COMMENTS TO THE AUTHOR:

amt-2020-517

GENERAL COMMENTS

This manuscript provides an evaluation of three noise-reduction algorithms for the "raw" equivalent black carbon (eBC) mass concentration data of the new 5-wavelength microAethelometer model MA200. This has been submitted for the special issue: New developments in atmospheric limb measurements: instruments, methods, and science applications (AMT/ACP inter-journal SI).

Below are general comments on the manuscript as a whole:

Appropriateness:

1. To the scope of the special issue (SI): If this is not an error, it must be aptly justified why this is submitted in this SI. The SI call is specifically for "new developments in atmospheric limb measurements" focusing on the stratosphere. This manuscript has very little relevance to the scope of the SI and is an outlier among the other preprints included in this SI. I understand that the MA200 has been and may be used in vertical profiling. However, the dataset used here were from ground-based mobile measurements in an urban area. In my opinion, this manuscript does not belong in this SI.

Response: Thank you for bringing this to our more immediate attention. We agree and will take efforts to move the manuscript to the regular edition of AMT.

2. To the scope of AMT: On the other hand, the manuscript does fall within the scope of AMT, in general. However, it lacks the detailed discussion on the technical aspects that is common with AMT publications, particularly if the presented "decision tree" is something the authors would like others to employ. The entirety of the comments on this manuscript are based on this manuscript fitting the scope of AMT and not of the SI to which it was submitted to.

Response: Thank you for your comments. We will take efforts to move the manuscript to the regular edition of AMT. We have also added more content to the paper surrounding technical discussion and justification for the novelty and utility of the material to the Introductory Discussion sections.

Scientific Significance and Quality:

The idea behind this investigation is understandably important for some users of the MA200, particularly those who use the read out directly. However, the following issues were insufficiently addressed in the manuscript:

1. The motivation for noise-reduction, in general, was not sufficient. There is a part of the community who prefer the data as it is (since the instrumental noise cancels out when averaged), and focus instead on making sure the measurements are set-up correctly to prevent artificial peaks in the data (Cai et al., 2013; Alas et al., 2019).

Response: Thank you for your correction and for pointing out the lack of clarity regarding our interests in "removing" negative values. We understand that a part of the community prefer the data as it is. Our paper is intended to serve and inform members of the community interested in smoothing out noise to produce more highly temporally resolved, unbiased estimates of eBC concentrations. We strongly agree that noise within the data, including negative values, contain valid information and that arbitrary removal of negative values may be detrimental to a dataset.

We have updated the text to more make this more clear (e.g. the 5th-to-last paragraph in the Introduction). Specifically, our motivation is that all measurements have noise, and in the case of the Aethalometer® method and thus with the microAeth®, some data points will be greater than the actual value and some will be lower than the actual value. To understand data at a temporal precision of the data collection (e.g. a 1 hz time-base) with greater likelihood of matching the "actual" value, unbiased noise-reduction methods must be applied.

We also understand that, in fixed black carbon monitoring, concentrations are often more stable and less noisy across time, and, as a result, noise reduction is less critical. However, when we perform mobile monitoring, due to the high heterogeneity of black carbon concentrations, noise in the data can (often) more meaningfully affect the interpretation of temporal and, thus, spatial variations in black carbon concentrations at higher sample time bases (e.g., 10 s). In the absence of noise reduction or by simply averaging the monitoring data over discrete time periods, it is difficult to accurately observe spatial pollutant properties and scale. For mobile monitoring, therefore, data noise reduction is considered necessary for the MA200 (and many of other air quality monitors).

2. It was mentioned in the text that the MA200 has an "on-board signal-processing that reduces the noise" of the MA200 raw data. Note that upon going through the user manual and quickly searching the MA200 website, this internal post-processing of the raw data is not mentioned (please, correct me if I'm wrong). This is an extremely important point that users need to know prior to using this instrument. This would mean that the output of the instrument is not "raw" anymore and, in the context of this study, the data has been "smoothed" out twice with the treatment of the noise-reduction algorithms being evaluated.

Response: Thank you for your concern. The raw black carbon concentration data are the data directly outputted by the instrument, and our analysis is only postprocessed once. To avoid misunderstanding, we deleted this sentence.

3. The measurement strategy was not explained in detail. Particularly, what measures were done to take into consideration the sensitivities of the MA200?

Response: Our main measurement strategy is how monitoring devices, noise, and the application of different data postprocessing methods affect the measurement of black carbon in different microenvironments, under different instrumentation interval times, and, specifically, for mobile monitoring. More detailed information has been added to the revised manuscript. Please refer to line 171-176 for confirmation.

4. The results were merely presented. Deeper discussion on why the algorithms performed as they did is needed.

Response: Thank you for your suggestion. We have provided a deeper discussion on the algorithms in the revised manuscript. Please refer to the result and discussion section for confirmation.

