We thank all reviewers for their comments. Our answers, in normal text, are presented below the comments, which are bold. Changes to the manuscript are in italic with line numbers of the modified manuscript.

REVIEWER #1

This manuscript (acp-2022-156) reports black carbon concentrations and physical properties in both urban and rural environments in Sweden. The study aims to investigate whether the rural air was influenced by traffic emissions and long-term aerosol transportation. I am glad to see that the authors have done various analyses using the available measurements (such as BC coating estimation, trajectory analysis, and OA source apportionment) and reported many observations. However, the Results and Discussion section missed many explanations, and the logistic of some subsections is not clear (See my general comments below). I suggest a major revision and restructuring of the paper before being acceptable.

Major comments:

1. I understand that the authors had to move the SP2 between the two campaigns, and some instruments were not available during some periods of the study. But this experimental limitation significantly hinders the authors' goal to explore the influence of urban emissions on the rural air quality, because the two campaigns were not conducted simultaneously. The authors use Figure S1a to justify that the BC concentrations at the rural site did not change drastically from July to October, but this result is not enough to support the research's goal because, besides BC, many other atmospheric components and meteorology can be changed between the two campaigns. I am not against the author's effort to compare the urban against the rural environments, but the author should be careful to use asynchronous observations to explore how urban emissions affect rural background air and draw the conclusion that "local abatement strategies aimed towards reducing BC emissions from traffic sources will thus have an effect in reducing the BC mostly limited to the urban population." Overall, I strongly suggest revising the goal and the conclusion.

The aim of the study was to compare the BC properties between the sites, and to look for influence of the urban are on the rural background air. We believe that it has been clearly shown that the difference in BC properties is driven by the local traffic emissions in the urban area (Figures 2, 3 and 4). When it comes to the influence of the urban area on the rural background air, single plumes were hard to distinguish during the campaigns and therefore we expanded the analysis to the full year using available eBC and trajectory data (Figure 7), hence the cited conclusion in the comment is NOT from asynchronous observations. However we agree that the last paragraph can be misinterpreted and removed that part from the conclusions and slightly rephrased the paragraph above to be more suitable as a final conclusion. It now reads as follows:

L381. *Plumes from the nearby urban site to the rural site were not clearly distinguishable during the field campaigns. Trajectory analysis of the full year of 2018 show that significant increases in eBC concentrations at the rural site for air masses passing over the Malmö/Copenhagen area are only clearly seen during days with low precipitation. This increase, however, is small in comparison with the influence of long-range transported BC. Transport of BC from continental, and especially eastern, Europe is what governs the BC concentrations in southern Sweden background air, when looking at eBC from the full year of 2018.*

2. Section 3.6 (origins of BC in the rural background air) is not well structured. The authors presented a lot about the origins of aerosols at the urban site (lines 331 -360), but didn't really answer why the BC is from at the rural site. It seems that the answer is related to Figure 7, but the paragraph above Figure 7 is hard to understand. The authors should consider not using a histogram to present Figure 7, then rephrase the paragraph to better justify how wind directions and precipitation affect the rural background air.

We have tried to simplify the formulations and refer to section 2.2.8 for details on the analysis, to make the first paragraph of section 3.6 more clear. Further, we also added the median numbers in the legend of Figure 7.

L302. Figure 6 shows the wind direction probability together with eBC from the five weeks rural campaign. The Malmö/Copenhagen area is in the direction 200 – 230 degrees as seen from the rural site. Based on the HYSPLIT trajectory analysis (Sect. 2.2.8) , some peaks in eBC, non-refractory PM1, and total particle number concentrations at the rural site coincide with air masses originating over Malmö/Copenhagen. These urban plumes were discernible when the air mass was relatively clean (typically of western or north-western origin). However, no conclusive results on the influence of the urban site on rural levels could be derived from analysis of the limited field campaign data. Also when analyzing the complete eBC dataset from 2018 there is no significant difference in the median eBC concentration for air masses with and without influence from Copenhagen and/or Malmö, according to the trajectory analysis. However, if we only consider air masses with insignificant precipitation within 48 hours upwind the rural site (i.e. less than <1 mm precipitation according the HYSPLIT model), the measured eBC median concentration is significantly higher in the air masses that have moved over Copenhagen/Malmö, 354.7 ng m-3 compared to 226.9 ng m-3 without influence from Copenhagen/Malmö. Similar results are also found when only the air masses originating from SW is considered (see Fig. 7 and Tables S2-S4). Most likely the contribution from the BC emissions from Copenhagen/Malmö is not apparent when analyzing the whole eBC dataset from

