

Performance of microAethalometers: Real-world field intercomparisons from multiple mobile measurement campaigns in different atmospheric environments

Honey Dawn C. Alas^{1*}, Thomas Müller¹, Kay Weinhold¹, Sascha Pfeifer¹,
Kristina Glojek², Asta Gregorič^{3,4}, Griša Močnik^{3,5}, Luka Drinovec^{3,5},
Francesca Costabile⁶, Martina Ristorini⁷, and Alfred Wiedensohler¹

¹ Leibniz Institute for Tropospheric Research, Leipzig, Germany

² Department of Geography, Faculty of Arts, University of Ljubljana, Ljubljana, Slovenia

³ Center for Atmospheric Research, University of Nova Gorica, Ajdovščina, Slovenia

⁴ Aerosol d.o.o., Kamniška 39 A, 1000 Ljubljana, Slovenia

⁵ Condensed Matter Physics Department, J. Stefan Institute, Ljubljana, Slovenia

⁶ Institute of Atmospheric Science and Climate, National Research Council, Rome, Italy

⁷ Department of Bioscience and Territory, University of Molise, Pesche, Italy

Abstract

Small aethalometers are frequently used to measure equivalent black carbon (eBC) mass concentrations in the context of personal exposure and air pollution mapping through mobile measurements (MM). The most widely used is the microAethalometer (AE51). Its performance in the laboratory and field is well documented, however, there is not sufficient data in the context of its performance in different environments. In this investigation, we present the characterization of the performance of the AE51 through field unit-to-unit intercomparisons (IC), and against a reference absorption photometer from three MM campaigns conducted in drastically different environments. Five IC parameters were considered: i) study area, ii) location of IC, iii) time of day, iv) duration of IC, and v) correction for the filter-loading effect. We can conclude that it is crucial where and how long the IC have been performed in terms of the correlation between the mobile and reference instruments. Better correlations ($R^2 > 0.8$, slope = 0.8) are achieved for IC performed in rural, and background areas for more than 10 minutes. In locations with more homogenous atmosphere, the correction of the loading effect improved the correlation between the mobile and reference instruments. In addition, a newer microAethalometer model (MA200) was characterized in the field under extreme cold conditions and correlated against another MA200 ($R^2 > 0.8$, slope ≈ 1.0), AE51 ($R^2 > 0.9$, slope ≈ 0.9), and a stationary Aethalometer (AE33) across all wavelengths ($R^2 > 0.8$, slope ≈ 0.7). For MA200, the loading effect was more pronounced, especially at the lower wavelengths, hence the correction of the loading effect is essential to improve the correlation against the AE33. The MA200 and AE51 proved to be robust and dependable portable instruments for MM applications. Real-world quality assurance of these instruments should be performed through field IC against reference instruments with longer durations in areas of slowly changing eBC concentration.

Keywords: Portable instruments; Mobile monitoring; Black carbon; Instrument intercomparisons

* Corresponding author. Tel: +49 341 2717-7372; Fax: + 49 341 2717 99 7060

E-mail address: alas@tropos.de

46 INTRODUCTION

47

48 Black carbon (BC) particles are an increasingly important air pollutant in terms of human
49 exposure to combustion-related emission sources such as traffic and wood burning. Since these
50 particles are highly variable in space (Peters et al., 2014a; Rakowska et al., 2014) due to their size
51 (20-300 nm), their spatial distribution should be determined with high resolution to estimate
52 different exposure scenarios. This has become possible with the rise of portable instrumentation
53 with fast measurements placed in mobile platforms.

54 The microAethalometer (microAeth® AE51 model, Aethlabs, San Francisco, CA) is
55 currently the most widely used portable absorption photometer for measurements of BC mass
56 concentrations aboard mobile platforms. The AE51 measures the attenuation of light (880 nm)
57 through a particle-loaded filter (T60 Teflon coated glass fiber) and converts this to an equivalent
58 black carbon (eBC; (Petzold et al., 2013)) mass concentration using a fixed mass attenuation
59 coefficient (MAC). The time resolution can be set from 300 seconds down to 1 second. It is also
60 small enough to fit in a pocket, making it extremely portable. This instrument is the most
61 characterized portable instrument for eBC measurements in terms of filter loading effect (Cheng
62 and Lin, 2013; Good et al., 2017), and sensitivity to sudden changes in relative humidity,
63 temperature (Cai et al., 2013), and vibration (Apte et al., 2011). To investigate the performance
64 of any instrument, it is often compared against “reference” instruments with operating principles
65 considered as standard method. While there is no standard method to measure BC, there are three

66 potential candidate methods to measure aerosol absorption ab-initio. These methods are capable
67 of measuring the absorption of particles suspended in the air, rather than collected on a filter. The
68 first method measures extinction and scattering, and calculates the absorption as the difference
69 between them. This method works for single-scattering albedo values below about 0.8. The
70 second is the photoacoustic method which measures the pressure waves generated by modulated
71 absorption of light by aerosols, subsequent heating and change in the density of the air. Usually, a
72 resonant cavity is employed to amplify the signal. This resonance needs to be tracked and is
73 sensitive to changes in ambient conditions. The signal is also dependent on the losses to latent
74 heat as the coating of the particles changes phase. The third is the photothermal interferometry
75 which employs a similar heating of the sample, measuring the change in the refractive index
76 following the change in density. All methods require drying of the sample. Filter photometers
77 are commonly used in the field. Light from an LED source passes through a particle-loaded filter,
78 and is detected by a photodiode. The amount of light attenuated by the light-absorbing particles
79 trapped in the filter is proportional to the concentration of these particles. The light absorption
80 coefficient of the particles is converted to mass concentration of light absorbing carbon by
81 dividing it with the MAC. The calculation assumes the filter properties in the derivation of the
82 absorption coefficient introducing uncertainties into the reported parameters. This method
83 provides “mass equivalent black carbon” or “eBC” as recommended by Petzold et al., 2013.

