Final author comments to the manuscript of "Sources of black carbon at residential and traffic environments"

Response to Referee comments

We thank the two Referees for their valuable comments on our manuscript "Sources of black carbon at residential and traffic environments". We think that the revision we made to the manuscript, based on the comment of the Referees, has improved the quality of the manuscript significantly. Most of the changes suggested by the Referees were implemented by adding requested details or discussion to the manuscript but we also added eleven figures to Supplemental material. In addition, some minor changes have been made throughout the manuscript to improve the grammar. All the changes to the manuscript have been made by "Track changes" mode. Point-by-point responses to the comments of the Referees are given below and also in separate author's responses.

Referee #1

We thank Referee #1 for her/his elaborate and valuable comments. We have responded all the comments below and the changes to the manuscript have been made in "track changes" mode.

Specific comments

1) 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?

    Reply and changes in manuscript: The quantification of BC by the SP-AMS was not the focus of this study since the results of PMF don't depend on the exact concentrations in general. Therefore, the results reported in this paper were calculated with a constant CE value of 1. However, for the calculation the coating factor, the accuracy of AMS concentrations is more important. Therefore, we have added to supplemental material a comparison of the sum of the SP-AMS species (excluding rBC) and BC from the aethalometer with the concurrent PM$_1$ measurements (Grimm model EDM 180, Grimm Aerosol Technik, Ainring, Germany) (residential site). The sum of the SP-AMS species and AE33 BC made on average 71% of Grimm PM$_1$ with a moderate correlation (Pearson's correlation coefficient r = 0.888; Fig. S4a). Similar comparison was done also by using the composition dependent CE (CDCE) calculated according to Middlebrook et al. (2012). With the CDCE, the sum of the SP-AMS species and AE33 BC was closer to PM$_1$ from Grimm (~85% of PM$_1$), however, the correlation was slightly poorer (r = 0.876; Fig. S4b). For the street canyon, the PM$_1$ comparison could not be calculated as there was no PM$_1$ mass measurement at that time. Based on these calculations, CE value of 1 was not changed, however, the discussion on the uncertainty in the coating factor due to the CE =1 was added to Chapter 3.3. Also a paragraph discussing the PM$_1$ and CE comparisons was added. The time series of CDCE values at the residential and street canyon sites were added to supplemental material (Fig. S4 and S5).

    As suggested by the Referee, the comparison of rBC from the SP-AMS with BC from AE33 have also been added to the supplemental material (Fig S3). It shows that at the residential site the SP-AMS gave roughly half of AE33-BC (48 %) with a strong correlation (r = 0.961). At the street

canyon, the correlation was poorer (r = 0.539), however, the difference between rBC from the SP-AMS and AE33-BC was smaller (rBC 65 % of BC).

Reply: SP-AMS run purely in dual mode.

Changes in manuscript: Additional correction was that the relative ionization efficiency for rBC was changed from 0.1 to 0.05 after the original rBC calibration data was inspected again in detail.

Reference

Middlebrook, A. M., Bahreini, R., Jiménez, J. L., and Canagaratna, M. R.: Evaluation of composition-dependent collection efficiencies for the Aerodyne aerosol mass spectrometer using field data, Aerosol Sci. Technol. 46, 258–271, 2012.

2) 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?

Reply: We believe that the discrepancies between BC detected by the SP-AMS and BC from the AE33 are mostly due to the poor laser alignment and therefore incorrect BC RIE calibration with Regal black. Also, the difference between SP-AMS and AE33 results can be due to the size limit of the aerodynamic lens that misses the smallest fraction of BC in the SP-AMS. That can be specially seen at the street canyon site that has the smallest BC particles and a large difference between BC_{HOA} and BC_{ff} (Fig. S39). It impact of imperfect laser alignment on BC RIE is discussed in detail in Referee #2 comment 3).

Changes in manuscript: The comparison between rBC from the SP-AMS and BC from the AE33 have been added to supplemental material (Fig S3).

Reply: MAAP measured only at the background site in Luukki that had no other BC instrument running at the same time.

3) 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.

