

Interactive comments on “Methodology for High Quality Mobile Measurement with Focus on Black Carbon and Particle Mass Concentrations” by Alas et al.

Referee comments are noted in black and denoted with “RC”. Author replies/comments are in blue and denoted with “AC”. Changes in the manuscript are in blue as well, italicized, and denoted with “Change in Manuscript”

We would like to thank the Referee for recommending this manuscript for acceptance and for the constructive comments. Please find our response to each of the comments below. Attached is the revised version of the manuscript with the changes marked.

## **Anonymous Referee #2**

### GENERAL COMMENTS:

RC: 1. It would be interesting if an estimate of errors would be provided “simulating” cases in which some of the steps suggested are not followed. This is because it is likely that during future campaigns similar to this all the instrumentation necessary for the detailed comparison might not be available or within the budget of a project. For example, what would be the error introduced if one would not have the option of duplicate runs? This could be easily quantified with the data at hand.

AC: Thank you for your comment. We understand the need to estimate errors for cases when the methodology proposed is not feasible. For the most part, specifically the quality checks in laboratory and field, an infinite number of technical errors can occur which would be difficult to estimate. One of the main goals of the method proposed is to minimize, detect, and address these errors as soon as possible. Although, we do agree that we can estimate the errors that we experienced for the following parts, especially the ones we have experienced ourselves:

- Parallel runs → without another unit when the instruments had unflagged technical errors or drifts, and in the absence of live viewing of the data, eBC mass concentrations and particle number concentrations had errors of 50% and 19-80%, respectively. We have mentioned this in the text but not quantified (Page 8 line 12 to 16).
- Refractive index and fine mode volume corrections for the OPSS data → The impact of these corrections on the size distribution derived PM<sub>2.5</sub> from the OPSS, we believe, is discussed comprehensively and quantified in Figure 13, page 22.

Change in Manuscript: Page 8 lines 12-16

*“For example, during the early stages of the campaign, analysis of the collocated measurements revealed that one AE51 was underestimating eBC mass concentrations by 50% due to weakening of the pump causing the flow to decrease. This was not flagged by the instrument, but because another AE51 was in operation, the error was identified and corrected immediately. Similarly, towards the end of the campaign, due to unidentified reasons, the sheath flow of one of the OPSS started to increase which resulted to an underestimation of the particle number concentration (PNC) across all size bins (19 – 80%).”*

RC: 2. The paper is mostly well written but there are several instances where verb-subject number agreement should be corrected (some examples in the specific comments section)

AC: Thank you for catching our grammatical mistakes. We have improved the manuscript in this regard and the changes are written below following your “specific comments”.

#### SPECIFIC COMMENTS:

RC: Abstract, Page 1, Line 18: “can provide following” should be “can provide the following”

AC: Changed.

Changes: Page 1, Line 18: *“The application of the methodology can provide the following results.”*

RC: Page 1, line 23: “physical meaningful” probably should be “physically meaningful”

AC: Changed.

Changes: Page 1, Line 23: *“...distribution using physically meaningful corrections.”*

RC: Page 1, line 26: “MPSS+APSS” should be “MPSS+OPSS”?

AC: In this sentence, we are referring to the “reference instrument” which is the combination of the MPSS and APSS (mobility and aerodynamic particle size spectrometers). For clarity, the following changes were made on the sentence involved:

Changes: Page 1, Line 24-26: *“Using size-resolved complex refractive indices and time-resolved fine mode volume correction factors of the fine particle range, the calculated PM<sub>2.5</sub> from the OPSS was within 5 % of the reference instruments (MPSS+APSS).”*

RC: Table 1: I wish they had also used photoacoustic or extinction minus scattering techniques to check for accuracy (not just precision).

AC: The MAAP and AE33 in the fixed station were both regularly quality-assured at the World Calibration Centre for Aerosol Physics in TROPOS, Germany. The instruments are quality-assured through intercomparisons of different filter-based instruments using generated pure black carbon particles of known mass absorption coefficients. This procedure is detailed in Müller et al., 2011, AMT.

Changes: No changes were made.

RC: Section 3.1.1: This is good, but on what particles will the aethalometer and MAAP comparison be carried out?

