Point-to-point answers to reviewers

The authors are grateful to the editor and reviewers who brought up important points and have spend their time in order to improve the manuscript. We are sorry that the review process had to be extended several times and we are grateful for the patience with this manuscript.

Below are point-to-point answers to each of the comments. The comments by the reviewers are marked bold, and the text copied from the revised manuscript is presented with cursive.

In addition updating the manuscript according to the comments, we updated Figs. 12 and 13, where we observed an error with reporting the value range of the temperature change variable. The change did not affect the shape of the curve. Therefore, fixing this error did not change the conclusions. Also, to improve clarity, we removed the "Both intercomparisons" -column from Table 4, as the values were not referred in the text or used in the data processing.

Reviewer 1

Review of ar-2024-12: "The applicability and challenges of black carbon sensors in dense monitoring networks"

The paper elaborates on the applicability of BC sensors in dense monitoring networks. The figures are clear and easy to understand and the topic is of high interest especially in the frame of the upcoming EU regulations. I still have some minor comments listed below.

General comments

To me, the word "dense" is a little misleading since the focus of the work is on BC small-scale sensors. I would remove the word dense. Because: What are the specific challenges with this BC sensors in the frame of a dense network over a stand-alone device included in a nation-wide network? Why is this emphasized?

We agree, removing word "dense" simplifies the title.

Regarding the "Results and Discussion" section: For the intercomparison, to check whether or not the offset in the orthogonal fit is real, please elaborate whether or not a sufficient zero test was conducted within the intercomparison period. Also, please provide statistically backed estimates on the validity of the coefficients of the orthogonal regression. If yes and successful, why the calibration curve was not forced through zero?

Unfortunately, no comprehensive zero tests were conducted during the intercomparison period or during the field deployment. Therefore, the intercept is included in the orthogonal regression. Standard errors of the orthogonal fit parameters were added in Table 4.

We added discussion of the studies that observed similar results of intercomparisons. To see these changes, please see the answer the Reviewer 2 comment starting with "The added paragraph discussing…" that is related to the same topic.

Also, the text needs a revision in terms of interpunctuation and harmonization of how units are given (with or without space).

We have modified the manuscript and paid focus on uniform notation for variables and units.

And last but not least, the paper would highly benefit from a more statistical analysis on the significance of the results (see below).

We understand this concern and we improved the statistical analysis, which has been added to Appendix A2 from the deployment phase. For the intercomparison periods error limits for fitting parameters have been added in Table 4, to show the accuracy of the fitting parameters.

References to statistical significance and to Appendix A2 have been added in the manuscript:

In Sect. 3.2.1: … *All locations had a statistically significant difference (Fig. A2.1), although the differences were not necessarily remarkable.*

In Sect. 3.2.2: … *At all the locations, Mon and Tue had statistically significantly (Fig. A2.2) higher concentrations than Wed and Thu. The most stable locations were F_{ground} and $SMEARIII_{ground}$, where there was no statistically significant difference between Mon, Tue, Fri, Sat and Sun. ….*

Please also set your work in context to other work like (Wu et al, 2023, https://doi.org/10.1016/j.jes.2023.05.044. who also checked the performance of protable BC monitors in field measurements

Text that addresses other similar studies were added in various sections of the manuscript. Discussion referring to other studies was added in Sects. 3.1. and 3.2.1. To see these changes, please see the answer the Reviewer 2 comment starting with "The added paragraph discussing…".

Also, following text was added to Sect. 1 "Introduction":

Previous studies have reported a good correlation between BC sensors and reference-grade instruments, but with varying slopes and intercepts depending on location and sensor implicating the need for onsite calibration (Alas et al., 2020; Kuula et al., 2020; Chakraborty et al., 2023; Wu et al., 2024). In previous studies, a common application for these sensors has been personal BC exposure as a carry-on measurement device (Delgado-Saborit, 2012; Li et al., 2015), mobile measurements (Alas et al., 2019; Pikridas et al., 2019), and sensor networks (Caubel et al., 2019).