2018 because the SW air masses are generally influenced by more precipitation (more BC wet scavenging) compared to other air masses. The HYSPLIT trajectory simulations for the complete year show that 66 % of all air masses that passed over Malmö/Copenhagen were influenced by >1 mm precipitation within 48 hours upwind the rural site, compared to 39 % for all other air masses. The lowest median eBC concentration is found in air masses from NW (101 ng m-3) followed by NE air masses (149.9 ng m-3) and SW air masses (192.0 ng m-3). SE air masses clearly stand out from any other air masses with a median eBC concentration of 563 ng m-3. This can partly be explained by the low probability of precipitation in SE air masses. Only 23 % of these air masses are influenced by >1 mm precipitation within 48 hours upwind the rural site. However, also when considering the effect of precipitation upwind Hyltemossa the SE air masses have a median eBC concentration that is ~2 times larger than the SW air masses.

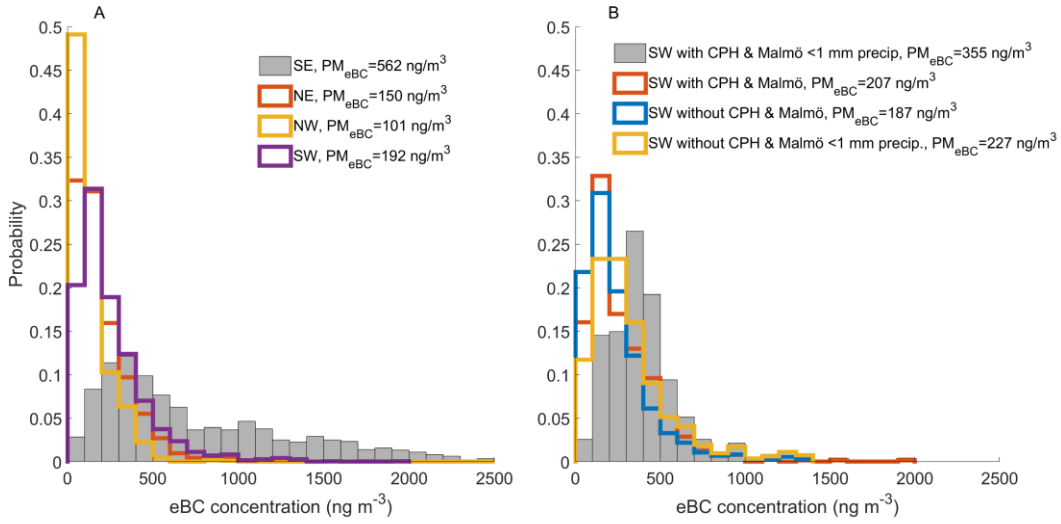


Figure 7. Trajectory analysis of different air mass origins, with corresponding median eBC mass concentrations in the legend. Panel A shows histograms with the observed eBC from the Aethalometer at Hyltemossa from year 2018 for air masses originating from SE, NE, NW and SW as defined in Fig. S3. Panel B shows the eBC histogram from all SW air masses with or without influence from the Copenhagen and Malmö region.

3. The introduction section should include more details. First of all, the authors should add references to the statements in the first paragraph. Second, the second paragraph is not directly related to the research question. The paragraph is a very general description of BC measurements and properties. More details should be added, including how BC properties vary from different emission sources, how BC properties change after mixing with other organic compounds, and what the authors mean by the importance of the BC mixing state. Third, the authors should add another paragraph to introduce the discrepancies between eBC and refractory BC measurements since the authors present such results in the Result and Discussion. Lastly, if the author wants to keep using the current title, an overview of the influence of long-range transportation of aerosols onBC properties from the other studies is necessary.