Reply: Coating factor was calculated similar to Drinovec et al. (2017) and the reference has been added. Coating factor was calculated by summing all the SP-AMS inorganic and organic species (excluding rBC) and dividing it by BC from the AE33. The assumptions in the coating factor calculations were: (1) CE of 1 for the AMS species, (2) default RIE for organics and inorganic species and (3) similar particle size for the AMS species and BC from the AE33. We can estimate that the uncertainty for CE=1 is 30% and RIE for the each AMS species is 20%. Based on these uncertainties, the total uncertainty due to CE and RIE was ~40 %. The uncertainty due to the different particle sizes of the SP-AMS and AE33 is difficult to estimate quantitively. Qualitatively, we can assume that the AMS is missing particle mass below <50 nm and therefore the coating factor can be slightly underestimated. The discussion on the uncertainties in the

coating factor has been added to manuscript. The comparisons of SP-AMS results with AE33 and PM$_1$ from Grimm have been added to supplemental material (Figs S3-S4) and discussed in text.

Changes in manuscript: the reference for the coating factor calculation and the discussion on the uncertainties in the coating factor have been added to manuscript.

4) Any reason for using only C_2^+, C_3^+ and C_4^+?

Reply: The reason for using only C_2^+, C_3^+ and C_4^+ was that the signal was close to the detection limits for the larger carbon fragments due to relatively poor laser alignment and small rBC concentrations at the sites. C^+ was excluded because it can have a significant contribution from organics.

Changes in manuscript: A sentence: "For the larger carbon fragments, the signal was close to the detection limit, and therefore, they were not included in the PMF analysis." has been added.

5) 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?

Reply: A large contrast in rBC contributions from 4 to 5 factors at the residential site can be explained by the splitting of OOA factors when changing from 4 to 5 factors. Moving from 4 to 5 factors, the rBC contribution in HOA increased slightly and BBOA decreased but those changes were not large. In terms of OOA factors, rBC seemed to be divided differently between OOA factors when changed from 4 to 5 factors as rBC contribution decreased in SV-OOA, increased significantly in LV-OOA and was small in the new factor LV-OOA-LRT. In total, the rBC contribution in OOAs was 0.040 in 5 factor solution that was larger contribution than in OOAs in 4 factor solution (0.029) or LV-OOA in 3 factor solution (0.011) showing that the rBC contribution increased in OOA factors when the number of factors increased.

The Referee is right that the composition of BBOA in terms of f$_{C2H4O2}$ differs at the sites, which could be due to the fact that BBOA detected at the street canyon is more aged than at the residential site as it is probably transported further away. Therefore its mass spectra resembles more SOA mass spectra.

Changes in manuscript: Sentences: "Also the mass spectra of organics in BBOA differed between the residential and street canyon sites as the contribution of $C_2H_4O_2^+$ (at m/z 60) and $C_3H_5O_2^+$ (at m/z 73) was much larger at the residential site compared to the contribution at the street canyon indicating that BBOA at the street canyon site was probably more aged with its mass spectra resembling SOA. That can make the separation of primary and secondary OA factors more complicated at the street canyon site." have been added to manuscript.

6) Line 469-470: "This suggests that $BC_{LV\text{-}OOA}$ 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)?

   Reply: That is a good statement. BC size distributions can give some clue regarding the source, but for example in terms of SOA, the size distributions do not give that much information on the original source. It is possible that BC at larger sizes was also related to SOA particles, however, the measurement period was conducted in autumn when the SOA formation in Finland is minor due to low UV radiation and small concentrations of SOA precursors.

   Changes in manuscript: "clearly" has been changed to "possibly"

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

   Reply: Calculating average angstrom exponent for BC_{BBOA} and BC_{HOA} dominating periods is a good idea and those values were calculated as suggested by the Referee.

   Changes in manuscript: Sentences: "Those cases corresponded to large BC_{BBOA} fraction as the α values calculated specifically for the periods dominated by BC_{BBOA} or BC_{HOA} (>50 % of BC_{BBOA} or BC_{HOA}) were 1.36 and 1.25 at the residential site and 1.24 and 1.12 at the street canyon, respectively." have been added to manuscript.

