Comment on acp-2021-231
Anonymous Referee #1

Referee comment on "Sources of black carbon at residential and traffic environments" by Sanna Saarikoski et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-231-RC1, 2021

Reviewer #1

General comments

This work provides interesting insights into urban emissions and their contribution to the BC loading in Helsinki, at two sites influenced by contrasting combustion sources. They apportioned BC sources by using and comparing two methods: (1) the model of Sandradewi et al. (2008) based on multiwavelength aethalometer measurements and (2) the PMF applied to organic and refractory black carbon components measured by a SP-AMS. The methodology regarding the SP-AMS and coating estimation would benefit from some elaboration.

The combined approach provides a better understanding of BC sources, as evidenced from how the BC observed on the residential site is mainly linked to local biomass burning, while the ones observed at the street canyon are mainly related to traffic emissions. At both sites, it has been shown that, in addition to local traffic, the BC associated with LV-OOA and related to more regional sources also contributes to the BC_{ff} estimated from AE33 measurements.

This study underlines the capacity of SP-AMS-PMF to identified specific BC sources, be they regional or local, in comparison to the AE33 model and especially in multi-influenced urban environments.

Specific comments

- Line 168: Why apply a CE of 1 on both datasets? Could more details about NR-PM_{1} and rBC quantification be provided (e.g. RIE might affect the contribution of rBC reported in Table S1 and S2)? Was the SP-AMS run purely in dual mode?
- Could comparisons between rBC from SP-AMS and eBC from AE33 and MAAP be added to the supplement? Could discrepancies between rBC and eBC and, later on, BC_{ff} / BC_{wb} and BC_{HOA} / BC_{BBOA}, be explained by either a low CE of rBC due to poor particle/laser
beam alignment and/or the fact that the smaller fraction of BC (< 70nm) is not detected by the SP-AMS?

- References regarding the coating factor calculation are missing as well as the assumptions behind this estimation and their potential impacts on your results should be discussed. Statistics about the SP-AMS and AE33 concentrations in the supplement could be useful alongside NR-PM$_1$ diurnal variations.

- Any reason for using only C$_2$+, C$_3$+ and C$_4$+?

- Lines 279-283: "At the residential site, the contribution of rBC was largest to the mass spectra of BBOA in three and four factor solution while in five factor solution the contribution of rBC was largest for LV-OOA. At the street canyon, the contribution of rBC was largest to HOA independent of the number of factors. The contribution of rBC to HOA was more than double at the street canyon compared to that at the residential site. In contrast, the mass spectra of BBOA had several times more rBC at the residential site than at the street canyon.” Why such contrast in rBC contributions from 4 to 5 factors at the residential site? Also, the f$_{C2H4O2}$ associated with the two BBOAs differs between the two sites, could it imply that one of the BBOA is more aged and harder to separate from other SOA?

- Line 469-470: "This suggests that BC$_{LV-OOA}$ was clearly associated with non-local sources.” Can BC size distribution alone lead to such a conclusion? Couldn’t SOA be formed by fast processing of locally emitted particles (e.g BC$_{HOA}$ at 100-150nm)?

- It could be interesting to report the average angstrom exponent when BBOA or HOA dominated periods at both sites.

- Line 511-513: "Biomass burning BC obtained from two source apportionment methods followed very similar time trend, however, BC$_{wb}$ calculated with the $\alpha_{ff}$ and $\alpha_{wb}$ values of 1 and 2, respectively, displayed smaller values than BCBBOA or BC$_{wb}$ calculated with the $\alpha_{ff}$ and $\alpha_{wb}$ values of 0.9 and 1.68, respectively.” How much smaller? Could any fit be done based on the two comparisons presented in figure 7.b? A parallel between the diurnal variations of AE33 (BC$_{ff}$ and BC$_{wb}$) and concentrations of the BC-PMF-factors – either by adding to figure 1.c and 5.b or having the equivalent of Figure S28 for BC$_{wb}$ and corresponding diurnal cycles for the residential area in the supplement – could provide a good support to the discussion in section 3.3.