We added references to the last sentence:

L39. *Soot measurements are complicated by the fact that the ambient aerosol is a dynamic mixture, and that soot from different sources may not possess the same properties in terms of chemical content (Malmborg et al., 2019), light absorption (Sandradewi et al. 2008), size (Schwarz et al., 2008), and toxicity (Hakkarainen et al., 2022).*

The second paragraph discusses the importance measuring not only absorption but size and mixing state of BC. This is in our opinion highly relevant properties that we want to compare between the two sites. The sentence on mixing state says that it is important to know to understand the light absorption. We added some details on BC transformation in the atmosphere. The paragraph now reads:

L43. *Although BC mass concentrations are routinely estimated from optical methods, BC number concentrations, size distributions and mixing state are rarely measured. In a previous study it was shown that global aerosol microphysics models underestimate the BC particle size, by a factor of ~2-3, while overestimating the number concentrations, by more than a factor of 3, compared to airborne measurements using single particle laser induced incandescence (Reddington et al., 2013). BC from different sources have different size distributions (Schwarz et al., 2008; Laborde et al., 2013; Saarikoski et al., 2021), that affects both transport (lifetime) and light interactions (Hinds, 2012), as well as deposited dose (Alfoldy et al., 2009; Rissler et al., 2012). Upon ageing in the atmosphere, fresh BC particles will obtain a coating layer consisting of*

condensed materials changing both their hygroscopicity (Swietlicki et al., 2008) and light absorption properties (e.g. Zhang et al., 2018). This transformation depends on atmospheric conditions and constituents but can be on the order of hours under favourable conditions (Eriksson et al., 2017). Recent studies have pointed to the importance of BC mixing state, governed by emission source and atmospheric ageing, in understanding the light absorbing properties, and hence climatic impacts, of BC containing particles (e.g. Liu et al., 2017; Liu et al., 2019; Fierce et al., 2020; Yuan et al., 2020). These properties of BC, that are crucial to understand both health and climate impacts, are not measured by the BC measurement techniques commonly used by monitoring networks.

The discussion on rBC and eBC is included in the Discussions section but we don't think it is necessary to include in the introduction.

Specific comments:

1. Line 39: The authors should specify what kind of severe climate and public health effects can be introduced by BC.

We prefer to refer the readers to the cited literature instead.

2. Line 66: Describe what the two campaigns are. Actually, the rural site, rural campaign, urban site, urban campaign are very confusing. I suggest renaming therural campaign and urban campaign. Maybe just Campaign #1 and #2.

We don't understand what is meant by "what the two campaigns are"? To satisfy another reviewer we added Figure 1 which shows an overview of the urban and rural measurements, when they were undertaken and what data was used. We don't think renaming the campaigns #1 and #2 would be less confusing, since we would still have to discuss urban and rural settings and sites.

3. Line 67: Is the results in Figure S1 for the rural or the urban site? The main text says rural, but the caption of Figure S1 says urban.

It's the rural site, thank you for noticing. We changed the caption.

4. Line 71 and Line 78: The two sites either measures PM2.5 or PM10. Does this affect any results?

This is our answer to a similar question in review round 1:

Good point. The bias that could have been introduced is that the measured BC concentrations at the urban site was too low. We don't believe a lot of BC mass is present in the larger particles. Especially the locally emitted particles will not have had time to coagulate and grow. Viidanoja et al. (2002) showed that typically more than 90% of BC resided in PM2.5 at an urban site in Finland. Either way, it is highly unlikely that this would affect our conclusions.

Viidanoja, J., Sillanpää, M., Laakia, J., Kerminen, V.M., Hillamo, R., Aarnio, P. and Koskentalo, T., 2002. Organic and black carbon in PM2. 5 and PM10: 1 year of data from an urban site in Helsinki, Finland. Atmospheric Environment, 36(19), pp.3183-3193.

5. Line 101: Was 10 and 1-5% of the data discarded or used in the analysis? Please specify.

That is what was saved. Changed to:

L101. 10 and 1-5 % was saved in the rural and urban campaigns respectively

6. Line 151: The authors should consider adding a time-series figure of BC and C4H9+concentrations during a traffic plume, then label the three windows and their durations.

We agree this could be a nice analysis, but feel it would not add to the current manuscript.

7. Line 186: I don't get how the authors concluded that "these corrected values are closer to the true values". What do the authors mean about "true values"? Please explain in the main text.