AC: For this case, to capture the performance of the AE51 in real world scenarios, the intercomparison between the MAAP and AE51 were carried out through parallel long-term measurements (overnight or over multiple days) of ambient air from the same inlet. The same procedure is done during in-field quality assurance as described in this study.

Changes: Page 6 Line 18-20 (Section 3.1.1) *“The AE51 units must be compared against a multi-angle absorption photometer, (MAAP Model 2012, Thermo, Inc., Waltham, MA USA), provided that both are connected to the same inlet, with ambient air to test the performance of the AE51 in real-world scenarios.”*

RC: Page 8, line 10: It would be good to provide the cause of the underestimation.

AC: The cause of the underestimation of one of the AE51 units was due to the weakening of the pump. The flow was decreasing and needed to be recalibrated which we did.

Changes: Page 8, Line 12-13 (previously line 10): *“For example, during the early stages of the campaign, analysis of the collocated measurements revealed that one AE51 was underestimating eBC mass concentrations by 50% due to weakening of the pump causing the flow to decrease.”*

RC: Page 8, line 13: Again, it would be nice to know the potential reason.

AC: Unfortunately, we never figured out why the sheath flow of one of the OPSS drifted during the campaign. We brought up the issue with the manufacturer but received no conclusive answer.

Changes: Page 8 Line 15-16 (previously line 13): *“Similarly, towards the end of the campaign, due unidentified reasons, the sheath flow of one of the OPSS started to increase which resulted to an underestimation of the particle number concentration (PNC) across all size bins (19 – 80%).”*

RC: Page 8, line 19-20: “The scatter plots on the right of each time series shows. . .” should be “The scatter plots on the right of each time series show” because the verb refers to plots (plural)

AC: Changed.

Changes: Page 8, Line 22-23 (previously line 19-20): *“The scatter plots on the right of each time series show the correlation between the two corresponding instruments.”*

RC: Page 8, line 25: “Large differences, on the other hand, were investigated further to determine if it is related. . .” should read “Large differences, on the other hand, were investigated further to determine if they are related” because the subject is “Large differences”

AC: Changed.

Changes: Page 8, line 28-29 (previously line 25-26): *“Large differences, on the other hand, were investigated further to determine if they are related to sources or technical malfunctions.”*

RC: Page 10: I had a little bit of a hard time to follow the section on “Convergence Analysis”

AC: My apologies. The text has been modified.

Changes: Page 10, Convergence Analysis section: *“The idea is to take the pollutant concentrations measured per run along a specific part of the route. Then take the cumulative (increasing number of runs) average (or median) of those concentrations. This procedure is done with high number of iterations to achieve high number of possible combinations of the runs. Convergence is achieved when the iterations has stabilized to an asymptotic behavior towards the desired metric (e.g. median concentration from that location from all runs). The number of runs when the iterations are within the specified threshold of deviation from the selected metric (criteria for convergence) then tells how many runs are needed to achieve the representative concentration.”*

RC: Page 11, line 11: “the data points has to be spatially” should be “the data points have to be spatially”

AC: Changed.

Changes: Page 11, Line 11: *“Therefore, to obtain the overall spatial distribution, the data points have to be spatially aggregated.”*

RC: Page 11, line 12: “data points that is not part of the route” should be “data points that are not part of the route”

AC: Changed

Changes: Page 11, line 12: *“Prior to spatial aggregation, the data cloud has to be cleaned by removing data points that are not part of the route (e.g. detours, inaccurate GPS points).”*

RC: Section 3.2.2: I would have liked some more guidelines on criteria to select background sites.