<u>Specific comments</u>

p. 2 l. 50: space between number and unit. Please also harmonize along the text.

We paid focus on uniform notation for variables and units throughout the manuscript.

p. 2 l. 56: small-scale versions of? Word missing

We added the missing word: *…small-scale versions of the filter-based instruments…*

p. 2 l. 58: In previous studies – add a comma there (check missing comma)

Added a comma.

p. 3 l. 70: Please elaborate on the assumed performance of the MAAP. How you know, that this device can act as a reference?

MAAP is deployed at SMEAR III for long-term measurements. MAAP widely used in eBC monitoring and it is accepted instrument e.g., in the ACTRIS measurement infrastructure and it has been used as a reference instrument in previous studies (Alas et al., 2019; Luoma et al., 2021a). With the angular scattering measurements and applying the radiative transfer scheme in eBC retrieval, MAAP is less sensitive to aerosol particle scattering and is more independent method compared to, e.g., AE33.

The text was modified accordingly in Sect 2.3:

The reference instrument MAAP is also a filter-based absorption photometer, but it differs from the measurement principle presented in Sect. 2.1 by additionally measuring backscattering from the filter at two angles to improve the accuracy of the σ_{ap} *and eBC. Additionally, the MAAP derives the* σ_{ap} *by applying a two-stream-approximation radiative transfer scheme (Petzold and Schönlinner, 2004). Therefore, it is somewhat more independent measurement method and is a good reference instrument for the eBC sensors. MAAP has been used as a reference instrument also in previous studies comparing filter-based instruments (Alas et al., 2019; Luoma et al., 2021a). The reported uncertainty and unit-to-unit variability of MAAP (at 16.67 l* min^{-1} *flow) are 12% and 3% (Petzold and Schönlinner, 2004; Müller et al., 2011). …*

p. 3 l. 79: An Aethalometer® is a device (and please use the registered trademark sign), not a method. Please rephrase.

Rephrased to: *Filter-based optical methods are widely used to measure BC concentration due to ease of operation and relatively low cost (Hansen et al., 1984).*

References to Aethalometer have been replaced with more generic language where appropriate. Trademark has been added when first referring to Aethalometer AE33 in Sect 2.1.

p. 3 l. 84: “The measured variable by the instrument is the attenuation coefficient bATN(λ) [m^{-1}] calculated from the measured attenuation” – if it’s calculated it’s not measured.

This was fixed and rephased to: *The attenuation coefficient* $b_{ATN}(\lambda)$ *[*m^{-1}*] is calculated from the measured light intensity and the operational parameters of the instrument as described in Eq. 2, …*

p. 6 l. 155: According to Müller et al. (2011), the MAAP LED differs from the manufactures given wavelength and is 637 nm.

This was corrected and the wavelength was changed to 637 nm and added a citation to Müller et al. (2011) in the text: *The instrument measures with only one wavelength at 637 nm (Müller et al., 2011) and applies MAC = 6.6* m^2 g^{-1} *(at 637 nm).*

p. 7 Table 1: The given detection limit is given for a defined sampling period. Please elaborate. E.g., MA200 is "30 ng BC/m 3, 5 min timebase., 150 ml/min flow rate, SingleSpot™" according to the technical specifications.

We have updated the Table 1 according to the technical specifications.

p. 12 l. 228: Please provide details on how the intercomparison was set up, e.g., whether or not the sensors sampled through the same inlet etc.

This is described now in more detail in Sect. 2.4.: *The small-scale sensors were all measuring on the same sample line (different with MAAP) that did not have any inlet pre-impactor. The separate measurement line was set up through the SMEAR III station wall at a height of 3 m from the ground.*

p. 22. L 368 and 369: "At all the locations, Mon and Tue had higher concentrations than Wed and Thu, and at most sites the highest concentrations were observed on Fri." Are these differences statistically significant – please add and/or comment within the text. This also expands to the later following analysis regarding the differences along sensors and weekdays.

Added appendix A2 where statistical significance is given with the Wilcoxon signed-rank test (using scipy.stats.wilcoxon) between the locations and between the weekdays for every location.