5. The broader significance of this study and how it relates to existing literature were not discussed.

Response: Thank you for your suggestion. We have updated to text to more directly address the significance of this study and how it relates to existing literature in the "3.5 practical implication" section (line 453-478), and have added more justification for the work in the introduction.

Presentation Quality:

There is much to be improved with the writing of this manuscript. The main points are often times hidden in a mix of redundancy, jargon, lack of proper sentence/paragraph structures, and poor grammar. This provides so much hurdles for the readers in understanding the thought process of the author(s). Important aspects such as the criteria for evaluating the noise-reduction methods were often vague and leaves so much to interpretation or misinterpretation of the reader. Only my personal experience with the microAethalometer and mobile measurements allowed me to extract the information the author wants to give, and even then, it was with such difficulty.

The figures and tables, as well as their corresponding captions were not informative enough for the reader to understand them even after reading the manuscript (much less without). The figures and tables must be intuitive. Use of informative legends would improve the figures significantly. In addition, the parameters used to evaluate these noise-reduction algorithms were not defined prior to presentation of results. For instance, "noise reduction effect" and "negative decline rate" were not defined prior to showing up in Table 2 and how they were calculated was also missing.

Response: Thank you for your suggestion. We have revised the manuscript, including text, some figures, and tables together with the captions following your comments and suggestions to improve the readability of our manuscript.

Some technical comments:

1. Define terms before abbreviating/ using initials (BC).

Response: Thank you for your correction. In order to improve the clarity of black carbon, we choose to not abbreviate this term in our revised manuscript. We have carefully checked the revised manuscript for other abbreviations and acronyms

2. Please consider using eBC (Petzold et al., 2013) consistently. It was anyway introduced in the text.

Response: Thank you for your correction. We have carefully checked and revised through our manuscript regarding your concern, including the following text in the paragraph: "In our study, the equivalent black carbon (eBC), the preferred term for describing black carbon assessed with mass absorption cross-section (MAC) facilitated optical absorption methods (Petzold et al 2013), was used when addressing quantitative values." Please refer to line 77-79 for further confirmation.

3. Use initialisms throughout the manuscript.

Response: Thank you for your correction. We have carefully checked and revised through our manuscript regarding your concern (e.g., CMA, ONA, LPR). We have highlighted all related initialisms term on blue color. Please refer to our revised manuscript for further confirmation.

4. Be consistent with terminologies used.

Response: Thank you for your correction. We have carefully checked and revised our manuscript regarding your concern. Please refer to our revised manuscript for further confirmation.

5. Please use “MA200” throughout the whole manuscript instead of switching from MA200 to “sample monitor” or “sampling equipment” every now and then.

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to our revised manuscript for further confirmation.

6. Use complete sentences in figure captions.

Response: Thank you for your correction. We have modified and used complete sentences in all figure captions including supplementary file in our revised manuscript. Please refer to our revised manuscript for further confirmation.

7. The texts (titles, labels, legends) in the figures are not all the same size.

Response: Thank you for your correction. We have checked and revised all the texts (titles, labels, legends) in the figures with the same size. Please refer to our revised manuscript for further confirmation.

8. I strongly suggest major revisions in the writing with a native English speaker contributing on and reviewing the manuscript prior to re-submission.

Response: We gave our revised manuscript to a native English speaker to improve the writing quality and readability.

SPECIFIC COMMENTS

MAIN MANUSCRIPT

ABSTRACT

1. line 32 “Our results showed CMA to be a good prospect…”

Response: Thank you for your correction. We have modified it combined with your comment. Please refer to line 32-33 for further confirmation.

2. line 33 This line is a little unclear. The readability may be improved. Here’s a suggestion for this sentence: “Based on the interval times used here, our results showed CMA to be a suitable algorithm to reduce the noise of raw BC mass concentration data based on the decrease of negative values and the retention of details attributable to microenvironmental changes.”

Response: Thank you for your correction. We have modified it combined with your comment. Please refer to line 32-35 for further confirmation.

3. line 34 Did you mean here “highest reduction OF peak values”?

Response: Yes it is. Please refer to line 35 for further confirmation.

4. line 35-36 “Furthermore, after background correction, the CMA results retained more detailed microenvironmental changes in pollutants than other methods.”

Response: Thank you for your correction. We have modified it combined with your comment. Please refer to line 35-37 for further confirmation.

5. line 38-39 “These findings provide new insights on the suitable noise reduction approach for

mobile monitoring data obtained from portable BC instruments."

Response: Thank you for your correction. We have modified it following your comment. Please refer to line 37-40 for further confirmation.

INTRODUCTION

General Comments

1. The jump in topics from line 48 to 49 is a bit big. I suggest to introduce at first the relevance of BC particles in air quality through its health effects. Then you continue with line 49. This will improve the motivation of your study.