84 However, MAC can vary widely as a function of particles' physical (i.e. coating) and chemical
85 (composition) properties across different locations and atmospheric conditions. Consequently,
86 this is a source of uncertainty for optically derived mass concentrations of eBC. Despite this,
87 light absorption instruments have been used in regular air quality monitoring in fixed locations
88 across the globe due to its ease of use, online measurements, and high time resolution. In
89 previous studies, the AE51 have been compared against rack-mounted versions of absorption
90 photometers as they have the same operating principle and feature high time resolution. For
91 instance, Viana et al., (2015) performed experiments on the unit-to-unit variability of the AE51
92 as well as intercomparisons (IC) against a reference instrument (multiangle absorption
93 photometer or MAAP model 5012, Thermo, Inc., Waltham, MA USA) in a single location.
94 However, this investigation was not done in the context of mobile measurements (MM). Birmili
95 et al. (2013) and Alas et al. (2019) among others, have emphasized the importance of doing field
96 IC between mobile devices and reference instruments during a mobile measurement round to
97 ensure the quality of the data obtained. This way, the eBC mass concentrations obtained from the
98 AE51 is harmonized with a more stable and quality-assured instrument. This has been done in
99 practice by previous studies, but rarely focused on. The question remains: how does the AE51
100 compare against a reference absorption photometer in the context of MM performed over
101 different environments and what factors influence their correlation?

102 The aim of this investigation is to determine the intercomparability of the AE51 against
103 each other and against reference instruments using the data from three mobile field studies
104 performed in multiple locations with varying sources and atmospheric conditions. These
105 measurement campaigns used the same mobile platform, and followed the same experimental set
106 up. All the mobile measurement routes included fixed stations that has absorption photometers
107 serving as reference instrument for eBC mass concentrations for field IC of varying duration.

108 In addition, the field performance of relatively new, portable, 5-wavelength absorption
109 photometers with PTFE filter material (microAeth® Model MA200, Aethlabs, San Francisco,
110 CA) was investigated in comparison to its predecessor (AE51) and to a rack-mounted 7-
111 wavelength aethalometer (Model AE33, Magee Scientific, Berkeley, CA, USA). This study
112 focuses on the unit-to-unit variability of MA200, its intercomparison with the AE51, and the
113 reference instrument, AE33. Sensitivity of this instrument to different factors such as temperature,
114 pressure, humidity, etc. will not be analyzed as these have been published elsewhere (Düsing et
115 al., 2019)

116 **METHODS**

117 **118**

119 *Locations*

120 This section briefly describes the instrumentation used as well as the mobile measurement
121 experiments performed in the following campaigns:

- 122 i. Metro Manila Aerosol Characterization Experiment (MACE-2015, “Manila campaign”),
123 Philippines
- 124 ii. Carbonaceous Aerosol in Rome and Environs (CARE-2017, “Rome campaign), Italy
- 125 iii. Loški Potok, Slovenia (LP-2018, “Loški Potok campaign”).

126 ***Mobile measurements (MM)***

127 The MM carried out for all three campaigns were more or less similar and descriptions
128 can be found in the references listed in the footnotes of Table 1. Briefly, the AE51 is placed
129 inside a hard-case, water-proof, backpack. The aerosol enters the system through a 1-m stainless
130 steel inlet. The aerosol sample then passes through a silica-gel drier, which dries the aerosol and
131 dampens the effects of sudden changes in humidity, before entering the AE51. A microcomputer
132 logs the data and synchronizes it with the location information obtained by the GPS unit. The
133 AE51 was operated with a flow of 100 mL min⁻¹ and time stamp of 1-s. However, to minimize
134 noise and still have high resolution data, the 10-second median of the eBC mass concentrations
135 were obtained from the 1-s data. For the case of the Loški Potok campaign, the MA200 was
136 additionally installed downstream of the silica gel dryer and the flow rate was set to 150 mL min⁻¹.

138 The MM were done along fixed routes, which covered different microenvironments.
139 Measurements were repeated along the routes to obtain representative information. All routes

140 passed by a fixed station which contained the reference absorption photometer where the
141 “runners”, who carry the backpack, can stop for some time to intercompare the AE51 and
142 MA200 (in the case of Loški Potok campaign) with the reference instruments (not on the same
143 inlet, but in the same vicinity).

144 The different transit times of the aerosol being sampled through the inlets of the backpack
145 and the container could influence the correlation between the two, especially in IC locations with
146 rapidly fluctuating eBC concentrations. This was addressed by synchronizing the data loggers
147 with internet time and aggregating the dataset. The 10-s median from the mobile devices were
148 further aggregated to 1-minute to be comparable to the reference instruments. This smooths out
149 the possible lag in the measurements.

150 *Field campaigns*

151 Table 1 describes the different campaigns and their mean ambient conditions. Table 2, on
152 the other hand, summarizes the IC parameters for each campaign. For summary of the description
153 of each instrument used in this study, including operating principles and technical specifications,
154 see Table A1. The number of IC periods done per site is listed in Table A2.

