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 #1
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Martinsson et al. present analysis of a yearlong dataset of carbonaceous aerosols at a rural background site. Long-term source apportionment studies of carbonaceous aerosols are rare, and in addition the authors propose a new modification to the 'aethalometer model' that they suggest can apportion biogenic sources in addition to wood burning and fossil fuel from aethalometer data. However, while the source apportionment results are promising for wood burning and biogenic carbon, there is a clear shortcoming in its ability to apportion carbon from fossil fuel. The authors are upfront with this issue and have tried alternative methods to improve the source apportionment model, but these did not offer any improvements. I would have liked to see more discussion on why the proposed aethalometer model over-estimated fossil fuel carbon, apart from the blaming the poor correlation between fossil fuel aerosol light absorption and carbon mass concentration. Why do the authors think there was such a poor fit?

We have performed a sensitivity analysis where we increased or decreased the carbon in Eq. 10-11, while not changing the b_{abs} parameters. This would be analogous to change the mass of non-light absorbing carbon. We have written a detailed discussion regarding this in section 3.4. Our results from this analysis suggest that interference of non-light absorbing carbon (presumably biogenic carbon) may be responsible for the observed overestimation of CM_{FF}. Hence, we have re-written our abstract, discussion and conclusion where we have added this information. We have omitted that the poor fit would be the reason for the overestimation.

Overall, the paper is well written and structured with a wide range of relevant references and I would recommend it for publication after consideration to the comments below.

Comments:

1. Page 11, line 15-20: While I would agree that the in Fig 3d, the BCFF diurnal trend is bimodal like traffic emissions (though with a peak at night?) I am not sure I agree that the diurnal trend in BCFF for winter especially (Fig 3c), is similar to NOx (Fig 3g). The diurnal trends in NOx in winter are more what would be expected for traffic emissions, and are dissimilar to that observed for BCFF. The flat diurnal trend in BCFF for winter instead to me suggests that the model did not apportion the FF fraction correctly.

We agree with the reviewer that the BC_{FF} during winter (Fig. 3c) appears to have less pronounced (if any) bimodal behavior in comparison to NO_X concentrations during winter (Fig. 3g). However, we would still claim that there are features in common. Both BC_{FF} and NO_X start with decreasing concentrations from 1:00-6:00, and then increase and stay on an elevated level until 22:00 when both concentrations show indications of decrease.

It is possible that the elevated precipitation during the winter (described in section 3.1) was responsible for increased wet deposition of BC while leaving the atmospheric NO_X unaffected, resulting in blurring correlations between the two parameters during the winter.

We have added a few words to indicate that the bimodal pattern is stronger for the NO_X concentrations compared to the BC_{FF} concentrations.

We have checked our data again and found no reason why the calculations here should be invalid.

2. Page 11, line 25: If NOx is being oxidized in transport to the site, then shouldn't the CMFF and NOx show the highest correlation during winter when there is less photochemistry compared to the other seasons? Why is there such a better correlation in spring compared to the other seasons?

This is a very good question and hard to explain. As we answered on the former comment, one can speculate that increased wet deposition during winter resulted in a scavenging effect on BC while leaving the atmospheric NO_X unaffected. Consequently, the correlations between BC and NO_X during winter may have been weakened.

Studying the precipitation of the other seasons we find that the spring had the lowest precipitation, hence possibly explaining the improved correlation between BC_{FF} and NO_X during this period (R^2=0.41; p<0.001). However, the precipitation during spring is not significantly different from the precipitation during fall and summer (seasons with much lower R^2 value between BC_{FF} and NO_X, R^2=0.07; p=0.021 and R^2=0.09; p=0.009, respectively). Hence, precipitation may only partially explain the increased correlation during spring.

3. Page 12, lines 1-3: I find it surprising that the CMwb was better correlated with NOx than CMFF. You explain this by stating that both NOx and CMwb have a seasonal dependence, but why would NOx and CMwb have the same seasonal dependence? Have you examined the correlation between CMwb and NOx to see if there is a change for the seasons?

NO_X is mainly emitted from traffic, which is a rather stable emitter throughout the year. Furthermore, NO_X has a longer lifetime during the cold/dark period of the year due to a lower rate of atmospheric photo-oxidation. Hence, NO_X concentrations can be expected to be elevated (due to longer lifetime) during winter, and lower during summer. The same pattern applies for CM_{WB} but for a different reason. Residents heat their homes through WB during winter, an activity that is almost absent during summer. We have clarified this in the text.

There are no changes in the correlation between CM_{WB} and NO_X, all seasons show significant correlations.

4. Page 12, line 17. In Fig S6, why there is a very large spike in the concentration of levoglucosan in late March that is not observed in AAE (fig 2)? This sharp spike in levoglucosan suggests a biomass burning event, and I would have thought you would observe a corresponding increase in AAE if wood burning aerosols have a high AAE? Investigating the cause of the spike may help guide the choice of AAE for wood burning at the site.