Added and modified text in Sect. 3.2.2.

A daily breakdown can be seen in Fig. 10. There is somewhat surprising variation on day-to-day basis, as no notable differences were expected between weekdays. At all the locations, Mon and Tue had statistically significantly (Fig. A2.2) higher concentrations than Wed and Thu. The most stable locations were F_{ground} *and* $SMEARIII_{ground}$*, where there was no statistically significant difference between Mon, Tue, Fri, Sat and Sun. Therefore, weekend and weekdays did not seem to have a clear difference in the medians to each other, which is differing compared to other studies that observed lower eBC concentrations during weekends at traffic and at urban background sites in Helsinki (Helin et al., 2018; Luoma et al., 2021b). Also,* *Caubel et al. (2019) reported lower concentrations during weekends especially on traffic influenced cites. It is to be noted that the variance in concentrations was higher during Fri–Sun than during Mon–Thu and the highest peaks were measured during the weekend. The unexpected similarity between the weekdays and weekends might be due to a rather short period (74 d) for such an analysis and the time of the deployment period, which is a vacation season in Finland, when the anthropogenic activities are expected to depend less on the days of the week.*

Also added text in Sect. 3.2.1:

All locations had a statistically significant difference (Fig. A2.1), although the differences were not necessarily remarkable.

p. 23 l. 387: I assume that the planetary boundary layer is thicker this time and the dilution of the emissions is enhanced. Please check.

This is now mentioned more clearly in the text in Sect. 3.2.3 where we modified the paragraph: *The diurnal variation of eBC can be seen in Fig. 11, which shows a similar diurnal pattern at all the locations. The variation is affected by the local and regional anthropogenic activities and meteorological conditions. The eBC concentrations sharply rose during the morning due to increase in traffic. The highest concentrations were reached between 9–10 after which the concentrations decreased due to smaller traffic rates, increased dilution in the convective boundary layer due to higher mixing height and increased wind speeds (e.g., Fig. S2 in Luoma et al., 2021b). Another rise in concentration was observed late in the evening around 21–23. This increase was much less compared to the morning peak. The increased levels during the evenings are probably caused by accumulation of pollutants in a more stable atmosphere when the mixing height is lower and the wind speeds are also generally lower. Based on observations made in Helsinki, (Järvi et al., 2009) reported that out of the meteorological parameters, the wind speed and mixing height had the greatest effect on eBC concentrations. Also, local wood combustion emissions, for example, evening activities at the close by community garden, can increase the eBC levels. Similar diurnal patterns with a peak in the morning and evening have been observed by previous studies during the warm period at traffic and urban background sites (Sahu et al., 2011; Backman et al., 2012; Caubel et al., 2019; Luoma et al., 2021b).*

p. 23 and p 24. Fig 12 and 13: The shown temperature clearly exceeds the operational temperature range given by the manufacturer. The question is whether or not the data is reliable. Also, the 880 nm channel is near IR and hence sensitive to temperature changes. Please provide insights, if the bias was also visible in the other channels of the MA200. To me, a MAAP at 637 nm wavelength indicates that the 625 nm channel of the MA200/MA350 would also be suitable to determine the eBC mass concentration. Since the MAs provide multiple wavelengths, and the question of applicability of those sensors in a dense network is stated, also other channels should be considered. The effect of BrC should be negligible still. Also, since the change rates of environment are of important I'd like to see also the change of the sample RH within the figure. In a supplementary material I'd like to see the time series, showing that Observair sensors is significantly less affected by environmental changes.