- Line 517-520: “In general, that trend suggests that the aethalometer model produces more constant BB% than PMF and is likely be less sensitive to the changes in the BC sources compared to PMF. Similar observation was done when the diurnal patterns of BC$_{wb}$ and BC$_{BBOA}$ were compared; BC$_{wb}$ had a rather flat diurnal trend whereas BCBBOA varied more clearly during the course of the day.” Could it mean instead that the constant angstrom exponent used to estimate BC$_{wb}$ is not really adapted when there are different biomass burning sources (such as BC$_{BBOA}$ and BC$_{LV-OOA-LRT}$) with potentially different absorption properties influencing the site?

Technical corrections and minor comments

- If it is possible, having a more detailed title which is more reflective of the content and novelty of the paper might help.

- The fact that the measurements in Street Canyon and residential areas took place years apart should be at least mentioned in the abstract and conclusions.

- Even though the two sites are described in other papers, it would also be useful to include in the supplement the map of the two sites and potential surrounding sources, and the distance between the two sites as well as the Luukki site.

- Line 22-23: “In general, the aethalometer model showed less variation between the sources within a day than PMF being less responsive to the fast changes in the BC
sources at the site.” Is it that the aethalometer model shows less variations, or that it
cannot distinguish between as many sources as AMS-PMF due to similar optical
properties despite different OA sources?

- Line 225: “BBOA concentration was a slightly smaller in daytime”, was slightly smaller?
- Line 272: “HOA correlated strongly with NO and NOx r being larger for four and five
  factor solutions than for three factor solution.” Missing punctuation?
- Line 361-362: “That is a slightly larger contribution that the campaign-average
  percentage obtained in this study”, changed to “larger contribution than”.
- Line 382: Change (Fig. 23) to Fig. S23.
- Why are the elemental ratios missing from Figure S6 to S8?
- Line 87: “the light absorption in different wavelengths”, changed to “at different
  wavelengths”
- Line 610-614: Barreira, L. M. F et al. 2021., has been accepted
- Line 393: “LRT episodes observed in Helsinki in April 2019 will be discussed in more
  detail in the other paper.” Are there any references for this paper? Otherwise, you
might want to rephrase as “in another paper”.
- Line 394-397: “BCSV-OOA concentration was smaller from 9:00 to 21:00 than at the
  other times of the day. BCSV-OOA concentrations did not depend on the ambient
  temperature (Fig. S23).” Any idea of the sources or mechanism leading to the
  formation of SV-OOA?
- Line 401 and Figure 2: specifying Lukki background site, instead of background site
  might avoid confusing it with Street Canyon site.
- Line 406: “probably caused by the local BC emissions at the background site
  accumulated in the boundary layer due to the temperature inversion.” Any data /
  references?
- Line 484-486: “n ambient measurements, a can be larger than 485 that measured
directly from the emission source as a values for biomass burning emissions have been
shown to increase due to the atmospheric oxidation processes simulated with a smog
chamber (Tasoglou et al., 2017).” Studies have also reported a decrease of BB
angstrom exponent by photooxidation during atmospheric aging (Nicolaie et al., 2013,
Dasari et al., 2019...), which could also explain the difference in AAE if the BBOA are
not locally emitted.
- Line 518-520: “Biomass burning BC obtained from two source apportionment methods
followed very similar time trend, however, BCwb calculated with the αff and αwb values of
1 and 2, respectively, displayed smaller values than BCbbo, or BCwb calculated with the
αff and αwb values of 0.9 and 1.68, respectively.” As mentioned before, the comparison
between the diurnal profiles of BCwb and BCbbo could be presented in the supplement.
- Figure 2.b: Y axis legend should be corrected from “ug (m^{-3})” to (ug m^{-3}).
- Figure 3.b and Figure 5.c: As mentioned in the caption, the two size distributions
correspond to the BC size distribution under BBOA influence and background conditions
respectively. The legend should reflect the same information, as BCbbo and BC_{LV+SVOOA}
size distributions cannot be completely deconvolved. Also, what about adding the size
distribution under LRT conditions? Could the corresponding size distribution of organic
and C_3+ for the same period be presented in the supplement for those periods? It could
provide some information on the mixing of particles, and help interpret the difference in
coating factors observed between the two sites.
- Figure S22: Could you add in the caption that the data corresponds to the residential
site?

References

Gustafsson, Ö.: Photochemical degradation affects the light absorption of water-soluble
brown carbon in the South Asian outflow. Sci. Adv., 5, eaau8066,