The levoglucosan peak is derived from a 72h quartz filter with a stop-date of 2015-03-19. Hence, the filter represents ambient air during 16-18th of March 2015. This is indeed a pollution episode. The OC and EC concentrations are also elevated as displayed in Fig. 1a. Air mass trajectory analysis revealed that southeasterly (SE) air masses totally dominated (92 %) during this three day period. As pointed out in section 3.5, air masses from SE are associated to higher levels of aerosol loading. Higher aerosol concentrations from the SE are further supported by the study by Kristensson et al. (2008).

During this three day period we actually had somewhat elevated AAE (although it is hard to see in Fig. 2.). The mean AAE during this three day period was 1.44 (±0.03 standard deviation), which is

higher compared to the average AAE for the whole month of March 2015 (mean=1.37±0.09). However, since the measured $F^{14}C$ from the same filter showed a value of 0.86 there are some obvious contribution from fossil fuel combustion. Hence, we would not regard this pollution episode as being totally dominated by wood burning.

Nevertheless, there is a discrepancy in apportioned wood burning between the aethalometer model and the radiocarbon + levoglucosan method during this three day period. The aethalometer model apportions 54 % of the TC to wood burning while the radiocarbon + levoglucosan method apportions 90 % of the TC into wood burning. However, entangling the causes for this discrepancy is difficult. For instance, combustion of lignite has been shown to emit large quantities of levoglucosan, although it is a fossil source (Fabbri et al., 2008). Further, 55 % of the heat and power generation in Poland (located in the SE direction of Vavihill) are generated from lignite combustion (Burmistrz et al., 2016). Hence, deriving any source specific AAEs from this pollution episode should be conducted with great caution.

5. Section 3.5: did you use annual means for your comparison of the aethalometer and radiocarbon and levoglucosan apportionment? Did you see any changes in agreement between the two methods for the seasons, e.g. was there better agreement in summer or winter?

In section 3.4 we are comparing mainly annual means between the two source apportionment methods. As displayed in Fig. 6, we are comparing the methods on a seasonal basis. We re-analyzed possible differences on a seasonal basis through analysis of variance (ANOVA). There was a significant difference between the fossil fuel apportionments between the two methods for all seasons. There were no significant differences in apportionment of wood burning and biogenic carbonaceous aerosol between the two methods in any of the seasons.

We have clarified in section 3.4 that these comparisons discuss annual means.

6. Page13, line 15: In the aethalometer model, to calculate C1 and C2 only winter data was used as it was assumed that there would be negligible CMbio. However, the results from the radiocarbon and levoglucosan model suggest that biogenic carbon was not negligible during winter. I think that you should therefore include some discussion on how the presence of biogenic carbon in winter affected the source apportionment by the aethaolometer model.

As described in our answer to the first question, we have performed a sensitivity analysis where we increased or decreased the carbon in Eq. 10-11, while not changing the b_{abs} parameters. This would be analogous to change the mass of non-light absorbing carbon. We have written a detailed discussion regarding this in section 3.4

7. Page 13, line 31: Why did you fix CMbio to -0.103 ug m-3 and not zero as you expect no CMBIO in winter?

In this case we are comparing our method (i.e. letting CM_{Bio} vary outside the linear regressions) to the model proposed by Sandradewi et al. (2008) where they suggested solving a bilinear regression model with an allowed intercept. Hence, in order to adopt the Sandradewi method, we need to fix our intercept.

8. Section 3.6: My understanding is that in the proposed aethalometer model, the influence of biogenic carbon needs to be minimized in order to calculate C1 and C2. In addition to only using winter data, could you not also select data for the calculation by wind direction? My reading of this section is that there are geographically distinct areas around the sampling

site, and that filtering by wind direction you could further decrease in the biogenic influence (e.g. removing data when the wind is from a forested area or from the NE?) in the data.

This is a good idea suggested by the reviewer. In our study we have the lowest amount of incoming NE air mass during winter (i.e. 10 % of the air masses were from this direction). Hence, we believe that the selected winter data in our study exhibit favorable conditions in order to minimize biogenic carbonaceous aerosol.

We have added a sentence in section 3.5 describing the low abundance of NE air masses during winter.

9. Page 14, line 19-20: Why would SW air masses have high NOx but not be associated with carbonaceous aerosols, when traffic emissions are a significant source of both?

This is indeed very hard to explain. One explanation might be that the SW air masses are associated to increased precipitation (R^2=0.19; p<0.01). This correlation was particularly high during winter (R^2=0.41; p<0.01), a time when NO_X can be expected to have increased lifetime due to low photochemical rates. Hence, it is possible that the increased SW-precipitation increased the wet deposition of carbonaceous aerosol particles while leaving the NO_X unaffected.

We have added a sentence to offer this explanation in section 3.5.

References

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