Anomalous activity is observed while below 40 °C between 6.00-8.00 (Fig. R2), which is within the operational temperature and should be considered reliable. We added a mention of this to Sect 3.2.4: *According to the instrument manual, the operating temperature for the instrument is 0-40 °C. In our deployment, the T increases even to 52 °C, which is above the operating temperature and could explain the behaviour. However, the anomalous activity is observed even below the 40 °C.*

We studied the observed behaviour also on other wavelengths, which are presented below in Fig. R1. The bias is visible on all wavelengths, but it is less pronounced at lower wavelengths. Other wavelengths were not used in the analysis to keep the scope of the study on *eBC* and to be comparable to the other sensor types. We added a mention of this to Sect 2.3:

… The other wavelengths of the MA-sensors can be used to differentiate between BrC and BC and the possible sources of these particles. In this study only the 880 nm wavelength was utilized to conform to the other sensors. …

To see also the variation in *RH*, we have now added the *RH* change rate to Figs. 12 and 13.

In Fig. R2 the Observairs seem more stable than the individual spots of the MA350 between 6.00–10.00 although the *T* gradient is less. The main problem arises from that the instability of the individual spots is amplified by the dualspot correction. This is relevant if the dualspot corrected data is considered the default output of the instrument. The spiking in Observair sensors around 11 is due to handling of the instruments (just inspecting that the measurement is running via opening the deployment box). With the handling it could be argued that the Observair sensors too are susceptible to sharp changes in *T*. This enforces the requirement of robust deployment with *T* and *RH* controlled deployment boxes and that the algorithmic solutions are suitable for lower temperature gradients but do also have a limit (which is not determined in this study).

The conclusions are updated to emphasize that the user needs to take into account the changing environmental parameters more carefully. See the answer the Reviewer 2 comment starting with "The added paragraph discussing…".

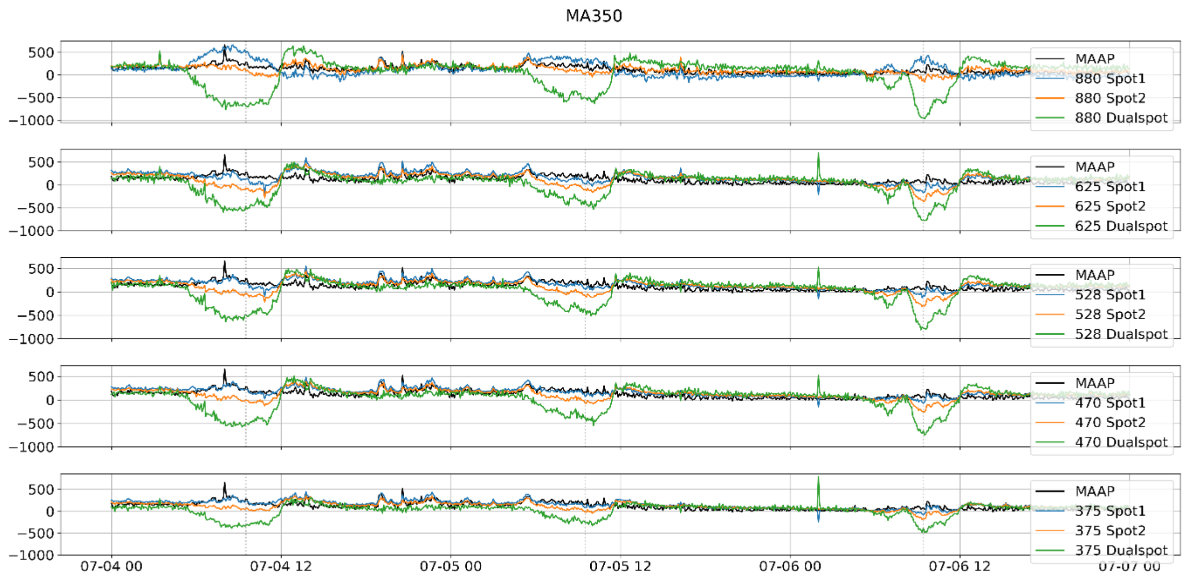

Figure R1. MA350 all wavelengths.

