Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-231

Anonymous Referee #1

Referee 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-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 $BC_{LV\text{-}OOA}$ 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)?
- 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 α_{ff} and α_{wb} values of 1 and 2, respectively, displayed smaller values than BCBBOA 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.
- 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?

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 (Nicolae 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, 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.
- 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 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.

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

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