8) Line 511-513: "Biomass burning BC obtained from two source apportionment methods followed very similar time trend, however, BC_{wb} calculated with the α_{ff} and α_{wb} values of 1 and 2, respectively, displayed smaller values than BC_{BBOA} or BC_{wb} calculated with the α_{ff} and α_{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.

   Reply: A comparison of BC_{wb} and BC_{BBOA} concentrations (in linear scale) with a linear fit have been added to supplemental material (Fig. S36). It shows that for the residential site, BC_{wb} calculated with the α_{ff} and α_{wb} values of 1 and 2 gives only 57 % of BC_{BBOA} or BC_{wb} calculated with the α_{ff} and α_{wb} values of 0.9 and 1.68, respectively. For the street canyon, BC_{wb} was approximately 3.8 and 7.9 times larger than BC_{BBOA} calculated with α_{ff} and α_{wb} values of 1 and 2 and 0.9 and 1.68, respectively. Also, the correlation between BC_{wb} and BC_{BBOA} was very poor.

   Changes in manuscript: A comparison of BC_{wb} and BC_{BBOA} concentrations with a linear fit have been added to supplemental material (Fig. S36). We also replaced "smaller values" by "half of the values". The diurnal variation for BC_{wb} and BC_{BBOA} at the residential site have been added to supplemental material (Fig. S37). We also added text: "In terms of diurnal variation (Fig. S37), BC_{BBOA} and BC_{wb} both had a maximum in the afternoon and a second peak in the morning, BC_{BBOA} increasing in both cases slightly later than BC_{wb}. The reason for the delay can speculated to be due to the fact that organics and BC can be emitted at different ratios at different stages of burning (e.g. Kortelainen et al., 2018), the time behavior of BC_{BBOA} being determined by biomass burning organics due to the used PMF method."

9) 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 $BC_{LV\text{-}OOA\text{-}LRT}$) with potentially different absorption properties influencing the site?

Reply: Sentence: "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 BC_{BBOA} varied more clearly during the course of the day." was incorrect as "BC_{wb}" and "BC_{BBOA}" should have been "BB% values from the aethalometer model and AMS PMF, respectively". This sentence has been corrected.

It is possible 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 $BC_{LV\text{-}OOA\text{-}LRT}$) with potentially different absorption properties influencing the site. We have added this remark to Chapter 3.3. (lines 601-602) and conclusions (lines 648-650).

Changes in manuscript: Incorrect sentence has been modified. Diurnal trends of BB% values from AMS PMF and aethalometer model at the residential site have been added to supplemental material (Fig S27). Lines 601-602 and lines 648-650 have been added.

Technical corrections and minor comments

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

   Reply and changes in manuscript: the title has been changed to " Sources of black carbon at residential and traffic environments obtained by two source apportionment methods"

2. 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.

   Reply and changes in manuscript: This was a good point. We added to abstract: "The measurement campaign was conducted at the residential area in winter-spring 2019 whereas the at the street canyon the measurements were carried out in autumn 2015." We also added to conclusions: "When comparing the sites, it should be remembered that the measurements were conducted at different years (residential site in 2019 and street canyon in 2015) that may also impact the source contributions. For example, Luoma et al. (2021) have shown that BC related to traffic has decreased in Helsinki area in recent years, which may overrate traffic related BC at the street canyon relative to year 2019."

3. 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.

Reply and changes in manuscript: A map showing the locations of the three measurement sites has been added to the supplement (Fig S1). The distance between the sites can be seen from the scale given in the bottom left corner. The potential surrounding sources were difficult to include in the map but for example the location of the major roads can be seen in the map.

4. 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?

    Reply and changes in manuscript: The Referee is correct that the difference between PMF and aethalometer model results can be due to the fact that the aethalometer model cannot separate as many sources as AMS-PMF due to similar optical properties of different OA sources. Therefore we have added to abstract …"or it could not distinguish between as many sources as PMF due to the similar optical properties of the BC sources." We also added to conclusions: "…or the aethalometer model was not able to distinguish between as many sources as PMF due to similar optical properties of the BC sources."