AC: We agree that more information regarding the selection of the background site is needed. We referred to the guidelines provided in the Air Quality Directive 2008/50/EC:

*“Urban background locations shall be located so that their pollution level is influenced by the integrated contribution from all sources upwind of the station. The pollution level should not be dominated by a single source unless such a situation is typical for a larger urban area. Those sampling points shall, as a general rule, be representative for several square kilometers”*

Changes: Page 13, Section 3.2.2: *“The fixed station containing reference instruments is crucial for the quality assurance of the mobile instruments and also for the determination of PM<sub>2.5</sub> mass concentration derived from the PNSD of the OPSS mobile measurements. Therefore, the selection of the fixed measurement site should be taken with care. An urban background location should be selected as fixed station, namely (as stated in the 2008/50/EC Air Quality Directive) a site located in an area that is not dominated by a single source and instead captures the combination of all the sources upwind of the selected site. For this study, the following criteria were followed: 1) the site should be inaccessible or has limited accessibility to vehicles; 2) the site should not be <100 m away from any main thoroughfare; 3) there should be minimal obstruction (e.g. buildings) in its immediate vicinity. The decision on the reference site location is also a balance between scientific aims and availability of space. Since this study was conducted in the city of Rome, the fixed station was placed inside a government-owned garden that is inaccessible to*

*most non-government vehicles and is 115 m away from the nearest trafficked road. The site can be considered representative of the fine particulate matter at urban background locations in Rome as its average values of PM<sub>2.5</sub> mass concentrations are consistent with typical values measured at the urban background sites of the local air quality monitoring network (cf. Table 4 in Costabile et al. 2017)."*

RC: Page 13, line 25: "measurements which uses" should be "measurements which use"

AC: Changed.

Changes: Page 13, line 25: *"...which is an advantage over filter-based measurements which use gravimetric analyses to obtain PM mass concentrations."*

RC: Page 13, line 28: "shape factor = 1" how good is this assumption?

AC: At the CARE urban background site, a large fraction of particles is expected to be aged in the atmosphere. This fraction is composed of submicrometer aged particles with diameters larger than approx. 100 nm, and is supposed to be spherical. Supermicrometer particles (e.g., dust) might be not spherical, but were excluded from the analysis.

Particles smaller than 100 nm might include shortly aged soot particles. The shape of these particles (usually fractal when freshly emitted) changes with their aging in the urban atmosphere. They can experience a conversion from fresh fractal to aged spherical shapes by becoming more compact and the overall particle spherical when coated in other inorganic and organic material. The shortly aged soot particles are supposed to be a fraction of the total BC (including aged biomass burning particles, as well). Therefore, we expect that the shortly aged soot particles account for less than approx. 10% of the PM<sub>10</sub> (i.e., average value estimated for BC/PM<sub>10</sub>). A smaller fraction of these shortly aged soot particles might be not spherical, but cannot be exactly quantified here.

Furthermore, PM<sub>2.5</sub> and PM<sub>10</sub> mass concentrations derived from this procedure with this assumption compared well with those measured by a beta-attenuation monitor at another urban background site in Rome as stated in the next paragraph. Although, we must say, the assumption that the particles are spherical could have also contributed to the lower correlation of PM<sub>10</sub> as larger particles becomes more irregular in shape with increasing size.

Changes: Page 14, line 6 – 7 (previously page 13 line 28): *"The conversion assumed aged spherical particles in the fine mode (shape factor = 1) as expected at urban background regions, and a size-dependent particle density (1.6 to 2 g cm<sup>-3</sup>)."*

RC: Page 14, line 18: "The agreement for PM<sub>10</sub> is lower" please quantify.

AC: Changed.

Changes: Page 14, line 18: "The agreement for PM<sub>10</sub> is lower ( $r^2 = 0.73$ ,  $y = 0.88x$ ), probably because of dust and marine aerosol events, which are supposed to modify the particle density used in the calculation of particle mass from particle number size distributions ..."

RC: Page 15, line 12: “AE51 data was” should probably be “AE51 data were”

AC: Changed.

Changes: Page 15, line 12: *“For the eBC mass concentration measurements, the AE51 data were averaged per minute to compare with the MAAP.”*

RC: Page 15, line 14: it should be “mass concentration is” or “mass concentrations are”

AC: Changed.

Changes: Page 15, Line 14: *“This increases our confidence that the measured eBC mass concentrations are reliable for the entire route.”*

RC: Page 21, line 7: either “unique CFf,vol are applied” or “a unique CFf,vol is applied”

AC: Changed.

Changes: Page 21, line 7: *“Finally, the deviation from the reference is significantly minimized when a size-resolved refractive index correction is used and a unique CFf,vol is applied for each run.”*