S.,
 Pöhlker, M. L., Pöschl, U., Rosenfeld, D., Voigt, C., Weinzierl, B., and Wendisch, M.:
 Sensitivities of Amazonian clouds to aerosols and updraft speed, *Atmos. Chem. Phys.*, 17, 10037–
 10050, 2017.

Chen, J., Li, Ch., Ristovski, Z., Milic, A., Gu, Y., Islam, M. S., Wang, S., Hao, J., Zhang, H., He, C.,
 Guo, H., Fu, H., Miljevic, B., Morawska, L., Thai, P., Lam, Y. F., Pereira, G., Ding, A., Huang,
 X., and Dumka, U. C.: A review of biomass burning: Emissions and impacts on air quality, health
 and climate in China, *Sci. Total Environ.*, 579, 1000–1034, 2017.

Cirino, G. G., Souza, R. A. F., Adams, D. K., and Artaxo, P.: The effect of atmospheric aerosol
 particles and clouds on net ecosystem exchange in the Amazon, *Atmos. Chem. Phys.*, 14, 6523–
 6543, 2014.

Claeys, M., Kourtchev, I., Pashynska, V., Vas, G., Vermeylen, R., Wang, W., Cafmeyer, J., Chi, X.,
 Artaxo, P., Andreae, M. O., and Maenhaut, W.: Polar organic marker compounds in atmospheric
 aerosols during the LBA-SMOCC 2002 biomass burning experiment in Rondônia, Brazil: sources
 and source processes, time series, diel variations and size distributions, *Atmos. Chem. Phys.*, 10,
 9319–9331, 2010.

Fabbri, D., Torri, C., Simoneit, B. R. T., Marynowski, L., Rushdi, A. I., and Fabiańska, M. J.:
 Levoglucosan and other cellulose and lignin markers in emissions from burning of Miocene
 lignites, *Atmos. Environ.*, 43, 2286–2295, 2009.

Favez, O., El Haddad, I., Piot, C., Boréave, A., Abidi, E., Marchand, N., Jaffrezo, J.-L., Besombes, J.-
 L., Personnaz, M.-B., Sciare, J., Wortham, H., George, C., and D'Anna, B.: Inter-comparison of
 source apportionment models for the estimation of wood burning aerosols during wintertime in an
 Alpine city (Grenoble, France), *Atmos. Chem. Phys.*, 10, 5295–5314, 2010.

Fine, P. M., Cass, G. R., and Simoneit, B. R. T.: Chemical characterization of fine particle emissions
 from the fireplace combustion of wood types grown in the Midwestern and Western United States,
Environ. Eng. Sci., 21, 387–409, 2004.

Forello, A. C., Bernardoni, V., Calzolari, G., Lucarelli, F., Massabò, D., Nava, S., Pileci, R. E., Prati,
 P., Valentini, S., Valli, G., and Vecchi, R.: Exploiting multi-wavelength aerosol absorption
 coefficients in a multi-time resolution source apportionment study to retrieve source-dependent
 absorption parameters, *Atmos. Chem. Phys.*, 19, 11235–11252, 2019.

Fraser, M. P. and Lakshmanan, K.: Using levoglucosan as a molecular marker for the long range
 transport of biomass combustion aerosols, *Environ. Sci. Technol.*, 34, 4560–4564, 2000.

988 Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., Denier van der Gon, H., Facchini, M. C.,
 989 Fowler, D., Koren, I., Langford, B., Lohmann, U., Nemitz, E., Pandis, S., Riipinen, I., Rudich, Y.,
 990 Schaap, M., Slowik, J. G., Spracklen, D. V., Vignati, E., Wild, M., Williams, M., and Gilardoni,
 991 S.: Particulate matter, air quality and climate: lessons learned and future needs, *Atmos. Chem.*
 992 *Phys.*, 15, 8217–8299, 2015.

993 Gelencsér, A., May, B., Simpson, D., Sánchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Caseiro,
 994 A., Pio, C., and Legrand, M.: Source apportionment of PM_{2.5} organic aerosol over Europe:
 995 primary/secondary, natural/anthropogenic, and fossil/biogenic origin, *J. Geophys. Res.*, 112,
 996 D23S04, doi:10.1029/2006JD008094, 2007.

997 Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J.,
 998 Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T.,
 999 Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W.,
 1000 McFiggans, G., Mentel, Th. F., Monod, A., Prévôt, A. S. H., Seinfeld, J. H., Surratt, J. D.,
 1001 Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic
 1002 aerosol: current and emerging issues, *Atmos. Chem. Phys.*, 9, 5155–5236, 2009.

1003 Hays, M. D., Smith, N. D., Kinsey, J., Dong, Y., and Kariher, P.: Polycyclic aromatic hydrocarbon
 1004 size distributions in aerosols from appliances of residential wood combustion as determined by
 1005 direct thermal desorption-GC/MS, *J. Aerosol Sci.*, 34, 1061–1084, 2003.

1006 Heal, M. R., Naysmith, Ph., Cook, G. T. Xu, S., Duran, T. R., and Harrison, R. M.: Application of ¹⁴C
 1007 analyses to source apportionment of carbonaceous PM_{2.5} in the UK, *Atmos. Environ.*, 45,
 1008 2341–2348, 2011.

1009 Hennigan, C. J., Sullivan, A. P., Collett Jr., J. L., and Robinson, A. L.: Levoglucosan stability in
 1010 biomass burning particles exposed to hydroxyl radicals, *Geophys. Res. Lett.*, 37, L09806,
 1011 doi:10.1029/2010GL043088, 2010.