Figure S2 shows that the losses in the turbulent flow inlet is 20-30% for particles of diameters between 100-1000 nm. The SP2 number and mass size distribution peaks at below 100 nm and ~150 nm respectively, but that does not include coatings. Hence from the calculated losses, we estimated and corrected for 25 and 30% losses in Number and mass concentrations.

We changed the wording to not confuse the reader:

L185. Based on these calculations and the measured rBC size distribution (discussed below), the SP2 mass and number concentrations measured at the rural site have been adjusted to correct for 25 and 30 % losses respectively since adjusting by size is not possible because the size of rBC cores including coating was not measured.

8. Line 216: Why does the greater concentration measured at the curbside suggest that these measurements are indicative of the city? Please explain in the main text.

It's actually the similarity in concentrations, shown in Figure S4, that we think point to the validity of the curbside site as an indicator for the whole city. The text now reads

L214. *The comparison between the urban street-level and urban background eBC levels are very similar in time-series but with a lower daily maximum for the roof-top measurements (Fig. S4). This suggests that the curbside measurements are indicative for the city.*

9. Line 266: Show statistical results to justify "statistically significant".

We added the p-value from a two tailed Z-test on mass size distribution GMD.

L265. The difference in mass GMD is statistically significant (P<0.01) and can be explained by different BC particle sources, air masses and coagulation.

10.Lines 275-293: Do the authors have any explanation about why 50 and 75 nm particles are unimodal, but 100 and 150 nm particles are bimodal?

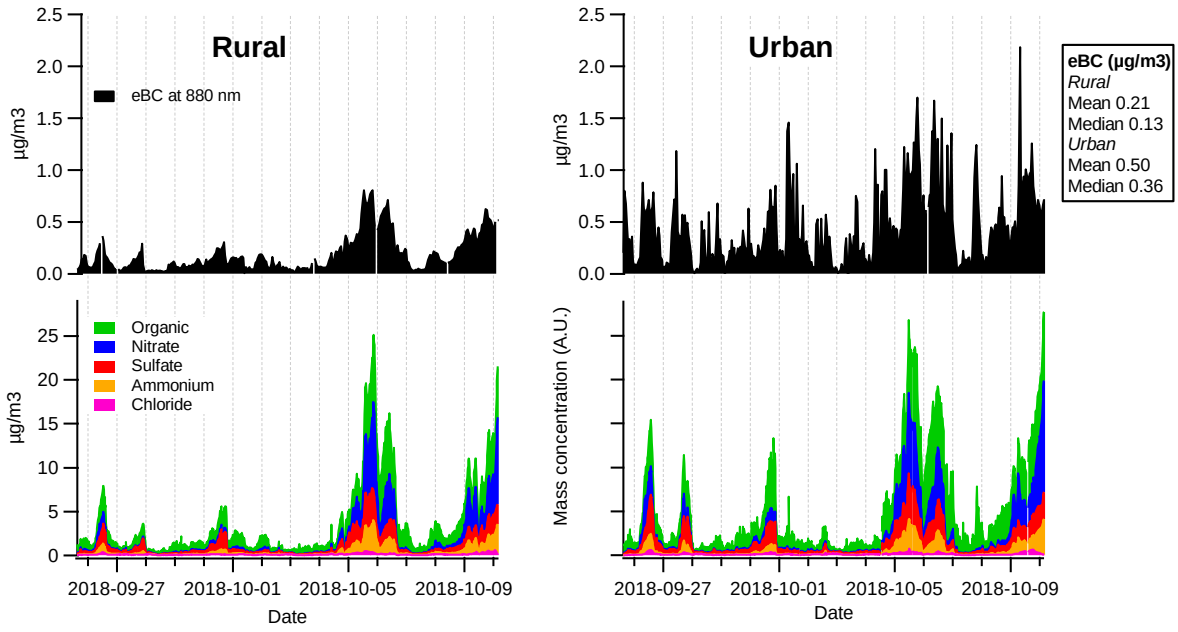
The APM measurements were taken during daytime, so there should exist both fresh and aged soot particles. The reason we didn't see bimodal distributions for the smaller sizes may have to do with the ambient total and soot size distributions (and chemical composition) but it is also harder to resolve effective densities of small particles which which requires smaller steps in voltage. Further, for fresh soot agglomerates, the smaller they are, the higher density they will have. This means that despite no coating the calculated effective density of fresh soot and Aitken mode aged particle may overlap.