155 *Manila, Philippines*

156 MACE-2015 was performed in the highly urbanized metropolitan (> 10M inhabitants) of
157 Metro Manila, Philippines (tropical) in the summer time (March to June). During these months,

158 the weather in Manila is typically warm (around 30 °C) with minimal rain and cloud cover.
159 Winds are generally blowing from the East and gradually switches to Southwest at the onset of
160 the monsoon season (middle of May). Here, the main source of eBC particles is traffic due to the
161 lenient regulation of vehicular emissions and high volume of vehicles. For more information on
162 MACE-2015, the readers are directed to the following publications (Kecorius et al., 2017; Alas et
163 al., 2018; Kecorius et al., 2018; Kecorius et al., 2019; Madueño et al., 2019). Reference
164 instruments were set-up permanently (for the duration of the experiment) inside a building within
165 an urban background area (university campus). Simultaneously, an aerosol measurement
166 container was placed 300 m away on a street side (phase 1) along Katipunan Avenue for more
167 than a month. Then, the aerosol container was moved closer to the urban background station for a
168 week of IC (phase 2). Finally, it was moved until the end of the campaign 20 km away to a street
169 side along Taft Avenue (phase 3), which has street canyon configuration. The aerosol container
170 and urban background station measured eBC mass concentrations with a MAAP. Two mobile
171 measurement experiments were performed, one during phases 1 and 2, (including the urban
172 background station) and one during phase 3. For both mobile measurement experiments, the IC
173 periods between the AE51 and the MAAP were shorter than 5 minutes.

174 IC was performed by placing the aerosol backpack(s) near the measurement container
175 where the reference instruments are. For the urban street site, the aerosol container was ~2 m

176 away from the backpacks and with ~ 3 m height difference between their inlets. For IC at the
177 urban background site, there were two instances: IC against the reference instruments on the 4th
178 floor of a building (as mentioned above), and IC against the aerosol container when it was moved
179 near the same building for a week. The inlet of the of building site was ~55 m from the inlets of
180 the backpacks during IC periods. IC periods at the urban street and urban background were done
181 within one run. Alas et al., 2018 have demonstrated that the difference in concentration between
182 the building site and the aerosol container when they were in the same location was not
183 significant. The mean eBC mass concentrations (standard deviation) at the building site was 6.9
184 (4.8) $\mu\text{g m}^{-3}$, while at the ground site with the aerosol container was 7.6 (4.9) $\mu\text{g m}^{-3}$ showing that
185 particles in this location, specifically eBC particles, were spatially homogenous.

186 For the IC performed at the urban street canyon site, the same aerosol container was used
187 but was mounted on 1-m cement blocks. Therefore, the vertical distance between the inlets of the
188 aerosol container and the backpacks were ~4 m. The horizontal distance, on the other hand, is
189 approximately 2 m.

190 *Rome, Italy*

191 CARE-2017 was performed in the city of Rome, Italy, which is home to more than 3
192 million people. The campaign was done in February 2017 with temperatures ranging from 7 to 15
193 °C. Minimal rain events occurred during this time. The main sources of eBC particles were

194 vehicular emissions and domestic heating. For this campaign, an aerosol container was placed
195 inside a gated garden, which is considered an urban background area (Costabile et al., 2017).
196 Collocated MM with two aerosol backpacks with identical instrumentation were performed
197 around this station with a 30-minute IC duration (Alas et al., 2019). During this IC periods
198 against the reference instruments, the backpacks were placed ~ 3 m from the aerosol container
199 inlets horizontally, and ~ 4 m vertically. The IC period was performed 30-minutes into the 2.5
200 hour run.

201 *Loški Potok, Slovenia*

202 The measurement campaign was performed in the model region Retje, Loški Potok,
203 Slovenia, a populated, forested karst hollow with frequent ground temperature inversions and
204 residential wood combustion as the main energy source. MM were performed from December
205 2017 to January 2018 with temperatures ranging from -17.7 °C to 14.2 °C in the hollow. Two
206 stations with reference instruments were set up in the studied area (Glojek et al., 2018), one at the
207 bottom of the hollow in the Retje village (715 m a.s.l., rural village) and one on top of Tabor hill
208 (815 m a.s.l., rural background site). At both stations, eBC mass concentrations were retrieved
209 with AE33. Along the hollow, simultaneous MM were performed with a 20-minute IC at the
210 station in the village and with a 10-minute IC on top of the hill. The following instruments were
211 intercompared: the AE51 and the MA200 with the AE33. IC at both stations were performed

212 within one run (one filter). For the IC at the rural village, the backpacks' inlets were ~8 m
213 horizontally and ~ 2m vertically away from the inlets of the fixed station. For the IC at the rural
214 background, the horizontal distance between the inlets was ~2.5 m and the vertical distance was
215 ~3 m.

216 ***Data processing***

217 The loading effect in filter photometers is a bias, which reduces the apparent
218 concentrations relative to the ambient ones. The apparent reduction depends on the loading of the
219 spot. The filter-loading effect (FLE) is a non-linearity due to the saturation of the attenuation
220 (ATN) as the amount of the sample on the filter in the photometer continually increases – the
221 eBC mass should depend only on the change of attenuation in time, but due to saturation, an
222 ATN dependence is observed (Park et al., 2010; Segura et al., 2014; Drinovec et al., 2015). The
223 FLE on eBC mass concentrations measured by filter-based photometers have been studied
224 extensively. The two most direct way to detect this in post-processing is to plot the raw eBC mass
225 concentrations as a function of the attenuation (ATN), and to compare against a reference
226 instrument. The FLE depends on the type of particles sampled. Another method is the one
227 outlined in Good et al. (2017), where an experimental set-up in the laboratory including a
228 photoacoustic extinctionmeter (PAX) was used as a reference instrument, which, being not filter-
229 based, is not susceptible to FLE. However, as this instrument was not used in any of the

230 campaigns in this study, the eBC mass concentrations measured by the AE51 and MA200 were
231 instead compared against the AE33 which has a real-time FLE correction, and the MAAP which,
232 compared to the AE51, is less susceptible to the FLE (Petzold et al., 2005).

233 For the Manila and Rome datasets, three approaches to assess the FLE were performed:

234 1. BC(ATN) approach: Assessment of FLE on the whole dataset (measurements from the
235 entire routes) by plotting the eBC concentrations as a function of the increasing ATN.