1012 Hoffmann, D., Tilgner, A., Iinuma, Y., and Herrmann, H.: Atmospheric stability of levoglucosan: A
 1013 detailed laboratory and modelling study, *Environ. Sci. Technol.*, 44, 694–699, 2010.

1014 Hopke, Ph. K.: Review of receptor modeling methods for source apportionment, *J. Air Waste Manag.*
 1015 *Assoc.*, 66, 237–259, 2016.

1016 Hoyle, C. R., Boy, M., Donahue, N. M., Fry, J. L., Glasius, M., Guenther, A., Hallar, A. G., Huff
 1017 Hartz, K., Petters, M. D., Petäjä, T., Rosenoern, T., and Sullivan, A. P.: A review of the
 1018 anthropogenic influence on biogenic secondary organic aerosol, *Atmos. Chem. Phys.*, 11, 321–
 1019 343, 2011.

1020 Ion, A. C., Vermeylen, R., Kourtev, I., Cafmeyer, J., Chi, X., Gelencsér, A., Maenhaut, W., and
 1021 Claeys, M.: Polar organic compounds in rural PM_{2.5} aerosols from K-puszt, Hungary, during a
 1022 2003 summer field campaign: Sources and diel variations, *Atmos. Chem. Phys.*, 5, 1805–1814,
 1023 2005.

1024 Janovics, R., Futó, I., and Molnár, M.: Sealed tube combustion method with MnO₂ for AMS ¹⁴C
 1025 measurement, *Radiocarbon*, 60, 1347–1355, 2018.

1026 Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Dingenen,
 1027 R. V., Ervens, B., Nenes, A., and Nielsen, C. J.: Organic aerosol and global climate modelling: a
 1028 review, *Atmos. Chem. Phys.*, 5, 1053–1123, 2005.

1029 Kirkby, J., Duplissy, J., Sengupta, K., Frege, C., Gordon, H., Williamson, C., Heinritzi, M., Simon,
 1030 M., Yan, C., Almeida, J., Tröstl, J., Nieminen, T., Ortega, I. K., Wagner, R., Adamov, A.,
 1031 Amorim, A., Bernhammer, A.-K., Bianchi, F., Breitenlechner, M., Brilke, S., Chen, X., Craven, J.,
 1032 Dias, A., Ehrhart, S., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Hakala, J., Hoyle, C. R.,
 1033 Jokinen, T., Junninen, H., Kangasluoma, J., Kim, J., Krapf, M., Kürten, A., Laaksonen, A.,
 1034 Lehtipalo, K., Makhmutov, V., Mathot, S., Molteni, U., Onnela, A., Peräkylä, O., Piel, F., Petäjä,
 1035 T., Praplan, A. P., Pringle, K., Rap, A., Richards, N. A. D., Riipinen, I., Rissanen, M. P., Rondo,
 1036 L., Sarnela, N., Schobesberger, S., Scott, C. E., Seinfeld, J. H., Sipilä, M., Steiner, G., Stozhkov,
 1037 Y., Stratmann, F., Tomé, A., Virtanen, A., Vogel, A. L., Wagner, A., Wagner, P. E., Weingartner,
 1038 E., Wimmer, D., Winkler, P. M., Ye, P., Zhang, X., Hansel, A., Dommen, J., Donahue, N. M.,
 1039 Worsnop, D. R., Baltensperger, U., Kulmala, M., Carslaw, K. S., and Curtius, J.: Ion-induced
 1040 nucleation of pure biogenic particles, *Nature*, 533, 521–526, 2016.

1041 Kiss, G., Varga, B., Galambos, I., and Ganszky, I.: Characterization of water-soluble organic matter
 1042 isolated from atmospheric fine aerosol, *J. Geophys. Res.*, 107(D21), 8339,
 1043 doi:10.1029/2001JD000603. 2002.

1044 Kourtchev, I., Hellebust, S., Bell, J. M., O'Connor, I. P., Healy, R. M., Allanic, A., Healy, D.,
 1045 Wenger, J. C., and Sodeau, J. R.: The use of polar organic compounds to estimate the contribution
 1046 of domestic solid fuel combustion and biogenic sources to ambient levels of organic carbon and
 1047 PM_{2.5} in Cork Harbour, Ireland, *Sci. Total Environ.*, 409, 2143–2155, 2011.

1048 Kuhlbusch, Th. A. J., Borowiak, A., Gelencsér, A., Genberg, J., Gladtké, D., Maenhaut, W., Pio, C.,
 1049 Popovicheva, O., Putaud, J. P., Quincey, P., Sciare, J., ten Brink, H., Viana, M., and Yttri, K. E.:
 1050 Measurement of elemental and organic carbon in Europe, JRC Scientific and Technical Reports,
 1051 European Commission, Joint Research Centre, Ispra, 2009.

1052 Lelieveld, J. and Pöschl, U.: Chemists can help to solve the air-pollution health crisis, *Nature*, 551,
 1053 291–293, 2017.

