Response to Referee #2

The authors thank Referee #2 for his/her detailed and valuable comments to further improve and clarify the MS. We have considered all recommendations, and made the appropriate alterations. Our specific responses to the comments are as follows.

Comments

1) Main added uncertainty to the classical method of radiocarbon analysis with the split of EC and OC before the analysis is included in predefined value of ECBB to OCBB ratio. Nevertheless, the authors do not comment uncertainty connected with this value at all. Moreover, although they found that one of the samples was probably contaminated by artificial 14C release, they omitted to discuss a possibility of such influence on the other samples.

A separate discussion part of the uncertainties connected with the apportionment method is now added to the text with a sensitivity calculation. We also like to indicate that the contamination occurred usually (and predominantly) for the back filters, and therefore, it may have an effect (be it limited) on the final uncertainty if it is not properly realised.

2) Finally, although the authors say that their OCBIO is in line with Simpson et al. (2007) the opposite is true due to different definition of fractions. The observed result was 0.17 μ g/m3 of OCBIO in K-puszta for winter (primary biogenic particles), which was roughly 0.2% of TC in PM10 and ca 20% of TC was attributed to non-fossil SOA. Therefore the reviewer suggests to call the last OC fraction same way (non-fossil SOA), also with regard to the text on page 12, line 44.

We decided finally to remove this part of the sentence to avoid discussions on primary and secondary particles, and indicated briefly the alternative attitude in the text.

3) The authors also spend quite a lot of space describing aethalometer and other on- line instrumentation results but they did not use an opportunity to test their method with aethalometer source apportionment method.

The authors regret that they did not indicate to the handling editor and the potential referees at the submission stage that another MS, which deals with and evaluates the data from the on-line measurements obtained by the AE, PAS and DMPS in focus, is under preparation. This other MS will include the evaluation methods referred to by the Referee, thus the so-called Aethalometer model (the wavelength dependence of the optical absorption coefficient) for both the AE (Sandradewi et al., Environ. Sci. Technol., 42, 3316–3323, 2008; Sandradewi et al.,

Atmos. Environ., 42, 101–112, 2008; Favez et al., Atmos. Environ., 43, 3640–3644, 2009; Favez et al., Atmos. Chem. Phys., 10, 5295–5314, 2010) and PAS–DMPS (Ajtai et al., Atmos. Environ., 122, 313–320, 2015) data sets. In addition, a new approach of combining the PAS, DMPS (number size distribution) and AE (soot) data sets into one model is also planned. Comparing the results, advantages and limitations of the filter-based approach (in the present paper) and several optical-related source apportionment methods based on the on-line data sets will be one of the key points of this other MS. We decided to split the whole matter into two parts (with a natural division line between the collection of aerosol samples and on-line measurements) because we consider that their joint presentation 1) could be too lengthy and perhaps even too complex and likely could have a too broad scope, and therefore, 2) it would detract the attention from the new apportionment scheme proposed in the present MS, and 3) our goal is not to prepare a comparative MS on a selection of apportionment methods. We indicated now these additional arguments, motivations and aims very briefly in the text. Nevertheless, it seems plausible and advantageous to describe the experiments of the aerosol sample collection and measurement campaign together in order to have a more comprehensive overview on the aerosol campaign and resulting analytical data sets as a whole, and to assist the future MS to focus the attention specifically on modelling issues and data validation.

4) page 4, lines 33-34: No blank uncertainty is given for mass on Nuclepore and quartz filters

The blank uncertainties are now added to the means. In an earlier study, the role and typical field blank uncertainties for Nuclepore, Teflon and quartz filters in the SFU sampler, which was also utilised in the present work, were determined from a larger number of sample sets, and they were discussed extensively (Salma et al., Atmos. Environ., 38, 27–36, 2004). It was concluded, for instance, that the uncertainty of the Nuclepore samples allows one to determine the PM mass more reliably than is the case with quartz fibre filters, and that the latter substrate is subject to several sampling and handling uncertainties. We refer now to this paper as well.

5) page 6, line 1-2: It seems unusual to combine online PM2.5 mass with offline filters (PM10-2.5) to construct PM10, especially when PM2.5 mass from filters is available even from the same filter pack. Moreover, fine fraction from Ghent SFU is rather PM2 than PM2.5 (Hopke et. al 1997) and therefore coarse fraction will be probably PM10-2 instead of PM10-2.5.

We decide to combine the $PM_{10\text{-}2.5}$ mass obtained from the Nuclepore filters with the corresponding mean on-line FDMS-TEOM $PM_{2.5}$ mass because the uncertainty for the $PM_{2.5}$ mass determined from the 12-h exposed fine quartz filters was considerably larger than that for

the mean on-line data. See also the answer to comment 4, and the paper referred there. The Gent-type stacked filter unit (SFU) sampler was indeed designed and realised by one of the co-authors of the present paper (W. Maenhaut) for collecting PM size fractions with nominal aerodynamic diameter (AD) of 10–2 and <2 μm. The cut-point of 2 μm results from drawing air through the top (coarse) Nuclepore filter with 8 μm pore size at a face velocity of 16.6 L/min. The separation between the two size fractions is, however, not very sharp (which is in contrast with the steep impaction collection curve of inertial impactors used for collecting $PM_{2.5}$ or PM_{10}). Furthermore, according to P.K. Hopke, the cut-point between the two size fractions in the Gent SFU is rather at 2.2 μm than at 2 μm (P.K. Hopke, personal communication to W. Maenhaut). Thus, the cut-point of 2 μm is rather approximate. In many published papers, in which the Gent SFU was used, including many papers co-authored hy P.K. Hopke and some co-authored by W. Maenhaut (e.g., the paper by Putaud et al. (2010) to which reference is made in our manuscript), the cut-off value between the two size fraction was reported to be 2.5 μm. In any case, several chemical species have a saddle point in their mass size distribution around the 2–2.5 μm diameter region, so that the difference between their masses in PM_2 and $PM_{2.5}$ is virtually negligible.

6) page 6, line 7 and elsewhere: The term “data lines” should be probably “data points”

The data files recorded by the on-line instruments usually consist of many rows/lines of data. A row contains usually a date/time stamp in the first columns, and various other measured quantities (e.g. optical absorption coefficients at several wavelength, or mass concentration and reference mass concentration) in the following columns from which the final data of interest are calculated. By writing the expression data rows, we referred to this structure of the collected data file.

7) page 12, Fig 3.: The scheme is quite unclear, needs improvement.

The apportionment scheme is now described more explicitly. We clarify all input data that are necessary for the apportionment and their actual entrance point into the scheme more systematically in both the text and Fig. 3. The arrows in Fig. 3 were originally selected in such way that they indicate the pathways and their major steps in the data treatment flow starting from the measured TC concentration toward the assessed end quantities of the five major carbonaceous species, i.e. EC_{FF} and OC_{FF}, EC_{BB} and OC_{BB}, and OC_{BIO}, which is considered

helpful in overviewing the idea of the proposed new approach for readers who are not very familiar with dedicated with source apportionment.

8) page 14, line 5: The term “tendentious” seems not properly used in the text several times (three times on this page), please check.

The term “tendentious” is removed from the text and it is replaced by other wording.

Imre Salma
for all co-authors