236 2. Deviation (ATN) approach: Assessment of FLE during the intercomparison periods
237 (based on Masey et al. (2020)) analyzing the ratio and difference of eBC measurements between
238 AE51 and reference instrument as a function of increasing AE51 ATN.

239 3. Virkkula correction approach: Assessment of FLE by correcting entire datasets using
240 the Virkkula algorithm (Virkkula et al., 2007) with a loading parameter from literature.

241 For BC(ATN) approach, the entire mobile measurement was used to assess the FLE. Raw, 1-s
242 data from the AE51 was analyzed by plotting it against the ATN. To determine if FLE is present,
243 the eBC measurements were binned in intervals of 1 ATN. A linear fit was performed for both
244 the mean and median eBC measurements per ATN bin over the whole ATN range. Another
245 experiment was to fit the mean and median values over only a specific ATN range. In Drinovec
246 et al. 2015, they did not include the lowest and largest ATN values in the fitting due to low
247 frequency of eBC measurements at those values. In this study, we did this by doing the BC(ATN)

248 plots only for eBC values below the 95th percentile of ATN. If the fit has a negative slope, the
249 eBC is decreasing with increasing ATN, hence, there is a loading effect. Normally, the loading
250 parameter to correct the AE51 raw concentrations can be derived from the slope and intercept of
251 the regression line.

252 The deviation (ATN) approach follows that of Masey et al., 2020 to assess FLE during IC periods.
253 The raw eBC mass concentrations measured by the AE51 during the IC periods were taken and
254 aggregated to 1-min averages. Two statistical parameters were used to investigate deviation of
255 the measurements between the AE51 and reference instruments as a function of the ATN of the
256 AE51: the ratio (AE51/reference) and the difference (AE51 – reference). Similar to the first
257 approach, the slope of the linear fit indicates the FLE.

258 The Virkkula correction approach was performed to investigate if correcting for the FLE
259 significantly improves the AE51 measurements. The FLE correction algorithm by Virkkula et al.
260 (2007) was used to correct the dataset. The loading parameter ($k = 0.005$) applied here was taken
261 from Drinovec et al. (2017) which is supposed to represent a diesel dominated aerosol type. In
262 most of the studies involving AE51 measurements, the algorithm presented by Virkkula et al.,
263 2007 is used as it is very simple (Cheng and Lin, 2013; Dons et al., 2013; Peters et al., 2014b;
264 Van den Bossche et al., 2015; Van den Bossche et al., 2016). Hence, it was also used in this study.

265 For the Loški Potok campaign, a different loading parameter has to be derived for the
266 MA200 as it uses a different filter material from the AE51. Also, due to characteristics of the
267 route measured with mobile devices, BC(ATN) plot alone was not enough to determine the level
268 of the loading effect for MA200 instruments. Since each run started at the rural background with
269 lower eBC concentrations, continuing towards the village, where concentrations were usually
270 higher, the BC(ATN) plot was biased. Therefore, two different measures had to be considered in
271 order to determine the filter loading effect:

272 - Observed jump in measured concentration after the tape advance should be minimized
273 after the correction for the loading effect.

274 - Considering the contribution of sources at both stations, rural background and village, and
275 measurements obtained by the AE33, the absorption Å ngström exponent (AAE) is expected to
276 increase from the rural background toward the village. Therefore, the compensation should not
277 result in decrease of AAE with ATN.

278 *Analysis*

279 Correlation analyses were performed to determine the comparability of the mobile with the
280 reference instruments. For the IC of the mobile devices against each other (MA200 vs MA200;
281 AE51 vs AE51; MA200 vs AE51), a reduced major axis (RMA) regression was used to include
282 errors in both instruments. For the IC of the mobile devices against the reference absorption

283 photometers in different environments a simple linear regression was applied. We also
284 investigated the impact of different parameters (i.e. location, time of day, filter loading effect
285 correction, and duration of IC) to the intercomparability of the instruments.

286 More information on the performance of the AE51 during these campaigns (time series, etc.) are
287 already published in Alas et al. (2018)(Supplementary Material), Costabile et al. (2017), and Alas
288 et al. (2019).

289 All calculations were performed in R (R Core Team, 2019) using lmodel2 (Legendre, 2018) and
290 lme4 (Bates et al., 2015) packages. Data manipulation was done using the package dplyr
291 (Wickham et al., 2018). For the visualizations, ggplot2 (Wickham, 2016) package was used.

292

293 **RESULTS AND DISCUSSION**

294

295 *Unit-to-unit comparability of AE51*

296 For campaigns in Rome and Loški Potok, two aerosol backpacks with identical instrumentation
297 were used to explore the unit-to-unit variability of two AE51 units in real-world MM. The
298 models used were the AE51 S5 and AE51 S6, where the former is an older model.

299 Figure 1 shows the correlation analyses (RMA) between the two models during both campaigns.

300 It must be noted that exactly the same models were used for both campaigns. The correlation of
301 the two units is slightly lower in the Rome campaign compared to the Loški Potok campaign. In
302 Rome campaign, The AE51 S5 was 5% lower than the AE51 S6. This can be attributed to the

303 study area in Rome, which was in an urban area with higher variabilities of sources. Nonetheless,
304 the unit-to-unit variability of the two AE51 units during MM is low at around 5% at 10-second
305 time resolution.

306 *Intercomparability of mobile devices to reference instruments in different environments*

307 In this section, we explore how the AE51 performed in different environments using data from
308 three different campaigns in comparison to rack-mounted, widely used absorption photometers
309 (MAAP 5012 and AE33), which are considered as reference instruments. The AE51
310 measurements were aggregated to 1-minute averages to compare against the reference
311 instruments with 1-minute time resolution. The entire IC dataset for each campaign was used for
312 this correlation analysis and the results are shown in Figure 2. From this figure, it appears that the
313 AE51s performed best in Rome, followed by Loški Potok, and lastly in Manila. To determine
314 which other factors may have influenced the correlations, the following parameters were
315 investigated: i) the location of IC, ii) the correction of the filter loading effect, iii) the time of the
316 day when IC was conducted, and iv) the duration of IC.

