Authors' response to reviewers' comments

We would like to thank the reviewer for his/her careful reading and comments which helped to clarify the message of the manuscript. Our responses to each of the reviewer's comments are written in blue. Changes in the manuscript are marked in yellow.

Interactive comment on "Carbonaceous aerosol source apportionment using the aethalometer model – Evaluation by radiocarbon and levoglucosan analysis at a rural background site in southern Sweden" by Johan Martinsson et al.

Anonymous Referee #2
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General comments

The authors present an evaluation of the aethalometer model for carbonaceous particle source apportionment using radiocarbon and levoglucosan measurements, and quantify wood burning (WB) and fossil fuel combustion (FF) aerosol for a year-long dataset from a rural station in southern Sweden. The model is modified to allow for apportioned non-light absorbing biogenic aerosol to vary in time, improving the aethalometer-based source apportionment compared to radiocarbon and levoglucosan data. This is an interesting and solid study. The manuscript is very well written (with a few grammatical errors here and there, I suggest having it checked by a native English speaker), and the analysis is sound. I therefore recommend publication in ACP after the following comments have been addressed:

Specific comments

P. 4, l. 13: How efficient are the active carbon denuders? Please add information on that and the expected background. Is there any information on the evaporation of semi-volatiles from the particles after disturbance of the gas-particle equilibrium due to the denuders?

Genberg et al. (2011) conducted a full year source apportionment study at Vavihill 2008-2009. They conducted tests on the installed denuders and found 90-95% denuder efficiency (Genberg et al. 2011).

Further, when denuders were installed, Genberg et al. (2011) observed that obtained field blanks contained a carbon content similar to that of the sampled back filters, indicating that the possible negative artefact (due to disturbance of the gas-particle equilibrium) was low. Hence, Genberg et al. (2011) did not consider nor corrected for this artefact. In our study, we did not obtain any field blanks, however we have adopted the same approach as Genberg et al. (2011) since we are performing a similar study at the same measurement site with the same sampling setup, i.e. we have not corrected for this artefact.

We have added information on the efficiency of the denuders and the presumably low negative artefact caused by the denuders in section 2.1.

P. 6, l. 1 – 4: Why were the AAE values not calculated based on the actual data, or at least calculated and compared to literature data? Assuming an AAEFF of 1, and plotting/fitting babs vs wavelength (either averaged, or time-dependent, more appropriate here) can be used to derive AAEWB.

We do not see how we can calculate an AAE_{WB} using the method suggested by the reviewer. To be able to derive AAE_{WB} we need to know AAE values from the wood burning, i.e. through emission inventories. Our actual measured data shows the *babs* from a mixture of different aerosol sources. Hence, we cannot see how we could select any AAE_{WB} based on these source mixtures of *babs*.

We instead obtained our source specific AAE values from emission inventories as displayed in Table 1. The mean AAE_{WB} are in line with selected AAE_{WB} values in previous aethalometer model source apportionment studies by Sandradewi et al. (2008) and Massabo et al. (2015). Further, our selected AAE_{WB} value (1.81) is rather close to the recently suggested AAE_{WB} (1.68) by Zotter et al. (2016).

P. 6, l. 11-12: If site specific sigma_abs were calculated based on linear regression of babs against EC, doesn't that imply an overestimation of sigma_abs, as light-absorbing OC is not included in EC? Please clarify.

This is true. We have added two sentences in order to clarify this.

P. 6, l.13: CM could also be SOA from WB and FF; should be mentioned here.

We have added this information.

P. 12, l. 2-3: This statement should be supported by references and more explanations.

We have developed and clarified this statement with some explanations.

P. 12, l. 29: Is the year-long time series correlated, or the diel evolution? I am assuming you are talking about the correlation of the time series. Apart from the non-optimal apportionment, a reason could also be, similar to the correlation of NOx and CMwb, a similar trend, but a different cause. CMFF is higher in winter than in summer, and so is NOx – potentially traffic emissions become more important in winter as well, or meteorological conditions favor the build-up of pollution episodes?

The correlation refers to the year-long time series since the levoglucosan data is in low time resolution (i.e. 72 h). This has been clarified.

This relation is hard to explain. It seems like the traffic emissions (judging from CM_{FF} and NO_X data) are relatively more important during winter compared to summer, at least in relation to TC. In absolute concentration, CM_{FF} shows highest values during the fall and similar values during the other seasons (Table 4).

However, studying the relation on a seasonal basis we can see that a large portion of the correlation is explained by high correlations during winter and spring (R^2=0.77; p<0.001 and R^2=0.62; p<0.001, respectively) as compared to summer and fall (R^2=0.04; p=0.3 and R^2=0.35; p=0.001, respectively). Hence, one can speculate that the stronger correlation during winter and spring are associated to increased wood burning where some of the generated aerosols are absorbing light with a spectral dependence, AAE, close to 1, thus being falsely apportioned as fossil fuel combustion aerosol. It is also possible that lignite combustion aerosols from continental Europe, containing levoglucosan and exhibiting a spectral dependence of an AAE close to 1, may show higher abundance during these seasons, however we have no data supporting this speculation.

P. 13, l. 15: CMbio was assumed to be 0 for calculations of C1 and C2. Could this be a reason for the overestimation of CMFF?

We have added a new discussion paragraph were we performed a sensitivity analysis of the impact of non-light absorbing carbon on the aethalometer model results. It is very likely that the overestimation in CMFF can be explained by presence of non-light absorbing carbon (possibly biogenic carbon).

Technical comments
P. 7, l. 14: typo in chloride (also in subsequent use of trimethylsilyl chloride) P. 7, l.16: typo, should be dichloromethane.
This has been corrected.

References

Genberg, J., Hyder, M., Stenström, K., Bergström, R., Simpson, D., Fors, E. O., Jönsson, J. A., and Swietlicki, E.: Source apportionment of carbonaceous aerosol in southern Sweden, Atmos Chem Phys, 11, 11387-11400, 10.5194/acp-11-11387-2011, 2011.

Massabo, D., Caponi, L., Bernardoni, V., Bove, M. C., Brotto, P., Calzolai, G., Cassola, F., Chiari, M., Fedi, M. E., Fermo, P., Giannoni, M., Lucarelli, F., Nava, S., Piazzalunga, A., Valli, G., Vecchi, R., and Prati, P.: Multi-wavelength optical determination of black and brown carbon in atmospheric aerosols, Atmos Environ, 108, 1-12, 10.1016/j.atmosenv.2015.02.058, 2015.

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