Line 43 "Black carbon particles with size ranging from …"

Response: Thank you for your correction. We have modified it following your comment. Please refer to line 43-50 for further confirmation.

2. Line 52: is it really the goal to propose a monitoring method or a method to analyze data from mobile monitoring? It would not hurt to already introduce here the ONA method by Hagler et al. and to motivate why it is necessary to explore other means of reducing noise from the eBC datasets from portable absorption photometers.

There is insufficient motivation on the purpose/advantages of noise reduction?.

Explicitly mention that the noise-reduction algorithms evaluated here are those that are readily available in the Aethlabs Dashboard. There are members of the community who log AE51 or MA200 data independently and do not use the Dashboard. In any case, it must be justified, why the LPR and CMA are options in the Dashboard as noise-reduction algorithms.

Response: Thank you for your correction. We have briefly described and confirmed regarding with the goal of our study "to propose a monitoring method and motivation of noise reduction". Following that, we briefly introduce the ONA method by Hagler et al. to explore other means of reducing noise from the eBC datasets from portable absorption photometers. And justification of the LPR and CMA are options in the Dashboard as noise-reduction algorithms was also mentioned. Taken together, please refer to line 54-66 for further confirmation.

3. Line 67-68 "…and simply removing negative values may introduce biases in the dataset." In this paragraph, it would be beneficial to inform the reader that these negative values are part of the instrumental noise, before you introduce noise reduction.

Response: Thank you for your suggestion. We have added related information that negative values are part of the instrumental noise before introduce noise reduction. Please refer to line 81-83 for further confirmation.

4. Line 70-72 "Moreover, high-time resolution measurements of air quality at roadside are susceptible to single events (e.g. occasional passing of heavy-duty diesel vehicles or cigarette smoke) that may not be representative of the street in study. This may result in over estimation of eBC levels when averaged over time/space as they introduce peaks in the dataset."

Line 72-74 "In addition, when the sampling equipment traverses from highly-polluted area to a low-polluted one, such as a park, the instrument produces strong negative peaks that is due to the measurement principle of the instrument and the strength of the pollution gradient between

microenvironments."

Line 75 "Therefore, the noise reduction method should also be evaluated based on the retention of actual peak concentrations and number of peak samples that are related to identifiable sources of pollution."

Line 78 Remove "including"

Response: Thank you for your correction. We have modified it combined with your comment. Please refer to line 86-95 for further confirmation.

5. Line 84 The rationale of doing background correction in relation to the performance of noise reduction methods is not clear to me. Could you further elaborate on the statement that background concentration could affect the noise of the instrument?

Response: The background concentration does not affect the instrument noise reduction, but rather the instrument noise reduction affects the true value of the background concentration. Therefore, noise reduction, which better reflects the background concentration, is one of the criteria for assessing noise reduction in this study.

6. Line 87 This study evaluated three methods for reducing the noise from the raw BC dataset obtained using the MA200 in mobile measurements along a trafficked street in an urban area. The methods investigated are ONA (please cite here Hagler et al.,), LPR, and CMA.

Response: Thank you for your correction. We have cited the related reference (Hagler et al., 2011) regarding ONA in the revised manuscript. Please refer to line 107 for further confirmation.

7. 89-92 "From these methods, the best noise reduction approach was selected by analyzing the post-processed results based on the following criteria:

(1) relative number of negative values left;

(2) retention of detailed information attributed to microenvironmental changes;

(3) reduction of artificial peak values (is this correct?); and

(4) retention of detailed information on microenvironmental changes after background correction (is this correct?)."

Response: Thank you for your correction. That is correct (3rd and 4th criteria). And we have made modification regarding the criteria on the sentence. Please refer to line 110-113 for further confirmation.

METHODS

General comments:

1. Some studies cited in this section used the older model AE51. Either clarify in one sentence or two that the studies you are referring here pertain to those that used a variety of portable absorption photometers for various applications, or remove citations which used the AE51 and not the MA200.

Response: Thank you for your correction. AE51 is a predecessor instrument to the MA200, and this instrument has demonstrated some sensitivity to mechanical shock during mobile measurements. Therefore, We mentioned the instrument AE51 as a reference for MA200.

COMMENTS TO THE AUTHOR:

1. What methods were taken to account for the sensitivities of the instrument? The AE51 is known to be highly sensitive to vibrations, and sudden changes in the environment like RH and temperature. It is highly likely that the MA200 would have similar sensitivities, as also shown by Düsing et al. (2019) when it comes to strong RH variability. There was no detailed description on how the instrument was handled during the mobile measurements. This is vital since one of the criteria of the study is the retention of signals due to microenvironmental changes after noise reduction and identification of "peak samples". It must be noted that the instrument may produce false peaks/signals (in either direction) as a result of vibration or sudden change in RH and T. This is even more significant given that very few or even data from only a single mobile measurement round was used in the analysis. Taking these sensitivities into account in the measurement itself, would strengthen arguments made on the retention of "peak signals/samples".

