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 cites 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 cites 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 PM_{10} 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 studied 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.

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