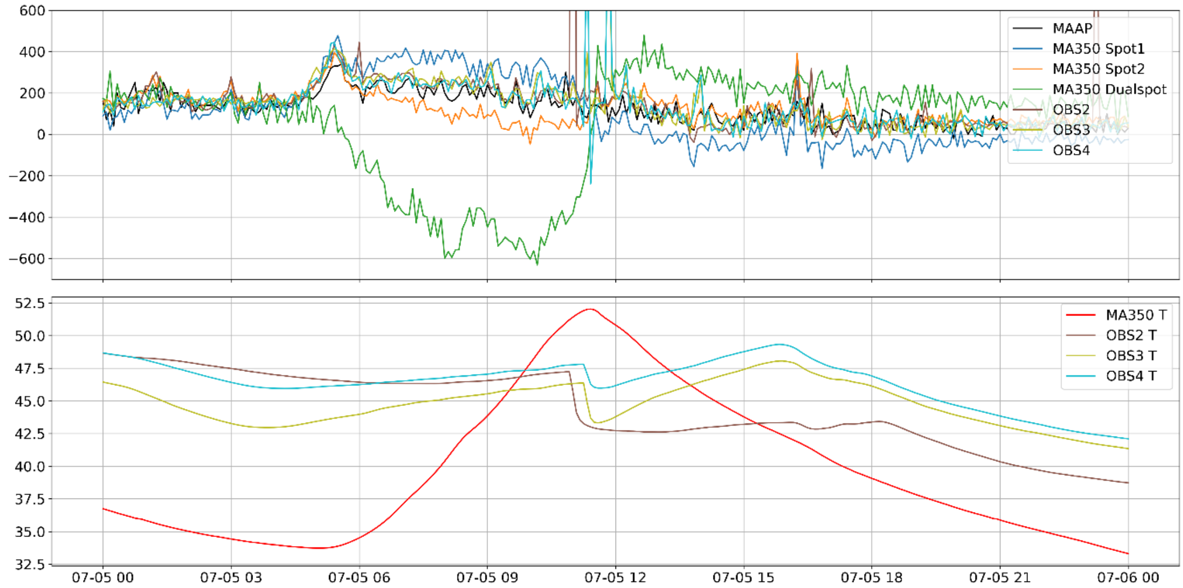

Figure R2. MA350 and OBS timeseries.

p. 27 Table A1. I am missing the respective information on the MAAP at SMEAR III ground station.

MAAP flow has been observed stable during long-term measurements at SMEARIII, and unfortunately it was not checked during the campaign. The operating flow is listed in Table 1.

Reviewer 2

The comparison of the performance of low-cost black carbon (BC) sensors, the correction algorithms and their impact on the measurements, as well as the sensitivity of these sensors to changes in relative humidity and temperature presented in this manuscript is valuable to the scientific community. Thus, I find this article suitable for publication, once the remaining points that need further attention are addressed.

General comments

Although the language in the manuscript has improved, there are still instances where the writing lacks clarity. I suggest another round of language revision. For example, sentences like "Unfortunately, the Observair sensors are not being produced as of the end of 2023…", "The assumptions are that with 880 nm light source the absorption" or "However, still in the field study, several issues were observed..." need to be revised for better readability and precision.

We have revised the marked sentences and read the paper carefully through aiming to improve unclear sentences and to unify notation.

In Figure 2, the dates on the x-axis have not been corrected as suggested by Referee #1. There is still a shift in the dates between late May and early June. I recommend using consistent labeling, as with the other dates (every 2 days). Additionally, there is a language error: "Timeseries" should be corrected to "Time series."

Corrected ticks to two-day frequency.

In Figures 12 and 13, there are errors in their descriptions. The units should be placed outside the parentheses, or the word "in" should be included within the parentheses.

The units in the figure captions were removed as they were unnecessary. Also, we found an error in the figures, so they were updated, but this did not change the conclusions.

The subsections on methodology and results were not fully renamed to reflect the context, as suggested by both referees. Below are some examples for improved subsection titles:

- **2.1. Measuring principle to obtain BC mass concentration with low-cost sensors**
- **2.3. Deployment of small BC sensors at the Kumpula Campus**
- **2.4. Description of the sampling site**
- **3.1. Intercomparison of BC sensors**
- **3.1.2. Adjusting differences between sensors for comparison**
- **3.2. Temporal and spatial variability during deployment**
- **3.2.2. BC levels during days of the week**
- **3.2.3. Diurnal variation in BC concentration**
- **3.2.4. Artifacts caused by sensor overheating**

The titles were improved according to the recommendations with small modifications. Also, the title of Sect. 3.2.1 was changed to *General features and spatial variability of eBC in Kumpula*.