11.Line 307: I don't find the results of HYSPLIT trajectories in the main text and SI.

These results are in section 3.6, Figure 7, and tables S2, S3 and S4.

12.Line 340: How do the authors draw the statement that "approximately half of the mass at the urban site is due to long-range transport"? First, how "half of the mass" is estimated? Second, what is the evidence for "long-range transport" in Figure 8?Please explain this sentence in more detail in the main text.

In Figure 8 we want to show the similarities in temporal pattern of pm1 non refractory components as well as the dissimilarity of eBC. The fact that approximately half of the mass is from long distance transport is deduced from the assumption of similar air masses (as is seen in non-refractory components) and the average concentrations at both sites. To help the reader, we added the mean and median concentrations to the figure. See below.



13.Figure 8: Why does panel d have a different y-axis label from panel c?

We only use the fractions of chemical species from the urban dataset. This is described in the methods, mentioned in the caption and also discussed with a previous reviewer, see below.

For the SP-AMS, there was confusion around the measured mass loadings (based on field calibration with 300 nm mobility diameter ammonium nitrate particles). The rural loadings were higher throughout the campaign, despite very similar time trends for all non-refractory species (see Figure 7). We found the concentration in the rural data well supported by DMPS (same site) and FIDAS (nearby background site). We do not have measurements for a similar evaluation of the urban SPAMS. Hence, the combined evidence suggests the urban absolute concentrations from SPAMS are too low, which we attribute to calibration issues. Notably this would not have been detected if we were not comparing with another SPAMS downwind (at the rural site) which was properly supported by auxiliary measurements.

This issue also affects Figure 9, where we also removed the absolute values now.

14.Line 353: The authors showed the strong correlation between non-refractory PM1 concentration and thickly coated BC. What is the correlation between non-refractory PM1 concentration and non-coated BC (i.e., fresh BC)? If the latter correlation is also strong, the statement "the BC coatings are similar in composition to nonrefractory-PM1" is over-interpreted.

From the delay time method of the SP2, described in section 2.2.2, the coating is divided into an either thickly or thinly coated fraction. Therefore, the thinly coated particles (fresh BC) is simply 1-thickly coated. Hence, there is an anticorrelation for this dataset.

15.Line 368: I don't think a factor of 2.2-4 between urban and rural environments is "surprisingly low". The authors should consider using another word.

We deleted this sentence.

16.Line 388: As I mentioned in the major comment #1, the authors should rephrase this sentence.

See answer to major comment 1 above.

REVIEWER #2

General

The paper reports BC measurements in southern Sweden including source analyses and interpretations. In my opinion it suits well to the journal. This is the corrected version of the manuscript where the authors have replied to other reviewer's comments. For me the replies look fine, I didn't find much to correct any more. However, I did still find some things that I think should be corrected. They are in the detailed comments below.

Detailed comments and questions

L 83. " An Aerosol Particle Mass Analyzer (APM) was at a later stage deployed ..." Show the deployment period also in Fig. 1, just like for all the other instruments.

The APM measurements were undertaken the year after. Including that in the figure would make it harder to resolve the intensive campaign dates, which we belive should be the main focus. We changed the caption to:

Figure 1. Overview of the data and instruments used. Not shown in the figure are DMA-APM measurements of effective density during spring 2019.

L123. "Default corrections for filter scattering (Cref = 1.39) and loading effects were used (Weingartner et al., 2003)." The Weingartner et al. (2003) correction was developed for the older AE version, AE31. It is good for it. However, the AE33 calculates the loading corrections already during the measurements by using the dual-spot method and the loading correction function presented by Drinovec et al. So, how have you really used the Weingartner et al. correction? Correction on top of another correction? Or have you used the raw data and reprocessed that according to Weingartner et al.? If so, why, what is the justification? That makes no sense. According to the "Data availability" section you have uploaded the EBC data to EBAS. They do not accept such double corrections for level 0, level 1 or level 2. So, if you really have done this you should recorrect and resubmit the data to EBAS. Follow the ACTRIS recommendations and refer to them in the paper. After all, this is a "measurement report" so you should properly refer to the corrections you have done.

