

## 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 PM<sub>10</sub> 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  $^{14}\text{C}$  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<sub>BIO</sub>? I would have expected to see highest OC<sub>BIO</sub> concentrations in summer.. or is there a reason why autumn OC<sub>BIO</sub> 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<sub>BIO</sub> (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  $OC_{\text{BIO}}$  in the regional background and insignificant dependencies at the urban sites. The links between them are rather complex and not fully uncovered. The  $OC_{\text{BIO}}$  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.

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