The authors did not provide the strong justification in line 71 as suggested by Referee #2 on the use of the MAAP as the reference instrument.

This has now been improved (see the answer to Reviewer 1 comment regarding “p. 3 l. 70”).

While presenting the calibrations results (F and orthogonal fit), a comparison with previous literature, as suggested for Referee #2 is missing. The authors report the results of the orthogonal fit (slope, correlation coefficients and intercept) in Table 4 but they do not include metrics from literature data with the type of calibration procedure adopted.

Parameters from previous studies and discussion have been added to Sect. 3.1 (See comment starting with "The added paragraph discussing…").

We found no previous studies that applied the fitting equations to correct data for black carbon sensors, but this approach has been used in sensor networks before (Petäjä et al., 2021). During the analysis phase this step was noted to be important as it reduced the differences between the locations.

The added paragraph discussing the results with literature on temporal and spatial variability has enhanced the revised version of the article, as suggested by the referee #2. However, the discussion could be further enriched by comparing these results in greater detail with findings from other studies conducted in urban areas across Europe using the same sensors.

The discussion has been improved in various sections by comparing the results to other similar studies.

Added to Sect. 3.1:

Alas et al. (2020) has reported similar results with AE51 compared to MAAP with reduced major axis (RMA) regression. In Manila during summer of 2015 AE51 had r of 0.845 with a slope of 0.871 ± 0.013 and in Rome during winter of 2017 better r of 0.983 and slope of 1.015 ± 0.003. In Loški Potok, with AE33 as the reference, the reported intercomparison values for rural background were r = 0.962 with slope = 0.876 ± 0.005 and for rural village r = 0.978 with slope = 0.826 ± 0.002. Varying slopes are most likely caused by different aerosol types that depend on the location and season.

…

Kuula et al. (2020) reported intercept, slope and r of -44, 0.85 and 0.97 of MA350 when compared to AE33. Chakraborty et al. (2023) reported r ~ 0.90 and slopes ranging from 0.736-1.01 for three MA300 units compared to AE33. As the AE33 measures slightly higher concentrations than MAAP (Pikridas et al., 2019; Wu et al., 2024) the intercomparison results seem to be inline with previous studies.

…

Previous studies have reported similar r and slightly lower slopes with AE33 as reference. r = 0.904 with slope = 0.57 (Wu et al., 2024) and r = 0.89 with slope = 0.87 (Caubel et al., 2018). Wu et al. (2024) noted that the low slope could be partially explained due to high loading of the filters. In this study filters were changed regularly and the average concentrations were much lower that in Wu et al. (2024) (mean 230 ng m^{-3}, 1465 ng m^{-3})….

Added to Sect. 3.2.1:

The overall concentrations were lower than previous studies conducted in the Helsinki region. Luoma et al. (2021b) reported annual means of 510 to 530 ng m^{-3} at urban background cites. Helin et al. (2018) reported average±standard deviation concentrations of 1940 ± 1530 ng m^{-3}

at a street canyon and 450 ± 420 ng m^{-3} at a detached residential area in the summertime of 2016. The concentrations were similar with corresponding values of urban background cites during summers of 2017-2019 within Northern Europe (~240 – 340 ng m^{-3}) and lower than in Western and Central Europe (330 – 1480 ng m^{-3}) (Table S2 in Savadkoohi et al., 2023).

In comparison to Caubel et al., (2019), who operated 100 eBC sensors for 100 days in a borough sized area, we observed only small variations within the much smaller campus area. Caubet et al., (2019) reported considerably larger differences between the sensor locations: for example, 200-400 ng m^{-3} in upwind locations that were less affected by the anthropogenic activities, and 500-1200 ng m^{-3} in a busy port environment. Residental concentrations were reported to be on average slightly higher 400-500 ng m^{-3} in comparison to the 250-400 ng m^{-3} measured in this study.