1054 Le Quéré, C., Andrew, R. M., Friedlingstein, P., Sitch, S., Hauck, J., Pongratz, J., Pickers, P. A.,
 1055 Korsbakken, J. I., Peters, G. P., Canadell, J. G., Arneeth, A., Arora, V. K., Barbero, L., Bastos, A.,
 1056 Bopp, L., Chevallier, F., Chini, L. P., Ciais, P., Doney, S. C., Gkritzalis, T., Goll, D. S., Harris, I.,
 1057 Haverd, V., Hoffman, F. M., Hoppema, M., Houghton, R. A., Hurtt, G., Ilyina, T., Jain, A. K.,
 1058 Johannessen, T., Jones, C. D., Kato, E., Keeling, R. F., Goldewijk, K. K., Landschützer, P.,
 1059 Lefèvre, N., Lienert, S., Liu, Z., Lombardozzi, D., Metzl, N., Munro, D. R., Nabel, J. E. M.
 1060 S., Nakaoka, S.-I., Neill, C., Olsen, A., Ono, T., Patra, P., Peregon, A., Peters, W., Peylin, P., Pfeil,
 1061 B., Pierrot, D., Poulter, B., Rehder, G., Resplandy, L., Robertson, E., Rocher, M., Rödenbeck, C.,
 1062 Schuster, U., Schwinger, J., Séférian, R., Skjelvan, I., Steinhoff, T., Sutton, A., Tans, P. P., Tian,
 1063 H., Tilbrook, B., Tubiello, F. N., van der Laan-Luijkx, I. T., van der Werf, G. R., Viovy, N.,

1064 Walker, A. P., Wiltshire, A. J., Wright, R., Zaehle, S., and Zheng, B.: Global carbon budget 2018,
 1065 Earth Syst. Sci. Data, 10, 2141–2194, 2018.
 1066 Lohmann, U., Feichter, J., Penner, J., and Leaitch, R.: Indirect effect of sulfate and carbonaceous
 1067 aerosols: A mechanistic treatment, J. Geophys. Res., 105, 12193–12206, 2000.
 1068 Maenhaut, W., Raes, N., Chi, X., Cafmeyer, J., Wang, W., and Salma, I.: Chemical composition and
 1069 mass closure for fine and coarse aerosols at a kerbside in Budapest, Hungary, in spring 2002, X-
 1070 ray Spectrom., 34, 290–296, 2005.
 1071 Maenhaut, W., Raes, N., Chi, X., Cafmeyer, J., and Wang, W.: Chemical composition and mass
 1072 closure for PM_{2.5} and PM₁₀ aerosols at K-pusztá, Hungary, in summer 2006, X-ray Spectrom., 37,
 1073 193–197, 2008.
 1074 Maenhaut, W., Vermeylen, R., Claeys, M., Vercauteren, J., Matheeuissen, C., and Roekens, E.:
 1075 Assessment of the contribution from wood burning to the PM₁₀ aerosol in Flanders, Belgium, Sci.
 1076 Total Environ., 437, 226–236, 2012.
 1077 Maenhaut, W., Vermeylen, R., Claeys, M., Vercauteren, J., and Roekens, E.: Sources of the PM₁₀
 1078 aerosol in Flanders, Belgium, and re-assessment of the contribution from wood burning, Sci.
 1079 Total Environ., 562, 550–560, 2016.
 1080 Major, I., Gyökös, B., Túri, M., Futó, I., Filep, Á., Hoffer, A., Furu, E., Jull, A. J. T., Molnár, M.:
 1081 Evaluation of an automated EA-IRMS method for total carbon analysis of atmospheric aerosol at
 1082 HEKAL, J. Atmos. Chem., 75, 85–96, 2018.
 1083 McFiggans, G., Mentel, T. F., Wildt, J., Pullinen, I., Kang, S., Kleist, E., Schmitt, S., Springer, M.,
 1084 Tillmann, R., Wu, C., Zhao, D., Hallquist, M., Faxon, C., Le Breton, M., Hallquist, A. M.,
 1085 Simpson, D., Bergstroem, R., Jenkin, M. E., Ehn, M., Thornton, J. A., Alfarra, M. R., Bannan, T.
 1086 J., Percival, C. J., Priestley, M., Topping, D., and Kiendler-Scharr, A.: Secondary organic aerosol
 1087 reduced by mixture of atmospheric vapours, Nature, 565, 587–593, 2019.
 1088 Minguillón, M. C., Perron, N., Querol, X., Szidat, S., Fahrni, S. M., Alastuey, A., Jimenez, J. L.,
 1089 Mohr, C., Ortega, A. M., Day, D. A., Lanz, V. A., Wacker, L., Reche, C., Cusack, M., Amato, F.,
 1090 Kiss, G., Hoffer, A., Decesari, S., Moretti, F., Hillamo, R., Teinilä, K., Seco, R., Peñuelas, J.,
 1091 Metzger, A., Schallhart, S., Müller, M., Hansel, A., Burkhardt, J. F., Baltensperger, U., and Prévôt,
 1092 A. S. H.: Fossil versus contemporary sources of fine elemental and organic carbonaceous
 1093 particulate matter during the DAURE campaign in Northeast Spain, Atmos. Chem. Phys., 11,
 1094 12067–12084, 2011.
 1095 Molnár, M., Rinyu, L., Veres, M., Seiler, M., Wacker, L., and Synal, H.-A.: EnvironMICADAS: a
 1096 mini ¹⁴C AMS with enhanced gas ion source interface in the Hertelendi Laboratory of
 1097 Environmental Studies, Hungary, Radiocarbon, 55, 338–344, 2013.
 1098 Nolte, C. G., Schauer, J. J., Cass, G. R., and Simoneit, B. R. T.: Highly polar organic compounds
 1099 present in wood smoke and in the ambient atmosphere, Environ. Sci. Technol., 35, 1912–1919,
 1100 2001.