5. Line 225: "BBOA concentration was a slightly smaller in daytime", was slightly smaller?

    Reply and changes in manuscript: "a" has been deleted

6. 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?

    Reply and changes in manuscript: comma added

7. Line 361-362: "That is a slightly larger contribution that the campaign-average percentage obtained in this study", changed to "larger contribution than".

    Reply and changes in manuscript: "slightly" removed

8. Line 382: Change (Fig. 23) to Fig. S23.

    Reply and changes in manuscript: changed as suggested

9. Why are the elemental ratios missing from Figure S6 to S8?

    Reply and changes in manuscript: Elemental ratios were missing in Figures S6-S8 because the CHO^+ fragment at mass-to-charge ratio (m/z) 29 needed to be excluded from the PMF data matrix, and CHO^+ is crucial for the source apportionment. The reason for that was that N_2^+ at m/z 28 was fluctuating due to the unresolved issue in the instrument. That fluctuation caused a large uncertainty for the determination of the isotope $^{15}NN^+$ at m/z 29 and also for CHO^+ that was overlapping $^{15}NN^+$. The exclusion of CHO^+ was explained in Chapter 2.3 and in figure captions S6-S8: "Elemental ratios were not calculated due to the exclusion of CHO^+ from the input matrix." No changes in the manuscript.

10. Line 87: "the light absorption in different wavelengths", changed to "at different wavelengths"'

Reply and changes in manuscript: "in" changed to "at"

11. Line 610-614: Barreira, L. M. F et al. 2021., has been accepted

    Reply and changes in manuscript: Barreira et al. (2021) has been changed

12. 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".

    Reply and changes in manuscript: Details on the LRT episodes have not been published yet. Therefore "in the other paper" was changed to "in another paper" as suggested by the Referee.

13. 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?

    Reply: The average contribution of BC_{SV-OOA} was quite small at the residential site (9%) and therefore we did not focus that much on its origin. Based on the mass spectra of organics, SV-OOA was rather oxygenated and therefore has probably been formed from the oxidation of HOA and BBOA, and because if its semi-volatile nature, the SV-OOA concentrations were smaller in daytime.

    changes in manuscript we added to sentence: "Based on the mass spectra of organics in the SV-OOA factor and its diurnal trend, it can be speculated that BC_{SV-OOA} was related to the aging of local traffic and biomass burning emissions."

14. Line 401 and Figure 2: specifying Lukki background site, instead of background site might avoid confusing it with Street Canyon site.

    Reply and changes in manuscript: "Luukki" added to text

15. 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?

    Reply: Unfortunately, we did not have any data supporting this statement. We know that there are some houses only few hundred meters from the Luukki station, however, there is a forest between so that the impact of the houses is supposed to be rather small.

    Changes in manuscript We added sentence: "Although the background site of Luukki is located in a sparsely inhabited area, there are some houses only few hundred meters away from the site, which can have an impact on the measured concentrations."

16. 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 (Nicolae et al., 2013, Dasari et al., 2019…), which could also explain the difference in AAE if the BBOA are not locally emitted.

Reply: That is a good addition from the Referee.

Changes in manuscript We added to sentence: "However, studies have also reported a decrease of α in biomass burning emissions by photooxidation during atmospheric aging (Nicolae et al., 2013, Dasari et al., 2019), which can explain the difference in α if the biomass burning particles are not locally emitted."

17. Line 518-520: "Biomass burning BC obtained from two source apportionment methods followed very similar time trend, however, BC_{wb} calculated with the α_{ff} and α_{wb} values of 1 and 2, respectively, displayed smaller values than BC_{BBOA} or BC_{wb} calculated with the α_{ff} and α_{wb} values of 0.9 and 1.68, respectively." As mentioned before, the comparison between the diurnal profiles of BC_{wb} and BC_{BBOA} could be presented in the supplement.

Reply and changes in manuscript: diurnal profiles have been added to supplemental material (Fig. S37).

18. Figure 2.b: Y axis legend should be corrected from "ug (m^{-3})" to (ug m^{-3}).

Reply and changes in manuscript: changed as suggested

19. 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 BC_{BBOA} 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.

