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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of carbonaceous aerosol in southern Sweden" *by* J. Genberg et al.

Anonymous Referee #1

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The publication "Source apportionment of carbonaceous aerosol in southern Sweden" by Genberg et al. describes the characterization of carbonaceous particulate matter from a rural site in Sweden based on analysis of radiocarbon, levoglucosan, organic carbon (OC) and elemental carbon (EC) concentrations. 35 weekly samples from April 2008 to April 2009 were attributed to the these sources: biogenic OC, biomass burning OC, fossil fuel OC, biomass burning EC and fossil fuel EC. Although the number of similar studies have increased recently, the available source information is still scare, especially considering the progress throughout a whole year. Furthermore, the authors discuss the seasonal contributions of these sources and compare their results with the EMEP chemical transport model, which leads to a critical review of both. Therefore, this publication is suitable for publication in ACP after revision based on the following comments. Unfortunately, this also includes additional measurements.

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 μg/m3, whereas Table 5 presents median particulate OC concentration as 1.1-2.3 μg/m3. 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 artefact correction. In summary, 14C measurements of at least a few back filters are essential for this study.

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.

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.

d) OCbio. 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.

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.

f) Levoglucosan results. Does ten-times more levoglucosan in winter compared to summer necessarily mean ten-times more OCbb (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.

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

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

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

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?

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13575, 2011.