Response: Thank you for your concern and suggestion. Firstly, the MA200's predecessor, the AE51 instrument, has shown some sensitivity to mechanical shock, relative humidity and temperature during mobile measurements. For mechanical shock, we have placed the MA200 in a carrying bag and secured it to a trolley, thus preventing mechanical shock during mobile sampling. For the sensitivity of the MA200 to relative humidity and temperature, the comparative measurements of the MA200 and the stationary Aethalometer (AE33, Magee Scientific, Berkeley, USA) showed good agreement between stationary measurements taken during each walk. Briefly, the AE33 was used to monitor black carbon at the same time as the MA200, following that, the AE33 was placed in a container (fixed monitoring site) while the MA200 was used outdoors (in a stroller) while walking alone with different relative humidity and temperature. This phenomenon had no effect on the accuracy of the black carbon concentration for either instrument (Pearson's r = 0.933, below figure). The brief description about mechanical shock, relative humidity and temperature has been added in the revised manuscript. Please refer to line 140-147 and 383-396 for further confirmation.

Mechanical shock. The following text has been added to section 3.2:

"It should be noted that a predecessor instrument to the MA200, the AE51, has demonstrated some sensitivity to mechanical shock during mobile measurements (Cai et al., 2013). Apte et al (2011) observed spurious, 1-3 second spikes of ± 200 - 2,000 μg/m^3 while monitoring black carbon in an auto rickshaw. When AethLabs took control of manufacturing the AE51, which was originally produced by Magee Scientific (Berkeley, CA, USA), instrument opto-electronics were redesigned to reduce such sensitivity (circa 2012). Researchers using redesigned AE51 demonstrated only a small effect on data. For example, Hankey (2014), using the same means of identifying such spurious measurements as Apte et al (2011), observed that approximately 1-2 % of their data collected via bicycle trailer were attributed to spurious mechanical shock. Supporting this improvement, Cai et al (2013) found evidence of a substantial improvement in data quality related to vibration-related spikes after an equipment upgrade by AethLabs, which reflected the aforementioned improvements to opto-electronics. In addition, there were no major mechanical shocks to or unique vibrational effects on the stroller and no major different of accelerometer data in the raw data, precluding these as potential con-founders on all 3 instruments."

RH and Temperature: The following text has been added to section 2.1:

In addition, it is worth noting that when the AE33 was used for monitoring black carbon at the same time as the MA200, the AE33 was placed in container, while MA200 was used outdoor (in the stroller) during the individual walks, which may have different relative humidity and temperature. This

phenomenon did not influence the consistency of eBC concentration measured with both instruments.

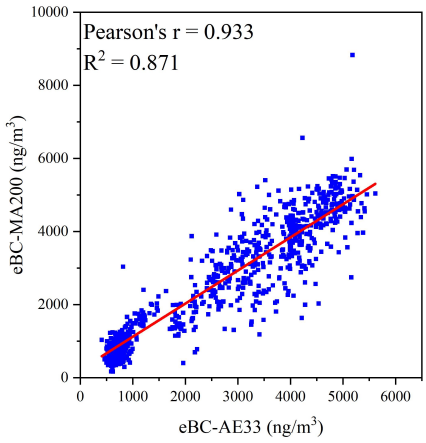

Figure R1 . The comparative measurements of the MicroAeth MA 200 with AE33.

2. You have to mention at some point that you only used data from one wavelength of the MA200.

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 119-121 for further confirmation.

3. Are the data already compensated? Was the internal DualSpot loading compensation operational during the measurements?

Response: Yes, the data are already compensated.

Yes, it was internal DualSpot loading compensation operational during the measurements

4. In which part of the analysis did you use Measurements 1-3?

Response: The measurements 1-3 was used for the average black carbon concentrations of raw data, ONA-processed, LPR-processed, and CMA-processed data (Table S2), negative value proportion and noise reduction (Table 2), and COV analysis. Please refer to line 281, 341 and 357 for further confirmation.

5. line 100 "In mobile monitoring, the MA200 can be used to estimate personal exposure and quantify eBC mass concentrations in different microenvironments."

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 123-125 for further confirmation.

6. line 108-109 "In order to reduce the noise of the data obtained with high time resolution, smoothing algorithms can be used."

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 132-134 for further confirmation.

7. line 112 "…this study analyzed BC data collected from…"

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 137 for further confirmation.

8. line 114 Why is it necessary to do further noise-reduction when there is already an on-board

signal-processing? What is the principle behind the on-board signal processing and how does it differ from the methods investigated in this study?

