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Reply on RC2

Sanna Saarikoski et al.

Author comment on "Sources of black carbon at residential and traffic environments obtained by two source apportionment methods" by Sanna Saarikoski et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-231-AC2, 2021

Author response to Referee #2

We thank Referee #2 for her/his valuable comments. We have responded to all the comments below and the changes to the manuscript have been made in "track changes" mode.

RC2: 'Comment on acp-2021-231', Anonymous Referee #2, 08 Jul 2021

1) the introduction could be shortened**,** especially regarding the exhaustive listing of SA methods for carbonaceous aerosols. I don't think it is needed here.

Reply: introduction has been shortened, especially in terms of source apportionment methods not utilized in this study.

2) it should be clearer in the text (eg abstract) that the measurement campaigns were not carried out at the same period of the year. For instance, biomass burning is the main BC sources at residential site in winter only.

Reply: the measurement season has been added to abstract and the difference between the measurement years has been emphasized in abstract and conclusions. See also comment 2) to Referee #1.

3) Some clarifications would need to be added about SP-AMS measurements. Why a different time resolution was applied for the 2 sites ? the CE of 1 applied on SP-AMS data is not well justified. Were they any co-located PM1 measurements which could validate SP-AMS concentrations ? Wouldn't the "imperfect laser-to-particle beam alignment" be included during rBC RIE calibration ? Is it a regular feature of SP-AMS to underestimate AE-derived BC compared to aethalometer measurements ?

Reply: There was collocated PM_1 measurement (Grimm) at the residential site but not at the street canyon. The comparison of the sum of the SP-AMS species (excluding rBC) and AE33-BC with PM_1 from Grimm at the residential site has been added to supplemental material and a paragraph of text has been added. See also comment 1) to Referee #1.

It is not a regular feature that the SP-AMS underestimates AE-derived BC. Imperfect laser alignment is one of the reasons that can cause underestimation. Imperfect laser-to-particle beam alignment is partly included in rBC RIE, however, if the laser beam is close to the edge of particle beam (laser beam is narrower than particle beam), small changes in laser alignment can change RIE. Another factor that affects the calibration of rBC is that rBC was calibrated with Regal Black particles with the mobility size of 300 nm, that can be aligned much better than smaller rBC particles measured e.g. at the traffic environment. Additionally, the composition of Regal black and ambient BC can differ, however, Regal black has been selected for the calibrations because its rBC mass spectra resembles ambient BC. As already mentioned in Referee #1 comment 2) also some of the BC particles in ambient air can be missed by the SP-AMS also due to the size limitations of the aerodynamic lens.

4) The strategy for PMF should be more explicit. For instance, the authors never mention the investigation of residuals, which is nevertheless a critical issue to adress. Was the uncertainty matrix calculated using the regular algorithm for AMS ?

Reply: New figures on residuals (Fig. S20 and S21) have been added to supplemental material as suggested by the Referee. Also a new paragraph of text regarding the residual analysis have been added.

Uncertainties were calculated using the regular algorithm for the AMS.

5) The authors present average values at different time averages (sometimes 1h, sometimes 10min). I suggest to choose one, and keep it throughout the paper.

Reply: The reason for different averaging times for residential and street canyon sites was the large difference between the length of the measurement periods. Measurements at the residential site lasted unbroken approximately three months whereas the measurements at the street canyon were not continuous and they consisted of eight separate time periods with a total of 195 hours of data (approximately 8 days). Additionally, the sites were very different in terms of particle dynamics; at the street canyon site, the contribution of various BC sources varied more rapidly than at the residential site.

One hour time resolution was used mostly for the α calculations (except histograms). The reason for longer averaging period was that the uncertainty in α values was considerable large the shorter averaging periods. 1-hour period was used also in temperature data and site-average BC concentrations from the AE33 to present general situation at the sites.

6) Long-range transport episodes and characteristics are not sufficiently supported. The author may want to investigate this issue with e.g. trajectory and/or wind analysis.

Reply: The characteristics of the long-range episodes have been analyzed in the paper of Teinilä et al. that is under review in Atmospheric Environment, and therefore, we decided to exclude the detailed analysis from this article. However, as suggested by the Referee, trajectories during the LRT episodes were added to the supplemental material as well as a sentence: "According to the calculated air mass back trajectories (Fig. S30), LRT aerosol arrived in Helsinki from agricultural burning or wildfires in Eastern Europe."

7) About Angstrom Exponent: why didn't the authors calculate the AAE from all wavelengths (should better take spectral dependance into account) ? The set of α values could have been calculated from diurnal variation of probability distribution function. Have the authors investigated this ? This work also highlights the limitation of the aethalometer model to characterize LRT of BC and coating, which may be associated to different α values. An interesting outcome would have been here to retrieve the AAE for each SP-AMS-PMF factors by either i) multi-linear regression, or ii) injecting all BC(1-7) within PMF. Have the authors ever considered this ?

Reply: α was calculated also by using all wavelengths but the results and conclusions did not differ much from α calculated only by using two wavelengths. α calculated with two wavelengths showed ~5% larger values, however there was a slight negative offset (see Figure R1 in supplement file to this author comment).

Diurnal variation of α probability distribution functions at both sites has been added to supplemental material (Fig. S33). It shows that α was smaller in daytime at both sites, the difference between day and night being larger for the street canyon data than for the residential area. At the residential area, α was smaller particularly during the morning rush hour. We added to text: "In terms of diurnal variation of the α values, α was smaller in daytime at both sites, the difference between day and night being larger for the street canyon than for the residential area (Fig S33)."

Regarding α values for each SP-AMS-PMF factor, we have investigated separate α values tentatively at the residential site by using multi-linear regression, however, it appears to be challenging as the α values seem to be close to each other, and additionally, the time series of the factors do not differ that much from each other. In attached file are scatter plot and average diurnal trends for the α values measured with the AE33 and reconstructed from the AMS PMF data. It shows that slightly different values are obtained depending on if we want to optimize linear fit or diurnal variation, however, it seems that the α values are approximately 1.05–1.1 for BC_{HOA}, 1.45–1.5 for BC_{BBOA}, 1.40–1.65 for $BC_{LV\text{-}OOA\text{-}LRT}$, 1.2–1.3 for $BC_{LV\text{-}OOA}$ and 1.25–1.42 for $BC_{SV\text{-}OOA}$. As this data is very preliminary, and has lot of uncertainties, we did not include it in the manuscript. However, because this topic is very interesting, we will continue this work and our tentative plan is to publish it in another paper.

Additional corrections

the citation details of Helin et al. (2021) has been changed

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-231/acp-2021-231-AC2-supplement.pdf