Good point. The Cref value was displayed on recommendation by a previous reviewer. We did not do any post-correction. We have however cited the wrong paper, as pointed out. We removed this sentence to not confuse readers.

L200. Caption of Figure 2. The word "trend" is wrong word here. Diurnal cycle is the right term. Trend is either increasing or decreasing in a longterm data. The same applies to the caption of Fig. S4, correct both.

Thank you, corrected both.

L214. "The AAE is similar between the sites, with small differences likely owing to different BC sources." Actually, the AAE values are different in a logical way. The AAE difference may be due to the BC size distribution and coating. At the rural site GMD is larger than at the urban site which may well lead to a lower AAE, in line with the simulations by Virkkula, Atmos. Meas. Tech., 14, 3707–3719, 2021.

We agree and modified this discussion accordingly:

L221. The AAE is slightly higher at the urban site, which could be due to the differences in BC size distribution and coating (Virkkula, 2021).

L215-219. Table 2. Add also the corresponding values of rBC from the SP-AMS. Discuss the differences between the rBC(SP2) and rBC(SP-AMS) also in the text, now there is no text about the rBC(SP-AMS). There are not many papers that would show both rBC(SP2) and rBC(SP-AMS), if any, I don't know (for you to find out...), so such a comparison would be very valuable.

In our experience rBC as measured by SP-AMS using the default calibrant (at the time of this study it was Cabot inc "Regal Black", size selected at 300 nm with a DMA) is biased low. This has been shown (willis et al 2014) to be due to incomplete overlap between particle beam and laser beam used to vaporize the particles (only vaporised components are detected in SP-AMS), referred to as low particle "collection efficiency" (CE)

The problem is that the calibrant particles have higher CE than ambient soot particles, and thus the instrument sensitivity obtained from calibration is not valid for ambient soot. The particle beam width, and thus CE, is dependent on particle shape which varies considerably for soot depending on e.g. coatings (aged soot particles are round or round-ish, fresh soot is highly aspherical). The SP-AMS normally measures "particle ensemble" properties, as opposed to single particle properties. In the urban air there is a mixture of old (spherical) and fresh (aspherical) soot particles which complicates rBC quantification by SP-AMS, as single particle mixing state comes into play. For that reason we opted to focus on rBC as measured by SP2 (intensive campaigns), and eBC as measured by AE33 (environmental monitoring) in this work.

We do agree that the issue is not properly tackled by the SP-AMS community yet, but it is outside the scope of this manuscript. It would make an interesting companion paper though (single particle data om coating thickness IS available from the SP2, and morphology state can be inferred to some extent from the size resolved SP-AMS data as well).

For the rBC modes in the table, give also their widths as GSD. And again, for both rBC(SP2) and rBC(SP-AMS).

See answer above on rBC (SP-AMS). We added GSD to the table:

	Rural	Urban
M_{rBC} (1h), µg m^{-3}	0.06 ±0.05	0.15 ±0.11
M_{rBC} (1h), µg m^{-3}	0.22 ±0.16	0.49 ±0.45
N_{rBC} (1h), cm^{-3}	31 ±21	100 ±80
GMD_M (24h), nm	168 ±12	141 ±11
GSD_M (24h), nm	1.74±0.08	1.77±0.04
$AAE_{370\text{-}950\ nm}$ (1h)	1.13 ±0.12	1.24 ±0.26

L275-276. "... mean mass of 0.062 ± 0.001 fg and 0.26 ± 0.01 fg, respectively, corresponding to effective densities of 0.95 ± 0.01 g cm-3 and 1.19 ± 0.02 g cm-3." How was this calculated? Give at least a reference, rather also the equation.

This is described in section 2.2.6 with references.

REVIEWER #3

The submitted manuscript focuses on the measurements of particulate matter in two locations, a rural and an urban location, in Sweden, during 2018. The authors presented measurements of rBC, eBC and investigated the chemical composition of non-refractory PM1. Furthermore, the authors conducted trajectory analysis to determine the influence of transport air pollution to the rural site.