Response: Thank you for your concern. on-board signal-processing in MA200 reduce the noise of the black carbon signals, not the black carbon concentration. So, on-board signal will not influence the black carbon concentration. And the black carbon concentration data acquired by the instrument is the raw data without post-processing. Therefore, our analysis was only smoothed once. To avoid misunderstanding, we deleted this sentence.

9. line 115 Remove "microAeth®", Please provide some summary statistics of this comparisons.

Response: Thank you for your correction. We have removed "microAeth®" and added statistics summary of this comparisons in our revised manuscript. Please refer to line 140 for further confirmation.

10. 116-117 Please clarify. Was the intercomparison between the MA200 and AE33 done during the walks (within one walk, the Ma200 stopped in the vicinity of the AE33 for short intercomparison)? How long were the intercomparisons? I understand that these results were presented in a previous publication, but summary statistics would aid readers.

Response: Thank you for your correction. Yes it was, the intercomparisons between the MA200 and AE33 was done before and after walks for about 30 min to 60 min. The statistics summary of this comparisons was added in our revised manuscript. Following that, we have modified and revised through our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 138-143 for further confirmation.

11. line 119 Delete this: "To give intercomparison between the instruments…" You may start this sentence immediately at "To demonstrate the unit-to-unit comparability between the MA200 units, we performed intercomparisons at fixed monitoring stations and during collocated mobile measurements."

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 145-149 for further confirmation.

12. line 138 What does "To control for relative patterns in environmental exposure" mean?

Response: We are sorry that this sentence is difficult for the reader to understand.. It means "To control the different land use types of microenvironment". In order to avoid misunderstanding of the reader, we have modified and revised through our manuscript. Please refer to line 164 for further confirmation.

13. line 139 "…the mobile measurements were carried out on the right side of the road simulating people's common habits…"

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 165-166 for further confirmation.

14. line 142 I suggest either removing "air" (as it is vague) or "exposure" in this sentence.

Response: Thank you for your correction. We have removed "air" in this sentence.

15. line 144 Remove “with” after 4 h.

Response: Thank you for your correction. We have removed “with” in this sentence.

16. line 149 COV and TPRS are not yet defined prior to this.

Response: Thank you for your correction. We have introduced the complete terms of COV and TPRS. Please refer to line 181-182 for further confirmation.

17. line 158-165 This section could improve to briefly describe HOW the ONA reduce noise in microaethalometer data. How are the three parameters used to do this noise reduction? I believe this could greatly help readers in understanding Fig. S2 and, of course, the following analyses. LPR and CMA were aptly described in the following subsections, it would be great to elaborate a bit on ONA, too.

Response: Thank you for your correction. We have briefly described how the ONA reduce noise in microaethalometer data and revised through our manuscript regarding your concern. Please refer to line 199-210 for further confirmation.

18. line 169-173 Please briefly describe “smoothing number”, as the determination of this “smoothing number” is similar for that of CMA. How did you arrive at the values 15, 7, and 3?

Response: The smoothing number of points is the number of data that need to be calculated from the original data. Following that the “smoothing number” is similar for that of CMA.

The smoothing number was obtained as the following formula

smoothing number = distance/ speed / interval time

100 (m)/1.3 (m/s)/5s=15.38≈15

100 (m)/1.3 (m/s)/10s=7.69≈7

100 (m)/1.3 (m/s)/30s=2.56≈3

19. line 182 “… the number of remaining negative values was determined.” Also, what “number” of remaining negative values would imply a “good” noise reduction method? Is it simply a comparison of the treated data and whichever has the least number of negative values gets the point?

Response: Thank you for your correction. The “number” of remaining negative values would imply a “good” noise reduction method, when the treated data has a few number of negative values.

20. Line 182-189 This paragraph could be greatly improved. I find the structuring of the sentence hard to understand, as well as the looping the same idea. Please simplify this and improve the writing for better readability. I suggest starting again here with the “criteria” you have for selecting the best noise-reduction approach. I understand you already enumerated them in line 145, but it was within the brief list of the process of the investigation.

Response: Thank you for your correction. We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 190-195 our revised manuscript for further confirmation.

21. line 207 It is unclear to me how the background estimation and correction is related to the investigation of the noise-reduction approaches.

Response: The background estimation and correction does not affect the instrument noise reduction, but rather the instrument noise reduction affects the true value of the background estimation and correction. We performed background estimation and correction to confirm which post-processing method performs better based on their background value and microenvironment character. Therefore noise reduction, which better reflects the background estimation and correction, is one of the criteria for assessing treated data in this study.

We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 263-267 our revised manuscript for further confirmation.

22. Line 217-220 This paragraph is better suited after the description of the noise reduction approaches. As for the 3rd criteria, it would help to specify what would make a noise reduction method "good". Is it it's ability to remove or retain these peaks? The criteria in judging which method is "good" should be crystal clear.

Response: Thank you for your correction. We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 190-195.