Reply: Unfortunately, the size distribution for the C_3^+ fragment needs the analysis of high resolution PtoF and that data is not available. Instead, we could present the unit mass resolution size distribution of m/z 36, however, that has an interference from HCl^+ fragment at m/z 36 that, based on the high resolution mass spectra, contributed 60, 54 and 88% of total signal at m/z 36 in BC_{BBOA} dominated, BC_{LV-OOA_LRT} dominated and $BC_{LV-OOA + SV-OOA}$ dominated periods, respectively. Therefore, the size distributions for the m/z 36 were not included in this article.

Changes in the manuscript: We have added "dominated" to legends in Figs 3b and 5c in order to make them more clear. The size distribution of rBC in the LRT dominated period has been added to Fig. 3b as suggested by the Referee. Also the average size distributions of organics in BC_{BBOA}, $BC_{LV-OOA-LRT}$ and $BC_{LV-OOA + SV-OOA}$ dominated periods at the residential site and BC_{HOA} and BC_{LV-OOA} dominated periods at the street canyon have been added to supplemental material (S31). We also added text regarding residential site: "The size distributions of organics were very similar to those of BC during the three periods (Fig. S31a) indicating that BC was mostly internally mixed with organics." and regarding street canyon site: "During the BC_{LV-OOA} dominated period, the size distributions of BC was similar to that of organics (Fig. S31b), but in the BC_{HOA} dominated period, there was slightly more BC in the first mode (at ~130 nm) relative to organics than in the second mode (at ~300 nm)."

In terms of coating factors, the size distribution of organics did not provide that much new information. We can see that rBC and organics from the SP-AMS are mostly in the same particle

size indicating that they seemed to be internally mixed, however, due to e.g. aerodynamic lens in the AMS, we may miss smaller (< 50 nm) particles especially at the street canyon site. These small BC particles are included in eBC measured by the AE33 that may result in the underestimation of coating factor. Uncertainties in coating factor calculation has been discussed in comment 3) in more detail.

20. Figure S22: Could you add in the caption that the data corresponds to the residential site?

Reply and changes in manuscript: "at the residential site" added

References

Dasari, S.; Andersson, A.; Bikkina, S.; Holmstrand, H.; Budhavant, K.; Satheesh, S.; Asmi, E.; Kesti, J.; Backman, J.; Salam, A.; Bisht, D. S.; Tiwari, S.; Hameed, Z.; Gustafsson, Ö.: Photochemical degradation affects the light absorption of water-soluble brown carbon in the South Asian outflow. Sci. Adv., 5, eaau8066, https://doi.org/10.1126/sciadv.aau8066, 2019

Nicolae, D., Nemuc, A., Müller, D., Talianu, C., Vasilescu, J., Belegante, L., & Kolgotin, A. : Characterization of fresh and aged biomass burning events using multiwavelength Raman lidar and mass spectrometry. Journal of Geophysical Research: Atmospheres, 118(7), 2956-2965, https://doi.org/10.1002/jgrd.50324, 2013

Sandradewi, J., Prévôt, A. S., Szidat, S., Perron, N., Alfarra, M. R., Lanz, V. A., Weingartner, E., and Baltensperger, U.: Using aerosol light absorption measurements for the quantitative determination of wood 825 burning and traffic emission contributions to particulate matter, Environ. Sci. Technol., 42, 3316–3323, https://doi.org/10.1021/es702253m, 2008

Referee #2

We thank Referee #2 for her/his valuable comments. We have responded 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 and changes in manuscript: 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 and changes in manuscript: 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 and changes in manuscript: 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 and changes in manuscript: 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 site 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. No changes in manuscript.

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.

Changes in manuscript: 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 Fig. R1).

Changes in manuscript: 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)."

