

Thank you for taking the time reading through the article. Your constructive critique has been valuable in the revision process.

a) Positive artefact correction. The authors state: “The F14C of the positive artefacts were assumed to be equal to the F14C of the OC fractions.” This procedure is not acceptable. Table 3 indicates that the average VOC contribution on the filters were 0.7 $\mu\text{g}/\text{m}^3$, whereas Table 5 presents median particulate OC concentration as 1.1-2.3 $\mu\text{g}/\text{m}^3$. Consequently, the positive artefact constituted 23-39% of the OC collected on the filter (i.e., OCp + VOC). This is such a large correction that a deviation of the assumption of identical F14C in OCp and VOC has substantial input on the results in Fig. 4 and Table 5. To me, this has direct impact on the discussion in chapter 3.4.2: If the hypothesis on page 13590, lines 18-20 (“This could be explained by the lower temperature in the winter altering the gas-particle equilibrium and thus suggests that a larger portion of the fossil OC during winter is secondary aerosol.”) is true, we should expect rather fossil VOC in summer which contradicts the assumption of identical F14C in OCp and VOC. With this respect, the statement on page 13590, lines 24-25 (“The lower F14C found in the winter cannot be interpreted as anything else but increased influence by fossil fuels.”) is not valid. I expect that this observation is not real but artificial from an inadequate positive artifact correction. In summary, 14C measurements of at least a few back filters are essential for this study.

That is true however it must be said that the values in table 3 only apply to the undenudered filters which are 8 in this study. These samples coincide with the EMEP campaigns in October 2008 and March 2009 which affect the autumn and spring values. These samples do not affect the general results of the paper since they focus on the summer and winter seasons.

We analysed 4 samples (2 times 2 pooled from each season and 2 times 1 from each season) and corrected the F¹⁴C for those samples accordingly. However, unfortunately the run did not go well and the result was not useable. The limited amount of material found on the back filters make a second attempt impossible. So we have to look elsewhere. In another study (Yttri et al., 2011a) we ran the back filters for the campaign. The F¹⁴C values found on the pooled back filters (4 times 4 filters) was not statistically different from the mean values of the front filters. Looking at the estimated OC part however, the back filters had a lower ¹⁴C value (5-20%). The campaign from Yttri et al. 2011 was set in the summer and the results may therefore not be transferable but that is the closest we can get since we denuder all our other samples

Actions taken: In the last sentence in sect 3.1 OC is changed to TC which is what has been used in the calculations.

b) Statistical significance of the data. The complex calculation of the sources according to chapter 2.5 bases in part on subtraction (e.g., for the determination of OCff, ECff and OCbio). This may lead to zero or even negative values as indicated in the discussion (page 13583, line 22) and by Figure 6. It should be emphasized for each individual sample (Figure 6) and for seasonal averages (Table 5 and Figure 4) if a source is to be considered as statistically insignificant. For example, this applies to me for fossil EC during winter so that this source should not appear in Figure 4 for this particular season and a detection limit should be given in Table 5.

The spread in data points from the simulations of fossil EC (Fig 6) is large in winter and for certain individual samples the median value is zero. However, as a mean (of the best estimates i.e. median values) over the whole season the values are greater than 0.

Actions taken: in figure 6 the point not regarded as significant is indicated by an unfilled marker. To be significant the 25th percentile of the simulations exceed 10% of the LOD of the superior fraction (OC or EC). In figure 4 and table 5 the best estimate is still used. The LOD for the OC/EC is easily calculated, but to what fraction a source must contribute is more arbitrary and in this case 10 % is chosen.

c) Equation (3). This formula provides large uncertainties in the final data as it employs two emission ratios in combination. This disadvantageous fact is discussed in chapter 3.4.4. Instead of $(EC/OC)/(lev/EC)$, I propose to use $(EC/lev)_{bb}$ ratios directly which should be achievable from the original papers. Hopefully, this procedure will confine the source apportionment outcome.

A similar approach to what is used in Yttri et al. (2011b), where TC_{bb} is first calculated by the levoglucosan values and thereafter separated into OC_{bb} and EC_{bb} , has been considered. I also recalculated the SA based of your suggestion and the three methods all give as large uncertainties.

d) OC_{bio} . This term is used in part incorrectly in the manuscript. On page 13584, line 1, it is defined as “non-fossil OC”, which is not correct, as the latter also includes biomass-burning OC. I found a repetition of this error on page 13591, line 3. The authors should screen for further misuses.

True. OC_{bio} is mostly of biogenic origin (thus the choice of name) however it may be influenced by other sources as well.

Actions taken: The error has been corrected and no further misuses have been found.

e) $(EC/OC)_{bb}$ ratios. The values from Yttri et al., *Atmos. Chem. Phys. Discuss.*, 11, 7375-7422, 2011 should be considered, as those were adapted to the situation in Scandinavia. That evaluation concludes quite narrow ranges for $(OC/TC)_{bb}$ so that the ranges of $(EC/OC)_{bb}$ in Table 2 seem to be unrealistically high.