For the 3rd criteria, after noise reduction, we compare the reduction values and the number of peak samples to further evaluate the noise reduction methods. Briefly, when the reduction of peak value is high, the treated data has a high peak noise reduction without removing the numbers of peak-samples. Therefore, the method with high reduction of peak value and retaining the number of peak-samples after postprocessing was selected as the best method.

We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 254-261 our revised manuscript for further confirmation.

RESULTS AND DISCUSSION

General comments

1. 3.1 please improve the structure of the sentences

Response: Thank you for your correction. We have restructured the 3.1 section sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 286-341 our revised manuscript for further confirmation.

2. 3.2 please explicitly distinguish between "peak samples" and "peak values"; and then in line 190 you also have "peak-value sample". These are all quite confusing.

How are the "proportions retained" calculated? For instance, in the 5-s data, 42.1% of the raw data were negative values. After post-processing, "negative values retained 33.3% for LPR and 26.1% for CMA". Are the 33.3% and 26.1% from the total amount of negative values or from the whole dataset? Please include in your methods how these numbers are calculated.

Response: Thank you for your correction. We have determined to use "peak-samples" in all the revised manuscript. We have added how are the "proportions retained" calculated in the main text.

Please refer to line 235-236 our revised manuscript for further confirmation.

3.3.4 why is background correction not applied to the Munich dataset? As I understand, one of the criteria for choosing CMA was its robustness to background correction.

Response: Thank you for your correction. We have analyzed the background correction of the Munich dataset and added it in the main text. As a result shown, after treated by CMA, the background concentrations showed few numbers of negative proportion (Fig. S8), suggesting the CMA method could be applied for black carbon postprocessing in another city. Please refer to line 449-450 and Fig. S8 our revised manuscript for further confirmation.

4. line 230 Please elaborate on the explanation. I find it quite insufficient, particularly, in the ONA paper of Hagler et al., 2011, they published results of applying ONA on 1-s data of SootGen, stove, and mobile monitoring. Van den Bossche et al. (2015) also used ONA on 1-s data from AE51 in field measurements. Is this an instrument issue? Or an algorithm issue?

Response: Thank you for your correction. As mentioned in section 2.2, the eBC average concentration is not high enough for this analysis in the city center of Augsburg, Germany, (measured at 2.62 $\mu g/m^3$ in winter by Gu, (2012)) thus in lower concentration, the ATN is more sensitive to the high time resolution. We have briefly elaborated this part in the revised manuscript. Please refer to line 295-298 for further confirmation.

5. line 237 I do not understand the last part of this sentence. I think, I know what you are trying to say, but it's not coming across to the reader clearly.

Response:Thank you for your correction. We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 320-323 for further confirmation.

6. line 240 Please be cautious of using the term "significant" here, particularly, that the analyses are based on comparability of statistical analyses of the raw data. I suggest the term "strong" here in place of "significant".

Response:Thank you for your correction. We have modified the related sentence. Please refer to line 324 for further confirmation.

7. line 242 This is not a complete sentence.

Response:Thank you for your correction. We have modified the related sentence. Please refer to line 325-327 for further confirmation.

8. line 243 Change "mitigating" to "decreasing".

Response:Thank you for your correction. We have modified the related sentence. Please refer to line 326 for further confirmation.

9. line 240-245 A deeper discussion on the differences of the 3 noise-reduction approaches could greatly improve this part. In essence, this part was merely a presentation of results which are already in Table 2.

Response: Thank you for your correction. We have added more discussion in this part to improve

the presentation of results. Please refer to line 324-331 for further confirmation.

10. Fig. 2 The unit should be nanograms. Am I right to assume that Fig. 2 is just same as Fig. 1 but only with the 10 s time resolution? If so, I do not see any added value in having this figure separated. The point you made in lines 240-245 is already clear in Fig. 1.

Response: Thank you for your correction. Fig. 2 (original version) with interval time 10 s is a part of Fig. 1. Therefore, following your suggestion, we removed the Fig. 2.

11. line 255 Table 2 It is unclear for me how the "noise reduction effect" was calculated. Please include in the methods section how these numbers are calculated and defined, including the "negative decline rate".

Response: Thank you for your correction. We have briefly described how to calculate the proportion of negative values and the reduction value of peak-samples in the method section 2.5.1 and 2.5.2, respectively. We have not longer used "negative decline rate" in our revised manuscript. Please refer to line 227-236 and 254-261 for further confirmation.

12. line 257 In this section, is my understanding correct? You want to evaluate two things about the "peaks": 1. # of peaks left after noise-reduction; 2. Magnitude of these peaks after noise-reduction Is this right?

Response: Yes, you are right. The text has been revised to make it more comprehensive.

13. line 264 How is the "reduction effect" calculated?