Added to Sect. 3.2.2:

Also, Caubel et al. (2019) reported lower concentrations during Sat and Sun especially on traffic influenced cites.

The following text was added in the very end of Sect. 3.2.1. related to the effect of the construction site:

Similar results have also been reported by Alas et al. (2019) who did not observe increased eBC concentrations close to construction sites.

The discussion on spatial variability has improved; however, there are still questions raised by referee #2 that have not been yet fully addressed, which would be of interest to the reader. For instance: "How is spatial variability captured by different devices?" or "Are there differences in performance, and how can spatial variability be effectively captured by these devices?" Additionally, some recommendations or comments are missing, such as identifying which devices performed best in specific contexts.

To address how the sensors were able to measure the variability, we added the following summary of the results in the conclusions and addressed the differences between the instrument designs:

During the intercomparison periods, the correlations between different eBC sensors and the reference instrument were good ($R \approx 0.8$, 5-min averages) but the slopes of the regression lines varied from 0.8 to 1.1 indicating a need for sensor-specific calibration. The eBC sensors observed the temporal variation well and the eBC levels varied according to anthropogenic activities in the local and regional area (e.g., in the nearby busy road), and meteorological conditions. For the spatial variation we observed only small variation. Surprisingly, the local construction site, which was assumed to cause an increase in eBC data, did not stand out in the results. Due to the lack of local emission sources in the studied area, the variation in eBC in an urban background location was observed to be minimal. Based on our results, the reference scale SMEAR III station, which is classified as an urban background site, represents the pollution levels in the campus area well. Taking the sensor network closer to local *anthropogenic sources (e.g., right next to a busy road), the gradients of eBC concentration are expected to be more remarkable.*

…

It is not possible to say which sensor performed the best as the sensor design differs significantly. The user needs to take into account the requirements of the measurement environment and the features of the individual sensor types (number of wavelengths, filter capacity and other maintenance needs, and price). In our conditions sensors performed near equally if single spot data is used for the MA-sensors. Observair performed slightly more stable in changing conditions. Comparing the instrument performance, it is to be noted that AE51 was run in temperature controlled environment while Observair and MA-series sensors were deployed in the deployment boxes. With AE51 and Observair the filter change needs to be done every few days whereas MA-sensors can measure independently for months. MA-sensors also offers the wavelengths for BrC, which weren't utilized in this study.

The authors have revised the conclusions in the manuscript; however, they need to update them by incorporating a few lines addressing the points raised here (previously asked by the referees). This would enhance the manuscript by providing valuable information and recommendations for the community. For example, identifying which device performed best for monitoring the spatial and temporal variability of BC mass concentrations under the specific ambient conditions of the site (e.g., high relative humidity) would be particularly useful. Additionally, positioning the sampling site as having high, medium, or low BC levels in comparison to other urban sites using small BC sensors would further enrich the conclusions. This revision would avoid discussing aspects not explored in the manuscript, as noted by Referee #2, such as BC source apportionment, which refers to determining BC sources through spectral dependency or positive matrix factorization in the scientific community.

We removed the section referring to BC source apportionment to keep the focus of the manuscript on BC monitoring and comparison of different sensor types.

In our set-up, the spatial variability was not remarkable due to lack of local sources. In this campaign, the location of the sensors were chosen based on easy access and availability for electricity. We agree that having sensors in locations with more varying eBC levels would have given interesting results. However, it was also interesting to see how little the eBC varies within the background area and if the SMEAR III station actually represents the campus area well. We have just submitted an article about air pollution gradients next to a busy road (Harni et al., submitted in 2025), but the article is not yet accepted for discussion. Also, in regard to recommendations to which device performed the best, general advice is given to consider the application and resources available to use the instruments. The instrument designs differ significantly, and the selection needs to be done with the strengths and weaknesses of the instrument design in mind. We updated the conclusions to address this (for the modified text, please see the answer to the previous comment).

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