317

318 *Location of IC*

319 The Manila and Loški Potok campaigns had multiple locations for IC. For Manila, IC was done
320 at an urban background site, one at a street side, and one at a street canyon. For Loški Potok, one
321 was done at a rural background region (up a hill) and one at a street side of a rural village. We
322 performed the correlation analysis again, this time not only as a function of the study area, but
323 also of where the IC was performed (Fig. 3). One can see now that, for Manila (Fig. 3(a)), the
324 low correlation ($R^2 < 0.5$, and slope = 0.75 and 1.5) between the AE51 and MAAP was due to the
325 IC done at the street side and street canyon. At the urban background region, the correlation is
326 high ($R^2 > 0.8$, slope > 0.8). In Rome, IC was only done at an urban background area so the

327 results are the same as in Fig. 2(b). Fig. 3(c) also shows good correlation between the mobile and
328 reference instruments, indicating that the area of Loški Potok has a homogenous distribution of
329 eBC particles. The poor correlation at street side IC in Manila is due to the higher variabilities
330 that arise from passing of vehicles, turbulence, and other local sources as well as the vertical and
331 horizontal distance between the inlets of the aerosol backpack and aerosol container. Although,
332 this does not mean that the AE51 do not perform well in areas with high spatial variabilities, it is
333 simply difficult to conduct an IC in such locations due to rapidly fluctuating concentrations. This
334 could be improved by connecting the backpack to the same inlet as the reference instrument, but
335 this would disrupt the MM. Therefore, to harmonize mobile instruments during mobile
336 measurement campaigns, IC done at atmospherically homogenous areas work best.

337

338 *The filter-loading effect*

339 All datasets were analyzed for filter-loading effect. From the three datasets, the measurements
340 from both AE51 and MA200 of the Loški Potok campaign were corrected for the filter-loading
341 effect. For the Manila and Rome datasets, the results of the three assessment approaches are
342 presented and discussed here. From the first approach, the BC(ATN) plots showed a dependency
343 on the route (Fig. A1 and A3), indicating that a single loading parameter cannot be derived,
344 because the area being studied has a very inhomogeneous atmosphere and specific areas with
345 different sources have to be analyzed separately. Unfortunately, there isn't sufficiently large data
346 set to derive an empirical k (Fig. A2). The deviation (ATN) approach showed similar results (Fig.
347 A4) to the first one. For the Katipunan route (Fig. A4 (a) and (b)), the ratio vs ATN plots shows a
348 negative slope (-0.0063) while the difference vs ATN has a positive slope (0.661). The Taft route
349 (Fig. A4 (c) and (d) shows negative slopes for both the ratio (-0.00117) and the difference (-
350 0.215). This is more indicative of an FLE. The Rome route (Fig. A4 (e) and (f)) shows positive

351 slopes as well (0.0028 and 0.0039). However, this approach may not be suitable for this study:
352 the IC periods (co-located AE51 and reference measurements) were performed in the middle of
353 the run – this means, that we would have only a fraction of the ATN range to analyze. To be able
354 to apply results from this approach, we would need eBC mass concentration data that is evenly
355 distributed over the whole ATN range, otherwise, it would be misleading to use a loading
356 parameter derived from this and apply it to the whole measurement route in urban areas. For the
357 third approach a value of 0.005 for k based on literature (Drinovec et al., 2017) was used,
358 representing a roadside aerosol for 880 nm. This was applied using the algorithm proposed by
359 Virkkula et al. (2007). Figure A5 shows that correcting for the FLE with the given k did not
360 significantly improve the eBC mass concentrations of the AE51 (3-8% increase).

361 Also, owing to the inhomogeneity of the study area, correcting the whole dataset with a single
362 loading parameter may cause an over/underestimation in specific parts of the route. Unlike in
363 Loški Potok, which is a rural area, the eBC levels vary widely and rapidly in urban environments
364 due to micrometeorology and high spatial variation of sources and their strengths. These
365 variations are also greater than the possible error caused by the loading effect. Hence, correcting
366 for it will not lead to any significant improvement of the AE51 eBC measurements.

367 Results of the three approaches suggest that there are no significant detectable FLE in the Manila
368 and Rome datasets. Dedicated experiments are necessary to develop methods that would lead to
369 derivation of a loading parameter appropriate for data obtained from MM in urban areas.

370 In this section, the impact of the FLE correction on the Loški Potok data is discussed. Figure 4
371 shows the IC between the AE51 and the AE33 in the two IC locations in Loški Potok for both
372 uncorrected and corrected AE51 eBC data. The correlations between uncorrected eBC measured
373 by microAethalometers (AE51_S5 and AE51_S6) and the reference instrument AE33 were good
374 at both stations, rural background and rural village, as seen on Figure 4 (red points). The slope of

375 uncorrected eBC measurements for both mobile instruments was higher at the urban background
376 station (0.88) than for the village station. This can be explained by taking a closer look into the
377 course of each run, since every single run started at the rural background, where the attenuation
378 of the filter was low, continuing towards the village, where filter attenuation was already high.
379 This leads to increased loading effect and consequently lower slope, when comparing to the
380 reference AE33 in the village: 0.81 and 0.84 for the AE51 S5 and AE51 S6, respectively (Fig. 4
381 II.). The same loading effect correction ($k=0.005$, also representative of freshly emitted particles
382 from wood burning (Drinovec et al., 2017)) was applied for the whole course of mobile run with
383 the AE51. This procedure improved the agreement between both instruments (AE51 and AE33)
384 with slopes close to unity: 0.92 (S5) and 0.93 (S6) at the rural background and 0.95 (S5) and 0.96
385 (S6) in the rural village. Variability and level of eBC concentrations was lower at the rural
386 background than at the rural village station, owing to more distant emission sources with lower
387 variability at the rural background station.