The presented measurement report is within the scope of the ACP journal, and can be consider for a publication. The authors have addressed the comments of the previous reviewers, but there are still some comments than need to be addressed.

Comments

• In Ln 320 and Figure S1, the authors presented source apportionment analysis, but they did not discuss in the methods section the methods/tools used. In Figure S1, what do the Other Sources represent?

Yes, it is described in the methods section, see below. Other sources are e.g. waste incineration, non-road-transport and fugitive emissions.

L200. The long-distance transported BC source contributions during the rural and urban measurement campaigns were estimated using the Lagrangian 1D-column chemistry transport model ADCHEM (Roldin et al., 2011; Roldin et al., 2019). ADCHEM was setup and run forward in time along pre-computed 14-days backward HYSPLIT air mass trajectories arriving 100 m a.g.l. at Hyltemossa, one new trajectory every hour. Source specific anthropogenic BC emissions along the trajectories were taken from the CAMS-GLOB-ANT v4.2 global emission inventory, which has a spatial resolution of 0.1°x0.1° (Granier et al., 2019). BC emissions from wildfires were estimated using the GFED4 emission inventory (Randerson et al., 2018).

• In Ln 156, the authors used a dryer while sampling with the particle sizers. Did they account for any particle losses in their analysis?

The particle losses are described in section 2.2.7. The dryer before the SP2 was accounted for by adding a 1.25 m equivalent pipe length (Wiedensohler et al., 2012). Either way, the losses in this section is not likely to have affected neither N or M concentrations in the range of the SP2.

Wiedensohler, A., et al.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmospheric Measurement Techniques, 5, 657-685, 2012.

• In Ln 241, a reference is missing regarding the measurement artifacts of the aethalometer due to the coating of BC core with non-absorbing material. Furthermore, one more reason for higher measurements is the coating of the BC core with absorbing organic aerosol, brown carbon. High relative humidity can also affect optical measurements.

We added a reference comparing the AE33 specifically for this:

Kalbermatter, D. M., G. Močnik, L. Drinovec, B. Visser, J. Röhrbein, M. Oscity, E. Weingartner, A. P. Hyvärinen and K. Vasilatou: Comparing black-carbon- and aerosol-absorption-measuring instruments – a new system using lab-generated soot coated with controlled amounts of secondary organic matter, Atmos. Meas. Tech., 15, 561-572, 2022.

• In Ln 214, the AAE is not similar between the two sites. The smaller core particles in the urban have higher AAE. Previous studies have discussed how the sizes of the core and the coating can affect the AAE.

We have rephrased this, see answer to reviewer #2

• In Ln. 205, the authors compare the measurements of the number concentration of the rBC although the SP2 number concentration measurements are not accurate, due to the lower size cut off. The authors should consider estimate the number concentration of the rBC by applying lognormal fit in the number size distributions.

We compare the two sites using the same instrument and same cut-off size which we think is fair. The size distribution of BC mass equivalent cores is generally very broad in ambient air, consisting of several different emission sources. Since the diameter where the SP2 detection efficiency decreases is often close to where the number size distribution of BC is at its maximum, the actual total number concentration is very uncertain. We have added the geometric standard deviation to table 1 so the size distribution can be computed by interested readers.

• The authors presented a comparison of the aethalometer and SP2 measurements. The authors should include in the manuscript the measurement uncertainty of the two methods.

We have added the estimated relative uncertainty for the SP2

L99. *The overall relative uncertainty in mass concentration by the SP2 was estimated by Sharma et al. (2017) to be within 25-38 %.*

The estimation of the AE33 is not so easy to make. We have added a reference looking at AE51 instruments and the following text.

L132. *The relative uncertainty of the absorption coefficient measured by the aethalometer increases with lower BC mass concentrations, and has been shown to converge around 30 % for an older version of the instrument (Backman et al., 2017). The uncertainty of eBC also includes the uncertainty in the MAC value which was not estimated in the present study.*

• In Ln.154 Please add references on the DMPS and the Hauke type DMAs.

We have added the following reference:

Wiedensohler, et al: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmos. Meas. Tech., 5, 657-685, 2012.

Minor Comments

• Figure 7: define CPH in the caption

Fixed.