1101 Nozière, B., Kalberer, M., Claeys, M., Allan, J., D'Anna, B., Decesari, S., Finessi, E., Glasius, M.,
 1102 Grgić, I., Hamilton, J. F., Hoffmann, T., Iinuma, Y., Jaoui, M., Kahnt, A., Kampf, C. J.,
 1103 Kourtchev, I., Maenhaut, W., Marsden, N., Saarikoski, S., Schnelle-Kreis, J., Surratt, J. D., Szidat,
 1104 S., Szmigielski, R., and Wisthaler, A.: The molecular identification of organic compounds in the
 1105 atmosphere: state of the art and challenges, *Chem. Rev.*, 115, 3919–3983, 2015.
 1106 Panteliadis, P., Hafkenscheid, T., Cary, B., Diapouli, E., Fischer, A., Favez, O., Quincey, P., Viana,
 1107 M., Hitzenberger, R., Vecchi, R., Saraga, D., Sciare, J., Jaffrezo, J. L., John, A., Schwarz, J.,
 1108 Giannoni, M., Novak, J., Karanasiou, A., Fermo, P., and Maenhaut, W.: ECOC comparison
 1109 exercise with identical thermal protocols after temperature offset correction – instrument
 1110 diagnostics by in-depth evaluation of operational parameters, *Atmos. Meas. Tech.*, 8, 779–792,
 1111 2015.
 1112 Pashynska, V., Vermeylen, R., Vas, G., Maenhaut, W., and Claeys, M.: Development of a gas
 1113 chromatography/ion trap mass spectrometry method for the determination of levoglucosan and
 1114 saccharidic compounds in atmospheric aerosols, Application to urban aerosols, *J. Mass Spectrom.*,
 1115 37, 1249–125, 2002.
 1116 Piazzalunga, A., Belis, C., Bernardoni, V., Cazzuli, O., Fermo, P., Valli, G., and Vecchi, R.: Estimates
 1117 of wood burning contribution to PM by the macro-tracer method using tailored emission factors,
 1118 *Atmos. Environ.*, 45, 6642–6649, 2011.
 1119 Putaud, J.-P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Flentje, H., Fuzzi, S.,
 1120 Gehrig, R., Hansson, H. C., Harrison, R. M., Herrmann, H., Hitzenberger, R., Hügl, C., Jones,
 1121 A.M., Kasper-Giebl, A., Kiss, G., Koussa, A., Kuhlbusch, T. A. J., Löschau, G., Maenhaut, W.,
 1122 Molnár, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz, M., Puxbaum, H., Querol, X., Rodriguez,
 1123 S., Salma, I., Schwarz, J., Smolik, J., Schneider, J., Spindler, G., ten Brink, H., Tursic, J., Viana,
 1124 M., Wiedensohler, A., and Raes, F.: A European aerosol phenomenology - 3: physical and
 1125 chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across
 1126 Europe, *Atmos. Environ.*, 44, 1308–1320, 2010.
 1127 Puxbaum, H., Caseiro, A., Sanchez-Ochoa, A., Kasper-Giebl, A., Claeys, M., Gelencsér, A., Legrand,
 1128 M., Preunkert, S., and Pio, C.: Levoglucosan levels at background sites in Europe for assessing the
 1129 impact of biomass combustion on the European aerosol background, *J. Geophys. Res.-Atmos.*,
 1130 112, D23S05, doi:10.1029/2006JD008114, 2007.
 1131 Rap, A., Spracklen, D. V., Mercado, L., Reddington, C. L., Haywood, J. M., Ellis, R. J., Phillips, O.
 1132 L., Artaxo, P., Bonal, D., Coupe, N. R., and Butt, N.: Fires increase Amazon forest productivity
 1133 through increases in diffuse radiation, *Geophys. Res. Lett.*, 42, 4654–4662, 2015.
 1134 Rosenfeld, D., Zhu, Y., Wang, M., Zheng, Y., Goren, T., and Yu, S.: Aerosol-driven droplet
 1135 concentrations dominate coverage and water of oceanic low-level clouds, *Science*, 363, eaav0566,
 1136 DOI:10.1126/science.aav0566, 2019.
 1137 Russell, L. M.: Aerosol organic-mass-to-organic-carbon ratio measurements, *Environ. Sci. Technol.*,
 1138 37, 2982–2987, 2003.

1139 Saarikoski, S., Timonen, H., Saarnio, K., Aurela, M., Järvi, L., Keronen, P., Kerminen, V.-M., and
 1140 Hillamo, R.: Sources of organic carbon in fine particulate matter in northern European urban air,
 1141 *Atmos. Chem. Phys.*, 8, 6281–6295, 2008.

1142 Saarnio, K., Niemi, J. V., Saarikoski, S., Aurela, M., Timonen, H., Teinilä, K., Myllynen, M., Frey,
 1143 A., Lamberg, H., Jokiniemi, J., and Hillamo, R.: Using monosaccharide anhydrides to estimate the
 1144 impact of wood combustion on fine particles in the Helsinki Metropolitan Area, *Boreal Environ.*
 1145 *Res.*, 17, 163–183, 2012.

1146 Salma, I. and Maenhaut, W.: Changes in chemical composition and mass of atmospheric aerosol
 1147 pollution between 1996 and 2002 in a Central European city, *Environ. Pollut.*, 143, 479–488,
 1148 2006.