388 *Time of IC*

389 The time of the day when the IC was performed was also investigated (Fig. 5) to determine if the
390 intercomparability of the AE51 and reference instruments is affected by the variability of the
391 meteorological conditions and sources within a day. The time of IC was segregated to morning,
392 afternoon, and evening as proxy to variations in incoming solar radiation, temperature, and height
393 of boundary layer. Fig. 5 shows that there is no obvious dependence of the intercomparability to
394 the time of IC. In all IC locations, the AE51s were able to capture the eBC mass concentrations
395 regardless of the variabilities within the day.

396

397 *Duration of IC*

398 Lastly, the duration of IC was investigated. Only data from rural background, rural village,
399 background and urban background regions were used for this analysis which is shown in Figure.
400 6. The duration of IC increases from Fig. 6 (a) to (d) and it shows that longer IC durations lead to
401 better correlation and harmonization of the mobile with the reference instruments. Longer
402 durations provided more time for the mobile instruments to adjust to its surroundings as they are
403 not in the same inlet as the reference instruments nor are they on the same height from the ground.
404 Therefore, IC should be done for more than 10 minutes in atmospherically homogenous areas to
405 achieve better harmonization between the mobile and reference instruments.

407 **MA200**

408 For the Loški Potok campaign, the 2 backpacks were equipped with both an AE51 and the new
409 generation, 5-wavelength microAethalometer MA200. This served as a field performance test of
410 the MA200 in extreme conditions (winter) in terms of unit-to-unit variability, IC against AE51
411 and finally, IC against the AE33.

412 *Unit-to-unit*

413 Figure 7 shows the results of the RMA regression between the two units of MA200 for all
414 wavelengths (uncorrected data). The two units have good agreement with each other except for
415 the blue channel (470 nm) where the R^2 is 0.57 which could be due to the noise from the light
416 source. In this experiment, the MA200 was compared against AE51. The MA200_75 was in the
417 same backpack as the AE51_S5, both downstream of the silica gel dryer. The same is true for the
418 MA200_69 and AE51_S6. Figure 7 shows the correlation between the MA200 (880 nm channel)
419 and AE51 during MM at Loški Potok with R^2 and slopes higher than 0.90. The real-world eBC
420 mass concentrations measured by the MA200 at 880 nm correlates well with the measurements
421 from the AE51.

422

423 *Against reference instrument*

424 Since the AE33 and the MA200 do not have the same number of wavelengths, the MA200 was
425 compared to only 5 channels of the AE33 which are listed in Table 3. In addition, since the
426 software versions of the MA200s still did not include the filter-loading correction algorithm, the
427 data were post-processed for the filter-loading effect with the offline method of Virkkula et al.
428 (2007) as explained in the Methods section. Fixed k (compensation; from here on k_{MA200})
429 parameters characterizing the loading effect were determined from the measurement data,
430 separately for each wavelength. k_{MA200} values used for loading effect compensation are listed
431 in Table 3. The data from AE33 were already corrected online.

432 The IC of measurements obtained with the MA200 and the reference AE33 for five different
433 wavelengths (UV, Blue, Green, Red and IR) at two stations in Loški Potok, showed a more
434 pronounced filter-loading effect in MA200 instruments as compared to the AE51. Moreover,
435 higher loading effect is seen for the lower wavelengths (Fig. 9).

436 Correcting for the filter-loading effect in MA200 makes a significant improvement of the
437 correlation against AE33 for all wavelengths, particularly for lower wavelengths. Less loading
438 effect was observed for the rural background station, due to low filter attenuation at the beginning
439 of each run. The slope between datasets for the UV wavelength increased after compensation
440 from >0.29 to >0.78 , with an increase of the R^2 from >0.80 to >0.93 . For the IR wavelength, the
441 improvement of correlation with corrected data was the smallest, yet with an important increase
442 of the slope at the rural village site from >0.74 to >0.87 . The loading parameter k_{MA200} differs
443 from the one featured in other Aethalometer instruments due to a completely different filter
444 material – it is not fibrous but rather a membrane. Loading effect for Teflon coated glass fiber
445 filters is mostly known, whereas this is one of the first studies, where the loading effect for

446 MA200 instruments is evaluated. As observed during the Loški Potok campaign, MA200
447 instrument experiences much stronger loading effect than the AE51. Therefore, loading
448 compensation should be applied to the raw data especially with high filter loading and when
449 AAE is calculated from the multi-wavelength data, since stronger loading effect in low
450 wavelengths leads to biased values of AAE.

451

452 **CONCLUSIONS**

453

454 microAethalometers, despite being widely-used for mobile measurements of eBC mass
455 concentrations, have hardly been assessed in real-world environments. In this study, two models
456 (AE51 and MA200) were assessed to determine how well they perform in the field during mobile
457 measurements when compared against a reference absorption photometer.

458 Data from three mobile measurement campaigns were used in this study: a highly urbanized
459 megacity during the summer (Manila, Philippines), a touristic but urbanized city in winter (Rome,
460 Italy), and a rural village in winter (Loški Potok, Slovenia). The assessment was in terms of its
461 comparability against another unit of the same model, and a reference absorption photometer.