Reply: 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. Below (Fig R2) 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 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 $BC_{LV-OOA-LRT}$, 1.2–1.3 for BC_{LV-OOA} and 1.25–1.42 for BC_{SV-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.
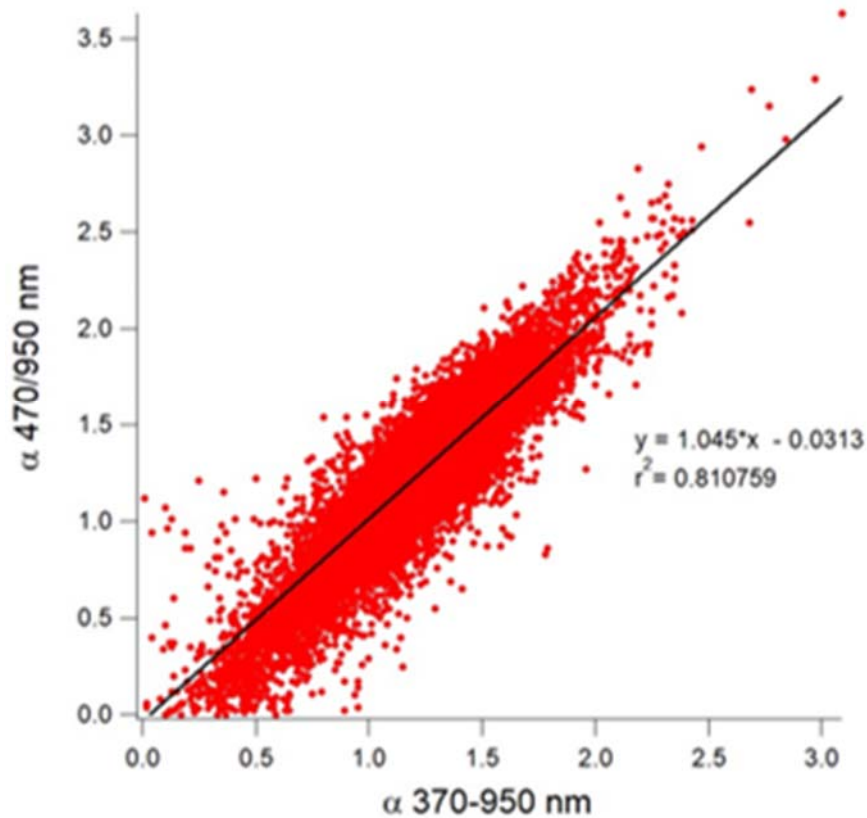
Figure R1*. Comparison of α values calculated with only the wavelengths of 470 and 950 and with all the wavelengths from 370 to 950 nm. One minute time-resolution.*
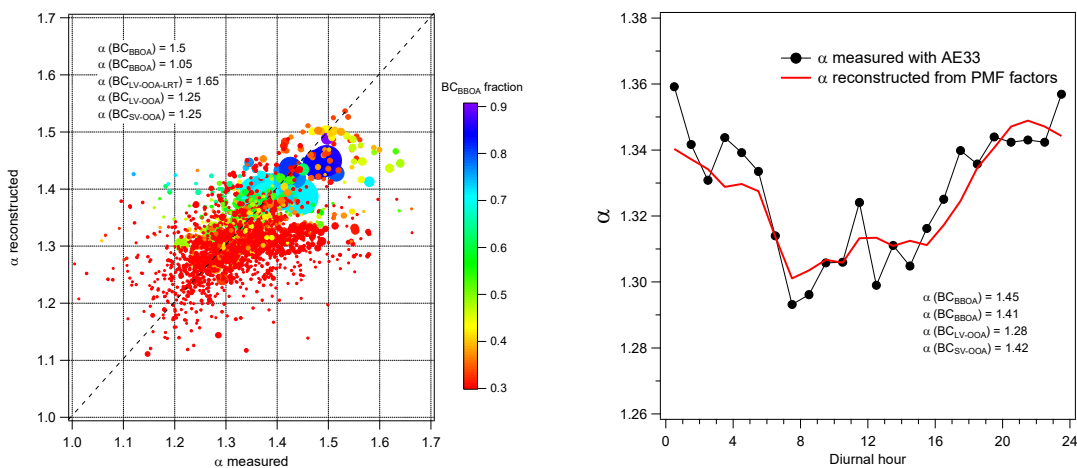

Figure R2*. Comparison of α values measured with AE33 and reconstructed from the AMS PMF data with multi-linear regression at the residential site. Scatter plot (left) and average diurnal variations (right).*