1149 ~~Salma, I. and Németh, Z.: Dynamic and timing properties of new aerosol particle formation and~~
 1150 ~~consecutive growth events, *Atmos. Chem. Phys.*, 19, 5835–5852, 2019.~~

1151 Salma, I., Maenhaut, W., Dubtsov, S., Zemplén-Papp, É., and Záray, Gy.: Impact of phase out of
 1152 leaded gasoline on the air quality in Budapest, *Microchem. J.*, 67, 127–133, 2000.

1153 Salma, I., Chi, X., and Maenhaut, W.: Elemental and organic carbon in urban canyon and background
 1154 environments in Budapest, Hungary, *Atmos. Environ.*, 38, 27–36, 2004.

1155 Salma, I., Ocskay, R., Chi, X., and Maenhaut, W.: Sampling artefacts, concentrations and chemical
 1156 composition of fine water-soluble organic carbon and humic-like substances in a continental
 1157 urban atmospheric environment, *Atmos. Environ.*, 41, 4106–4118, 2007.

1158 Salma, I., Mészáros, T., and Maenhaut, W.: Mass size distribution of carbon in atmospheric humic-
 1159 like substances and water-soluble organic carbon for an urban environment, *J. Aerosol Sci.*, 56,
 1160 53–60, 2013.

1161 Salma, I., Németh, Z., Weidinger, T., Kovács, B., and Kristóf, G.: Measurement, growth types and
 1162 shrinkage of newly formed aerosol particles at an urban research platform, *Atmos. Chem. Phys.*,
 1163 16, 7837–7851, 2016a.

1164 Salma, I., Németh, Z., Kerminen, V. M., Aalto, P., Nieminen, T., Weidinger, T., Molnár, Á., Imre, K.,
 1165 and Kulmala, M.: Regional effect on urban atmospheric nucleation, *Atmos. Chem. Phys.*, 16,
 1166 8715–8728, 2016b.

1167 Salma, I., Németh, Z., Weidinger, T., Maenhaut, W., Claeys, M., Molnár, M., Major, I., Ajtai, T.,
 1168 Utry, N., and Bozóki, Z.: Source apportionment of carbonaceous chemical species to fossil fuel
 1169 combustion, biomass burning and biogenic emissions by a coupled radiocarbon–levoglucosan
 1170 marker method, *Atmos. Chem. Phys.*, 17, 13767–13781, 2017.

1171 Sandradewi, J., Prévôt, A. S. H., Szidat, S., Perron, N., Alfara, R. M., Lanz, V. A., Weingartner, E.,
 1172 and Baltensperger, U.: Using aerosol light absorption measurements for the quantitative
 1173 determination of wood burning and traffic emission contributions to particulate matter, *Environ.*
 1174 *Sci. Technol.*, 42, 3316–3323, 2008a.

1175 Sandradewi, J., Prévôt, A. S. H., Weingartner, E., Schmidhauser, R., Gysel, M., and Baltensperger,
 1176 U.: A study of wood burning and traffic aerosols in an Alpine valley using a multi-wavelength
 1177 Aethalometer, *Atmos. Environ.*, 42, 101–112, 2008b.

1178 Schmidl, C., Marr, L. L., Caseiro, A., Kotianova, P., Berner, A., Bauer, H., Kasper-Giebl, A., and
 1179 Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of
 1180 common woods growing in mid-European Alpine regions, *Atmos. Environ.*, 42, 126–141, 2008.

1181 Simoneit, B. R. T., Schauer, J. J., Nolte, C. G., Oros, D. R., Elias, V. O., Fraser, M. P., Rogge, W. F.,
 1182 and Cass, G. R.: Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles,
 1183 *Atmos. Environ.*, 33, 173–182, 1999.

1184 Simoneit, B. R. T., Elias, V. O., Kobayashi, M., Kawamura, K., Rushdi, A. I., Medeiros, P. M.,
 1185 Rogge, W. F., and Didyk, B. M.: Sugars-dominant water-soluble organic compounds in soils and
 1186 characterization as tracers in atmospheric particulate matter, *Environ. Sci. Technol.*, 38, 5939–
 1187 5949, 2004.

1188 Stefanelli, G., Pospisilova, V., Lopez-Hilfiker, F. D., Daellenbach, K. R., Hüglin, C., Tong, Y.,
 1189 Baltensperger, U., Prévôt, A. S. H., and Slowik, J. G.: Organic aerosol source apportionment in
 1190 Zurich using an extractive electrospray ionization time-of-flight mass spectrometer (EESI-TOF-
 1191 MS) – Part 1: Biogenic influences and day–night chemistry in summer, *Atmos. Chem. Phys.*, 19,
 1192 14825–14848, 2019.

1193 Swietlicki, E., Hansson, H. C., Hämeri, K., Svenningsson, B., Massling, A., McFiggans, G.,
 1194 McMurry, P. H., Petäjä, T., Tunved, P., Gysel, M., Topping, D., Weingartner, E., Baltensperger,
 1195 U., Rissler, J., Wiedensohler, A., and Kulmala, M.: Hygroscopic properties of submicrometer
 1196 atmospheric aerosol particles measured with H-TDMA instruments in various environments – a
 1197 review, *Tellus B*, 60, 432–469, 2008.

1198 Szidat, S., Jenk, T. M., Synal, H. A., Kalberer, M., Wacker, L., Hajdas, I., Kasper-Giebl, A., and
 1199 Baltensperger, U.: Contributions of fossil fuel, biomass-burning, and biogenic emissions to
 1200 carbonaceous aerosols in Zurich as traced by ^{14}C , *J. Geophys. Res.*, 111, D07206.
 1201 doi:10.1029/2005JD006590, 2006.