462 The AE51 showed a unit-to-unit variability of 5% in urban areas, and lower in rural areas. This
463 was also reflected by the intercomparison (IC) against the reference instruments, where R^2 are
464 higher and slopes closer to unity for IC's done at the rural background, rural village, background,
465 and urban background locations than at urbanstreet and urban street canyon. The
466 intercomparability of the AE51 to the reference instruments showed dependence on the location

467 of the IC, filter-loading effect correction, and duration of IC, but not on the time of day when the
468 IC was done. This implies that the AE51 performs well in different environments and can capture
469 the variabilities of the eBC mass concentrations within the day which are caused by the varying
470 strength of sources and meteorological conditions. Also, for mobile measurements,
471 harmonization of the AE51 with the reference instruments should be done in an atmospherically
472 homogenous environment at longer duration (10-30 minutes) where the spatial variabilities are
473 much lesser than at the street side.

474 In addition, the field performance of a newer microAethalometer with 5-wavelengths (MA200)
475 was also assessed in terms of its intercomparability against another MA200, the AE51, and a 7-
476 wavelength Aethalometer. The MA200 has low unit-to-unit variability (~2%) across all
477 wavelengths as determined at the rural sites. The variability is greater at the rural village,
478 especially at lower wavelengths (UV = 15-22%, blue = 12-18%, green = 11-15%, red = 0-8%,
479 and red = 0-3%). The MA200s (880 nm channel) showed good agreement with the AE51s. In the
480 environments with similar conditions as in Loški Potok, where biomass burning is an important
481 source of eBC, correcting the raw data for filter-loading effect is of exceptional importance for
482 reliable data interpretation. In the study in Loški Potok, Slovenia, compensation parameter k was
483 determined for each wavelength and applied with the post-processing method (Virkkula et al.,
484 2007) assuming a constant k value for the whole winter measurement period. This assumption

485 can introduce systematic errors in the data, which can be avoided by determining highly changing
486 k values using the online algorithms (e. g. Drinovec et al., 2015; Drinovec et al., 2017). In
487 addition, further laboratory, as well as real-world experiments, are necessary to obtain a range of
488 k values characteristic for MA200 instrument.

489 Further investigations on the field performance of the MA200, similar to the ones done for the
490 AE51 in this study, especially in other environmental conditions, is necessary to fully assess its
491 capabilities in reliably capturing the spatial variability of eBC mass concentrations.

492 Loading effect on eBC measurements done at urban areas in the context of mobile
493 measurements proved challenging to detect as the relationship between eBC and ATN showed
494 dependency on the route. Dedicated experiments have to be done to develop a method capable of
495 detecting loading effect and apply an offline correction on the eBC measured through mobile
496 measurements.

497 Finally, the AE51 and the MA200 are reliable instruments that can perform well in drastically
498 different environments. Fully understanding the how these instruments perform in the field can
499 increase our confidence in the data gathered through mobile measurements which are necessary
500 for accurate personal exposure estimates and air pollution mapping.

501

502 **ACKNOWLEDGMENTS**

503

504 Authors from the LP-2018 campaign would like to thank the following: Slovenian Research
505 Agency, Municipality of Loški Potok, Environment Agency of Republic of Slovenia, and the
506 grant from COST Action CA16109 COLOSSAL. Authors from the CARE-2017 campaign
507 acknowledge: Antonio di Ianni, Dr. Luca di Liberto, and Dr. Gian Paolo Gobbi. Authors from the
508 MACE-2015 campaign acknowledge: Dr. James Simpas, Dr. Mylene Cayetano, Dr. Edgar Vallar,
509 Dr. Wolfram Birmili, Dr. Simonas Kecorius, Everlyn Tamayo, and Leizel Madueño. We
510 acknowledge the efforts of the all the people and institutes involved in the three campaigns.

511

512 **DISCLAIMER**

513

514 Reference to any companies or specific commercial products does not constitute endorsement
515 by the authors and their affiliations.

516

517 **APPENDICES**

518 *Appendix A*

519 *Assessment of FLE in Manila and Rome datasets*

520

521 Here, the details of the BC(ATN) approach performed to assess the FLE for the Manila and
522 Rome datasets are presented. For the other two approaches, the information is provided in the
523 Methods section of the main manuscript. They require the same data preparation as below.

524 For the BC(ATN), the 1-s raw data from the AE51 was compiled and given IDs pertaining to
525 each mobile measurement period (1 completion of the route = 1 “run”). As ATN should start at 0

526 when the new filter is inserted at the start of each run, we deducted the initial value for each run
527 (ATN at $t=0$, ATN_0) from the ATN values during the measurements: the corrected ATN
528 (ATN_{corr}) was calculated as the difference between the ATN measured at the next point in time
529 ($ATN_{t=1}$) and ATN_0 . ATN does not start at 0 when the filter is inserted due to ununiform
530 illumination of the sample and reference spots in the filter photometers. Then, the BC mass
531 concentrations were binned according to ATN_{corr} with intervals of 1 ATN. The BC mass
532 concentration (with mean and median concentration per bin) was then plotted as a function of the
533 ATN_{corr} . To detect the loading effect, a linear fit of both the mean and median values of the BC
534 mass concentrations was performed over whole ATN range. Another experiment was to fit the
535 mean and median values over only a specific ATN range. Drinovec et al. (2015) did not include
536 the lowest and largest ATN values in the fitting due to low frequency of BC measurements at
537 those values. In this study, we fitted the BC(ATN) plots only for BC values below the 95th
538 percentile of the ATN. If the fit featured a negative slope, and BC is decreasing with increasing
539 ATN, we interpreted this as the presence of the loading effect, that is the dependence of BC on
540 ATN rather than just on the change of ATN in time. Normally, the loading parameter to correct
541 the AE51 raw concentrations can be derived from the slope and intercept of the regression line.

542 *Investigation of the filter-loading effect on AE51 data from Manila and Rome campaigns*

543 In this section, the loading effect on the AE51 measurements from the Manila and Rome
544 campaign was investigated following three approaches presented in the manuscript. The
545 prerequisites for applying the filter loading effect correction using a loading parameter derived
546 from a single period of analysis are having sufficient measurement data points and homogenous
547 sources of particles.