Yttri et al. (2011b) uses a $(EC/OC)_{bb}$ ratio of 0.22-0.37 which is in the higher end of the ratio we have used since the EC/OC values is set in a beta distribution which favours the lower values. Compared to the Yttri values the values used are lower. The ratios used in this study are a more narrow range than in the carbosol project (Gelencsér et al., 2007) yet larger than found by Yttri. We think it is motivated to use a wide range since the measured values differ so much it should be taken into account during the calculation even though the spread becomes large.

f) Levoglucosan results. Does ten-times more levoglucosan in winter compared to summer necessarily mean ten-times more OC_{bb} (page 13589, lines 17-18)? $(lev/OC)_{bb}$ emission ratios are not identical for summer and winter, as the former has a larger influence from wild fires and long-range transport, whereas the latter is rather controlled by domestic heating.

Very good question. Long range transport of open burning aerosol would, with the degradation of levoglucosan suggested by Hoffmann et al 2010, lead to significant loss of levoglucosan and thus underestimate the burning influence (if POC from biomass burning is considered stable). The ratios from Zdráhal in table 1, which are collected during dry period in the amazon does however not show a big difference in levoglucosan/ OC_{bb} ratio. It has however lower EC/OC ratio which might be due to the condition of the fire or the formation of SOA from VOC_{bb} . But what is most important is that the ratios used intend to determine the primary emission from the biomass burning and does not take into account the SOA formation from biomass burning VOCs.

As mentioned the atmospheric stability of levoglucosan is uncertain. But if a different set of ratios would be used it would have to include both the stability of levoglucosan as well as POC_{bb} , both are which are uncertain and thus not feasible. No good datasets have been found to use seasonal ratios.

g) Citation Putaud et al., 2010. A better reference should be given than a conference presentation.

First, the reference does not point at the EUSAAR project, there are better references for that. It aims at the evaluation of positive artifacts at European sites. This reference is so far the only place these data points have been used. The reference have been remove since the data being used is our own, but the measurements were conducted within EUSAAR in Putaud's WP and it should therefore be credited. A manuscript is being prepared and it will be included if ready in time.

Action taken: citation removed.

h) Table 1. According to page 13585, lines 18-20, median values should be shown instead of mean values.

The median values of the 3000 simulations are considered the best estimate, not the median values of the ratios.

Action taken: Clarified in the text (Sect 2.6).

“ All variables were allowed to vary within a set distribution, and calculations were performed with 3000 random sets of variables. The median value from the calculations was considered the best estimate... ”

Also, the variables used in the calculations are found in table 2. Table 1 are results from other studies. A reference to table 2 has been added to sect. 2.6 to minimize the risk of misunderstandings.

i) Figures 2+3+6. Seasons should be indicated by vertical lines.

changed accordingly

j) Figure 3. What does “The error bars illustrate one standard deviation” mean? A deviation implies measurement repetitions. Is this really the case or do the bars represent measurement uncertainties?

The standard deviation of the levoglucosan samples are calculated by multiple extractions of the filters (2), derivatisation (2) and injections (2). In total every sample is injected 8 times.

Action taken: *“The samples were extracted, derivatised and injected using duplicates, i.e. every sample was injected 8 times.”* added to sect. 2.4.

The standard deviation of the F¹⁴C is calculated using counting statistics in the AMS for several measurement runs for the samples as well as the blanks and primary standards. No replicates were conducted for the samples; however secondary standards are used to ensure that the values obtained are reliable. The secondary standard can be seen as “between sample” variation.

Action taken: The sentence *“The uncertainty of the measured value is determined by combining counting statistics of the AMS run of the sample, blanks and standards.”* has been added to sect. 2.3.

References

Gelencsér, A., May, B., Simpson, D., Sánchez-Ochoa, A., Kasper-Giebl, A., Puxbaum, H., Caseiro, A., Pio, C., and Legrand, M.: Source apportionment of PM_{2.5} organic aerosol over Europe: Primary/secondary, natural/anthropogenic, and fossil/biogenic origin, *Journal of Geophysical Research-Atmospheres*, 112, 1-12, 10.1029/2006JD008094, 2007.

Yttri, K. E., Simpson, D., Nøjgaard, J. K., Kristensen, K., Genberg, J., Stenström, K., Swietlicki, E., Hillamo, R., Aurela, M., Bauer, H., Offenberg, J. H., Jaoui, M., Dye, C., Eckhardt, S., Burkhardt, J. F., Stohl, A., and Glasius, M.: Source apportionment of the summer time carbonaceous aerosol at Nordic rural background sites, *Atmos. Chem. Phys. Discuss.*, 11, 16369-16416, 10.5194/acpd-11-16369-2011, 2011a.

Yttri, K. E., Simpson, D., Stenström, K., Puxbaum, H., and Svendby, T.: Source apportionment of the carbonaceous aerosol in Norway – quantitative estimates based on ^{14}C , thermal-optical and organic tracer analysis, *Atmospheric Chemistry and Physics*, 11, 9375-9394, 10.5194/acp-11-9375-2011, 2011b.