1202 Szidat, S., Ruff, M., Perron, N., Wacker, L., Synal, H.-A., Hallquist, M., Shannigrahi, A. S., Yttri, K.
 1203 E., Dye, C., and Simpson, D.: Fossil and non-fossil sources of organic carbon (OC) and elemental
 1204 carbon (EC) in Göteborg, Sweden, *Atmos. Chem. Phys.*, 9, 1521–1535, 2009.

1205 Tatai, Zs., Szőke, B., and Körmendi, K.: Budapest zöldinfrastruktúra koncepciója (Concept on green
 1206 infrastructure of Budapest, in Hungarian), Municipality of Budapest, BFVT Kft., 2017.

1207 Tian, H., Lu, C., Ciais, P., Michalak, A. M., Canadell, J. G., Saikawa, E., Huntzinger, D. N., Gurney,
 1208 K. R., Sitch, S., Zhang, B., Yang, J., Bousquet, P., Bruhwiler, L., Chen, G., Dlugokencky, E.,
 1209 Friedlingstein, P., Melillo, J., Pan, S., Poulter, B., Prinn, R., Saunois, M., Schwalm, C. R., and
 1210 Wofsy, S. C.: The terrestrial biosphere as a net source of greenhouse gases to the atmosphere,
 1211 *Nature*, 531, 225–228, 2016.

- Turpin, B. J. and Lim, H.-J.: Species contributions to PM_{2.5} mass concentrations: revisiting common assumptions for estimating organic mass, *Aerosol Sci. Technol.*, 35, 602–610, 2001.
- [Turpin, B. J., Saxena, P., and Andrews, E.: Measuring and simulating particulate organics in the atmosphere: problems and prospects, *Atmos. Environ.*, 34, 2983–3013, 2000.](#)
- Viana, M., Chi, X., Maenhaut, W., Cafmeyer, J., Querol, X., Alastuey, A., Mikuška, P., and Večeřa, Z.: Influence of sampling artefacts on measured PM, OC, and EC levels in carbonaceous aerosols in an urban area, *Aerosol Sci. Technol.*, 40, 107–117, 2006.
- Vicente, E. D. and Alves, C. A.: An overview of particulate emissions from residential biomass combustion, *Atmos. Res.*, 199, 159–185, 2018.
- Von Schneidmesser, E., Monks, P. S., Allan, J. D., Bruhwiler, L., Forster, P., Fowler, D., Lauer, A., Morgan, W. T., Paasonen, P., Righi, M., Sindelarova, K., and Sutton, M. A.: Chemistry and the linkages between air quality and climate change, *Chem. Rev.*, 115, 3856–3897, 2015.
- Wacker, L., Bonani, G., Friedrich, M., Hajdas, I., Kromer, B., Némec, M., Ruff, M., Suter, M., Synal, H.-A., and Vockenhuber, C.: MICADAS: Routine and high-precision radiocarbon dating, *Radiocarbon*, 52, 252–262, 2010.
- Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The fire inventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, *Geosci. Model Dev.*, 4, 625–641, 2011.
- Yttri, K. E., Schnelle-Kreis, J., Maenhaut, W., Abbaszade, G., Alves, C., Bjerke, A., Bonnier, N., Bossi, R., Claeys, M., Dye, C., Evtyugina, M., García-Gacio, D., Hillamo, R., Hoffer, A., Hyder, M., Iinuma, Y., Jaffrezo, J.-L., Kasper-Giebl, A., Kiss, G., López-Mahia, P. L., Pio, C., Piot, C., Ramirez-Santa-Cruz, C., Sciare, J., Teinilä, K., Vermeylen, R., Vicente, A., and Zimmermann, R.: An intercomparison study of analytical methods used for quantification of levoglucosan in ambient aerosol filter samples, *Atmos. Meas. Tech.*, 8, 125–147, 2015.
- Zdráhal, Z., Oliveira, J., Vermeylen, R., Claeys, M., and Maenhaut, W.: Improved method for quantifying levoglucosan and related monosaccharide anhydrides in atmospheric aerosols and application to samples from urban and tropical locations, *Environ. Sci. Technol.*, 36, 747–753, 2002.
- Zhang, Y. L., Perron, N., Ciobanu, V. G., Zotter, P., Minguillón, M. C., Wacker, L., Prévôt, A. S. H., Baltensperger, U., and Szidat, S.: On the isolation of OC and EC and the optimal strategy of radiocarbon-based source apportionment of carbonaceous aerosols, *Atmos. Chem. Phys.*, 12, 10841–10856, 2012.
- Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S., and Prévôt, A. S. H.: Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol, *Atmos. Chem. Phys.*, 17, 4229–4249, 2017.