Response: Thank you for your correction. We have briefly described how to calculate the "the reduction value of peak-samples" in the method section 2.5.2. Please refer to line 254-261 for further confirmation.

14. line 263-269 It was not apparent right away that these results are already in Table 2. This could be solved by adding more information in the Table caption. Again, please give more information as to how these numbers are calculated or defined. Also, include the mean values in the table and not just the range so the readers can connect the numbers in this paragraph to the table.
Do these numbers mean that CMA reduces the magnitude of the peak values greater than the other two noise reduction approaches? If so, what is the main criteria here? Do you want a noise-reduction algorithm that retains the magnitude of these peaks? Do you have a threshold where you say the algorithm diminished the peaks "too much"? A bar graph comparing raw and processed data for all your parameters would help clarify these compared to Table 2 alone.

Response: Thank you for your suggestion. We have modified the Table 2 (adding the mean values) and added more information in the Table caption. As mentioned before, we have added how the proportion of negative values and the reduction value of peak-sample are calculated in the method section 2.5.1 and 2.5.2, respectively.
Yes, these numbers mean that CMA reduced the magnitude of the peak values greater than the other two noise reduction approaches, but retained the all number of peak-samples. Therefore, the criteria is the method with high reduction of peak value and retaining the number of peak-samples after postprocessing was selected as the best method. Following that, there is no specific threshold for the magnitude of peak reduction. However, it is same with the criteria as mentioned before. Following your

suggestion, we have added a bar graph comparing raw and processed data for the negative values proportion and average reduction value of peak-sample (Fig. S4).

15. line 273-274 This sentence is not clear. Did you mean to say, that based only on the # of remaining "peak samples", CMA performed better than the other approaches?

Response: Thank you for your concern. Comparing the three postprocessing methods, CMA retains all number of peak samples despite the highest reduction in their magnitude, which highlights other micro-environmental characters and is helpful to identify the actual peak-sample location and further identify the source of pollution. However, ONA has lowest reduction, but it may omit micro-environmental characters, while LPR has higher reduction than ONA, but it retained higher proportion of negative value. Therefore, CMA performed better than the other approaches. We have restructured and modified the sentence regarding your concern to make the reader easier following our manuscript. Please refer to line 358-362 for further confirmation.

16. line 288-289 I do not understand how CMA, which "greatly reduces" the peaks (magnitude and number) is helpful in identifying "hotspots", in a sense. For instance, if this peak that is related to a source happens a few moments before or after a lower (below the COV threshold) peak, and it is greatly reduced by the CMA method, wouldn't that further blur the impact of this single source? I believe, a better criterion is a noise-reduction method that does not greatly reduce the magnitude of these peaks, particularly for exposure studies where every real signal is important.

Response: Thank you for your concern. After reanalysis for paek-samples identification, the three postprocessing methods have retained all the number of peak samples, but they have different reduction pattern of peak-samples after postprocessing. In this regard, CMA retained all peak samples despite the highest reduction in their magnitude. Therefore, CMA highlights microenvironmental trends while preserving the identity of peak-samples, facilitating the identification of local pollution sources, and may thus be a better postprocessing method than ONA or LPR (Table 2, Fig. S4b). Moreover, after CMA postprocessing, the treated data did not blur the effect of a single source and was useful to identify more sources or hotspots of air pollution. In order to avoid misunderstanding this part, we have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 358-362 for further confirmation.

17. Fig. 3 The statement that these "spatial peaks" (Fig. 3a) are due to traffic and street canyon configuration could be better justified with a map that has spatially averaged eBC mass concentrations along the route. This also would prove the quality of the collocated measurements of the three MA200 and assure the reader that the peaks are due to local sources and not an instrumental artifact. I mean, you already have the data (running with 3 MA200 at the same time). Please provide more information in the figure caption such as the measurement number, to inform the readers that this is data from one run only. Please also improve Fig. 3a by adding time stamps

in the map to help readers reconcile the spatial plot with the time series.

Response: Thank you for your suggestion. We have analyzed the three different MA200 to prove the peaks are due to local sources and not an instrumental artifact. The results showed there were no major differences in the hot spot areas shown by the measurements of the 3 instruments (Fig. S4). it further justified that these peak-samples were due to traffic and street canyon configuration. Following that, we have briefly described this part in our revised manuscript. In addition, we have provided more information in the figure caption and added time stamps in the map (Fig. 2a, revised manuscript), to help readers reconcile the spatial plot with the time series. Please refer to Fig. 2 and our supporting information for further confirmation.

18. Line 295 This sentence can be simplified for better readability.

Response: Thank you for your correction. We have restructured the sentence and revised our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 398-400 for further confirmation.

19. line 306 What is "minus absolute value"?

Response: minus absolute value refers to *most*-negative values (i.e. negative values of the greatest absolute magnitude). We have revised this term in our manuscript regarding your concern to make the reader easier following our manuscript. Please refer to line 410 for further confirmation.