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

Imre SALMA¹, Anikó VASANITS-ZSIGRAI¹, Attila MACHON², Tamás VARGA³,
István MAJOR³, Virág GERGELY³, Mihály MOLNÁR³

¹ Institute of Chemistry, Eötvös University, Budapest, Hungary

² Air Quality Reference Center, Hungarian Meteorological Service, Budapest, Hungary

³ Isotope Climatology and Environmental Research Centre, Institute for Nuclear Research, Debrecen, Hungary

Correspondence to: Imre Salma (salma@chem.elte.hu)

Table S1. Mean apportionment multiplication factors of the coupled radiocarbon-levoglucosan marker model with SDs for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Factor	Site type	October	January	April	July
f_1 (%)	Region	69±3	75±3	63±13	74±9
	Suburb	68±6	76±7	50±6	57±11
	Centre	76±10	74±2	50±9	58±7
f_2 (%)	Region	7.6±1.3	14±1	1.6±0.5	0.41±0.15
	Suburb	8.3±1.7	15±2	3.5±1.1	0.50±0.24
	Centre	6.0±1.0	13±1	2.7±1.1	0.61±0.18
f_3 (%)	Region	49±9	93±6	10±3	2.4±1.0
	Suburb	53±12	97±16	21±7	2.9±1.5
	Centre	38±7	90±8	16±7	3.6±1.1
f_4 (%)	Region	28±14	5.3±2.4	35±13	24±14
	Suburb	14±11	6.8±2.0	36±8	33±5
	Centre	29±17	19±4	45±12	31±6

Table S2. Median atmospheric concentrations of SO₂, NO, NO₂, CO, O₃ and PM₁₀ mass for regional background in the Carpathian Basin, suburban area and city centre of Budapest for the different months of seasons.

Pollutant	Site type	October	January	April	July
SO ₂ (µg m ⁻³)	Region	0.44	n.a.	n.a.	n.a.
	Suburb	2.2	2.0	1.35	1.50
	Centre	7.0	5.3	4.7	2.7
NO (µg m ⁻³)	Region	n.a.	n.a.	n.a.	n.a.
	Suburb	8.4	4.7	1.40	1.05
	Centre	74	51	17.3	10.6
NO ₂ (µg m ⁻³)	Region	2.9	n.a.	n.a.	n.a.
	Suburb	36	25	19.3	12.8
	Centre	52	37	57	31
CO (mg m ⁻³)	Region	n.a.	n.a.	n.a.	n.a.
	Suburb	0.50	0.60	0.78	0.69
	Centre	0.69	0.56	0.61	0.33
O ₃ (µg m ⁻³)	Region	n.a.	37	103	92
	Suburb	12.4	15.5	67	74
	Centre	3.4	2.7	38	55
PM ₁₀ mass (µg m ⁻³)	Region	14.5	n.a.	n.a.	n.a.
	Suburb	30	29	39	21
	Centre	38	41	42	23

n.a.: not available

Table S3. Mean atmospheric concentrations and SDs of SO₂, NO, NO₂, CO, O₃ and PM₁₀ mass for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Pollutant	Site type	October	January	April	July
SO ₂ (µg m ⁻³)	Region	0.67±0.38	n.a.	n.a.	n.a.
	Suburb	2.4±0.5	2.1±0.7	1.35±0.60	1.55±0.35
	Centre	7.0±0.2	5.8±0.9	4.7±0.8	2.7±0.1
NO (µg m ⁻³)	Region	n.a.	n.a.	n.a.	n.a.
	Suburb	12.7±10.7	11.1±10.1	1.44±0.75	1.08±0.46
	Centre	75±16	50±17	17.6±3.2	10.2±2.3
NO ₂ (µg m ⁻³)	Region	3.1±1.4	n.a.	n.a.	n.a.
	Suburb	36±20	31±12	18.9±8.7	14.9±7.3
	Centre	50±8	39±6	52±11	32±7
CO (mg m ⁻³)	Region	n.a.	n.a.	n.a.	n.a.
	Suburb	0.48±0.20	0.66±0.18	0.82±0.13	0.61±0.17
	Centre	0.68±0.09	0.60±0.09	0.61±0.07	0.34±0.06
O ₃ (µg m ⁻³)	Region	n.a.	41±21	96±17	90±11
	Suburb	21±18	16.7±13.1	67±12	71±13
	Centre	4.0±2.0	3.5±2.1	40±11	56±9
PM ₁₀ mass (µg m ⁻³)	Region	17.4±12.7	n.a.	n.a.	n.a.
	Suburb	36±25	30±15	35±11	24±6
	Centre	40±13	40±12	42±9	27±7

n.a.: not available

Table S4. Mean air temperature (T), relative humidity (RH), wind speed (WS) and daily maximum global solar radiation (GRad_{max}) with SDs for regional background in the Carpathian Basin, suburban area and city centre of Budapest during the aerosol sampling campaign for different months of seasons.

Property	Site type	October	January	April	July
T (°C)	Region	8.4±1.9	1.8±2.9	17.1±2.6	23±2
	Suburb	10.0±2.2	2.5±3.3	19.0±2.0	24±2
	Centre	12.9±1.9	2.5±3.5	19.9±1.0	24±2
RH (%)	Region	88±10	89±10	64±8	78±9
	Suburb	75±11	79±8	48±9	56±8
	Centre	79±4	82±4	49±9	64±9
WS (m s ⁻¹)	Region	1.61±1.14	1.52±0.71	1.45±0.57	1.55±0.72
	Suburb	1.64±1.10	1.63±0.49	1.27±0.23	1.32±0.68
	Centre	1.82±0.84	2.1±0.4	2.5±0.5	3.9±1.7
GRad_{max} (kW m ⁻²)	Region	n.a.	n.a.	n.a.	n.a.
	Suburb	0.31±0.15	0.186±0.106	0.77±0.09	0.79±0.11
	Centre	n.a.	n.a.	0.74±0.11	0.76±0.15

n.a.: not available

The differences in the mean WS can be explained by non-equal heights of the meteorological sensors (3 m above the ground in the regional background and at the suburban area, and 12 m above the street level in the city centre).