20. Fig. 4 It is unclear if the figure 4 a and b are background concentration or background corrected data? Please specify in the figure caption. What is "actual detection concentration"? What are the those encircled in dash black lines mean? Are they values below 1ug/m^3? If so, it would help to draw a zero-line, or magnify the scale such that the data around 0 ug/m^3 would be more visible. Improve figure caption.

Response: Thank you for your suggestion. We have modified the Fig. 4 (Fig. 3 after revised) following your suggestion together with the caption to improve the readability of the figure. The "actual detection concentration" is the measured concentration of the black carbon. The dash black lines (black circle lines after revised) indicated the background-corrected results after the ONA processing that values below 1ug/m^3. In order to make the reader easier following our manuscript, we draw a zero-line in the Figure 3 c and d. Please refer to line 419-423 for further confirmation.

21. Line 318 Change "certify" to "verify".

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 424 for further confirmation.

CONCLUSIONS

General comments

1. The broader significance of this study should be explicitly mentioned here.

Line 353-355 The first sentence is misleading. As I understand, it was not the goal of this study to "assess BC pollution", but to determine a suitable noise reduction algorithm for the new MA200.

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 480-481 our revised manuscript for further

confirmation.

2. line 369 “The data is available upon request by contacting the first author of the paper.”

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 489 for further confirmation.

3. line 375 “The authors declare no conflict of interests.”

Response: Thank you for your correction. We have modified and revised through our manuscript regarding your concern. Please refer to line 496 for further confirmation.

SUPPORTING INFORMATION

1. Table S1 Are these numbers mean or median of the 5040 data points? Either way, please indicate and provide range, either quantiles, minimum and maximum, or standard deviation. How long were the measurements?

Response: Table S1 are mean of the 5040 data points, the measurements were performed for 14 h. In our perspective, the standard deviation has very limited meaningfulness, because these data are ambient air measurements with a diurnal pattern, so that the standard deviation is very high, nevertheless, we have analyzed and shown in the following able.

Table S1 Comparative measurements of different MA200 in the fixed monitoring station (unit: ng/m^3, total N=5040 for each MA200, each measurement 14 h).

Measurements	375 nm	470 nm	528 nm	625 nm	880 nm
MA200-0051	818±183	833±226	812±224	810±232	774±251
MA200-0053	827±115	838±121	814±124	815±128	783±164
MA200-0059	870±186	866±155	830±163	840±161	814±213
MA200-0060	872±121	881±135	857±126	857±135	822±169
MA200-0155	856±103	855±112	842±107	840±115	830±138
MA200-0153	846±153	850±180	822±109	832±117	795±152
MA200-0159	825±108	845±148	818±108	832±110	780±157
Mean	844.9±22.1	852.6±16.6	827.9±16.5	832.3±15.9	799.7±22.2

2. Table S2 Another new terminology: “peak values number” Why is there no information for measurement numbers 5 and 7, 8-10?

Response: Thank you for your correction. After reanalysis all of the raw data and all postprocessing data (measurements 1-10), the number of peak samples did not change before and after postprocessing. Therefore, this table is no longer used in our revised manuscript. The detail information about it, please refer to line 356-357 for further confirmation.

3. Fig. S1 Did you use standard major axis regression here to account for the error on both axes? Technically, none of these instruments are “reference” instruments to merit the use of simple linear regression.

Response: Figure S1 (Fig. S2 after revised) is presented to demonstrate the unit-to-unit comparability between the MA200 units in the black carbon concentration during collocated mobile measurements. The results showed that there were no significant wavelength dependence between different instruments in different interval times. Therefore, in our perspective, the

standard major axis regression and “reference” instruments are very limited meaningfulness.

4. Fig. S2 Improve figure caption, indicate that this is for ONA.

Response: Thank you for your correction. We have improved Figure S2 (Fig. S3 after revised) caption. Please refer to our revised supplementary for further confirmation.

5. Fig. S3 Indicate that this is from CMA treated data.

Response: Thank you for your correction. We have improved Figure S3 caption (Fig. S6 after revision). Please refer to our revised supplementary for further confirmation.

6. Fig. S4 So, the measurements in Munich were not simultaneous like in Augsburg? The figure labels are too big. Why is there no analysis of the “peak values” and “peak samples” for the Munich dataset? As I understand, you were testing the applicability of the CMA method to a different dataset, but fail to run the entire series of tests which “proved” CMA to be the suitable method.

Response: Thank you for your correction. We have analyzed the “peak samples” (Fig. S8) and background concentration (Fig. S9) for the Munich dataset. The result showed that after treated by CMA, the peak-samples can be identified in different interval time (Fig. S8, and the background concentrations showed few numbers of negative proportion (Fig. S9).

Please refer to line 449-451 and supplementary for further confirmation.