Table S5. Mean atmospheric concentrations and SDs 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), 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 ($\mu\text{g m}^{-3}$)	Region	16.3±8.3	18.4±6.4	9.7±2.7	10.5±1.7
	Suburb	24±13	25±9	10.4±2.1	11.6±2.8
	Centre	29±11	22±7	13.5±3.1	8.8±1.6
EC ($\mu\text{g m}^{-3}$)	Region	0.41±0.24	0.37±0.13	0.21±0.06	0.131±0.043
	Suburb	0.53±0.30	0.73±0.26	0.51±0.19	0.41±0.16
	Centre	1.08±0.37	0.77±0.22	0.84±0.22	0.40±0.10
OC ($\mu\text{g m}^{-3}$)	Region	3.0±1.8	3.4±1.5	2.1±0.5	2.3±0.4
	Suburb	4.9±3.2	5.4±2.4	2.4±0.5	2.7±0.4
	Centre	6.4±1.9	4.1±1.1	2.8±0.8	2.6±0.4
WSOC ($\mu\text{g m}^{-3}$)	Region	2.1±1.4	2.1±1.0	1.13±0.30	1.62±0.29
	Suburb	2.6±1.7	3.4±1.5	1.27±0.34	2.0±0.3
	Centre	2.8±1.4	2.6±0.7	1.61±0.53	2.0±0.4
LVG ($\mu\text{g m}^{-3}$)	Region	0.189±0.134	0.44±0.21	0.0020±0.0013	0.0081±0.0022
	Suburb	0.34±0.23	0.74±0.44	0.048±0.029	0.0151±0.0100
	Centre	0.38±0.06	0.49±0.15	0.049±0.024	0.0113±0.0027
MAN (ng m^{-3})	Region	19.2±5.7	24±14	2.6±0.2	<1.2
	Suburb	32±23	46±25	3.4±1.7	<1.2
	Centre	29±8	23±7	5.5±2.6	<1.2
GAN (ng m^{-3})	Region	n.a.	11.9±6.0	1.27±0.25	<0.5
	Suburb	14.1±10.1	22±11	1.47±0.84	<0.5
	Centre	12.3±2.7	14.1±3.9	1.32±0.35	<0.5
f_c (%)	Region	69±3	74±3	63±13	74±9
	Suburb	68±6	76±7	50±6	57±11
	Centre	76±6	74±2	50±9	59±7
K ($\mu\text{g m}^{-3}$)	Region	0.188±0.071	0.26±0.09	0.107±0.043	0.081±0.003
	Suburb	0.22±0.11	0.28±0.14	0.105±0.031	0.075±0.016
	Centre	0.27±0.07	0.28±0.05	0.112±0.031	0.057±0.013
Ni (ng m^{-3})	Region	1.01±0.63	0.71±0.13	1.31±0.32	1.23±0.48
	Suburb	0.97±0.30	0.98±0.59	1.26±0.21	1.35±0.54
	Centre	1.25±0.36	0.86±0.47	1.60±0.23	1.08±0.13
Pb (ng m^{-3})	Region	3.9±0.9	4.1±2.4	3.5±1.9	3.9±3.0
	Suburb	6.4±3.0	7.7±3.9	4.8±1.8	4.5±2.9
	Centre	6.7±1.6	5.5±2.5	5.2±1.9	2.4±1.5

Table S6. Coefficients of correlation between EC_{FF}, OC_{FF}, EC_{BB}, OC_{BB} and OC_{BIO} on the one side and K, NO, and *T* on the other side for regional background in the Carpathian Basin, suburban area and city centre of Budapest.

Variable (unit)	Site type	EC _{FF} (µg m ⁻³)	OC _{FF} (µg m ⁻³)	EC _{BB} (µg m ⁻³)	OC _{BB} (µg m ⁻³)	OC _{BIO} (µg m ⁻³)
K (µg m ⁻³)	Region	0.02	0.73	0.95	0.95	-0.26
	Suburb	-0.69	0.51	0.90	0.91	0.20
	Centre	-0.22	0.44	0.86	0.86	0.20
NO (µg m ⁻³)	Region	n.a.	n.a.	n.a.	n.a.	n.a.
	Suburb	-0.36	0.93	0.60	0.60	0.42
	Centre	-0.17	0.39	0.76	0.77	0.44
<i>T</i> (K)	Region	-0.37	-0.58	-0.87	-0.85	0.60
	Suburb	0.59	-0.24	-0.85	-0.87	0.24
	Centre	0.53	-0.08	-0.79	-0.80	0.34

Table S7. Mean contributions with SDs of fossil fuel (FF) combustion, biomass burning (BB), biogenic sources (Bio) and unaccounted part (UnA) to the PM_{2.5} mass in % as rough estimates for regional background in the Carpathian Basin, suburban area and city centre of Budapest for different months of seasons.

Environment	Source	October	January	April	July
Regional background	FF	12±3	8±2	13±6	8±3
	BB	14±3	21±3	3±1	<2
	Bio	14±5	<2	23±5	24±4
	UnA	60±8	70±4	61±8	67±3
Suburban area	FF	14±5	8±3	20±5	15±4
	BB	16±6	23±3	5±3	<2
	Bio	13±5	3±2	16±4	24±3
	UnA	58±13	67±4	59±9	61±5
City centre	FF	9±5	9±2	18±6	20±4
	BB	11±2	22±4	4±2	<2
	Bio	16±3	3±1	18±6	31±6
	UnA	63±3	67±4	60±8	48±6