548 For the BC(ATN) approach, again, the whole datasets (not just the data points during the
549 intercomparison (IC) period) were used for a complete loading effect assessment. The results are
550 shown in Figure A1. The blue and red dots represent the median and mean eBC mass
551 concentration per ATN bin, respectively, while the error bars represent the standard deviation. To
552 detect if there is a loading effect, a linear fit was performed over the whole ATN range and the
553 ratio of the slope and the intercept represents the loading parameter k . If the slope of the fit is
554 negative and its absolute value is greater than 0, then there is a loading effect.

555 However, Fig. A1 shows a positive slope which could be a statistical artifact (Drinovec et al.,
556 2015). Hence, to determine an appropriate range of ATN for fitting, the frequency distribution of
557 the number of measurements per ATN bin was plotted and are shown in Figure A2. From here,
558 the ATN range for fitting was adjusted to include only everything below the 95th percentile of
559 the ATN as the frequency of the measurement decreases towards higher ATN.

560 The BC(ATN) was plotted again, this time fitting within the range of ATN reflecting 0-95th
561 percentile of the data (Figure A3). For the Taft and Rome routes, the slopes are still positive.
562 Refitting with ATN range down to < 85th percentile still resulted to positive slopes (not shown).
563 For the Katipunan route, fitting the median values for an ATN range covering up to 95th and up
564 to 85th percentile of the data gave negative slopes which could indicate a loading effect.
565 However, from these plots, it can be observed that the dependency of BC on ATN seem to be
566 affected by the route itself.

567 This indicates that there are clearly different sources throughout the route, which means probably
568 different aerosol compositions. According to Drinovec et al., 2015, when the frequency
569 distribution is not unimodal, this is indicative of different periods or in this case “area types”
570 which could mean different source compositions and should be analyzed separately. However, as
571 can be observed from Fig. A2, the number of measurements per ATN bin are not enough to
572 derive loading parameters that are dependent on specific parts of the route.

573 The results of the deviation (ATN) approach are shown in Fig. A4. Figures on the left panel are
574 AE51/Ref ratios vs AE51_ATN and on the right panels are AE51-Ref vs. AE51_ATN. The plots
575 for the Katipunan dataset show inconclusive results with negative slope for the ratio vs ATN and
576 positive for the difference vs ATN. The Taft dataset, on the other hand, show negative slopes for
577 both, indicating a possible FLE. However, it must be noted that the number of datapoints used for

578 this analysis is quite low (222 for Katipunan, and 383 for Taft) with the IC periods of less than 5
579 minutes each. This is evident in the figures with ratios much greater than 1 and large differences.
580 As mentioned in the manuscript, the IC periods occurred in the middle of a run, hence, this
581 analysis do not cover a uniform dataset over the whole ATN range. Deriving a loading parameter
582 from this analysis would also be misleading as we do not expect that the loading parameter in one
583 point in space would be representative of the rest of the route in inhomogeneous atmospheres.
584 The loading parameter depends on the whole collected sample on the spot.

585 As a last attempt, a fixed k value of 0.005 was used to correct the Manila and Rome datasets (as
586 was done for the Loški Potok AE51 data). This value represents the loading effect of a diesel
587 exhaust dominated atmosphere as well as from fresh ambient wood burning (Drinovec et al.,
588 2017). The corrected eBC was then plotted against the uncorrected eBC and is show in Fig. A5.
589 This shows that the correction did not change the eBC measurements substantially (6%, 8%, and
590 3% overall differences between corrected and uncorrected measurements for the Katipunan, Taft,
591 and Rome routes, respectively). As a result, no filter-loading effect correction was applied on the
592 Manila and Rome datasets. As for the Manila dataset, the discrepancy between the mobile AE51
593 and the reference instrument is due to the high variabilities of different factors (wind, sources,
594 etc.) characteristic of an urban area.

595

596

597 **Appendix B**

598 <Tables of regression results>

599

600

ACCEPTED MANUSCRIPT

601 **REFERENCES**

602

603 Alas, H.D., Müller, T., Birmili, W., Kecorius, S., Cambaliza, M.O., Simpas, J.B.B., Cayetano, M., Weinhold, K.,
604 Vallar, E., Galvez, M.C. and Wiedensohler, A. (2018). Spatial characterization of black carbon mass
605 concentration in the atmosphere of a southeast asian megacity: An air quality case study for metro manila,
606 philippines. *Aerosol Air Qual Res* 18: 2301-2317.

607 Alas, H.D.C., Weinhold, K., Costabile, F., Di Ianni, A., Müller, T., Pfeifer, S., Di Liberto, L., Turner, J.R. and
608 Wiedensohler, A. (2019). Methodology for high quality mobile measurement with focus on black carbon
609 and particle mass concentrations. *Atmospheric Measurement Techniques Discussions*: 1-27.

610 Apte, J.S., Kirchstetter, T.W., Reich, A.H., Deshpande, S.J., Kaushik, G., Chel, A., Marshall, J.D. and Nazaroff,
611 W.W. (2011). Concentrations of fine, ultrafine, and black carbon particles in auto-rickshaws in new delhi,
612 india. *Atmospheric Environment* 45: 4470-4480.

613 Bates, D., Mächler, M., Bolker, B. and Walker, S. (2015). Fitting linear mixed-effects models using lme4. *Journal of*
614 *Statistical Software* 67.

615 Birmili, W., Rehn, J., Vogel, A., Boehlke, C., Weber, K. and Rasch, F. (2013). Micro-scale variability of urban
616 particle number and mass concentrations in leipzig, germany. *Meteorologische Zeitschrift* 22: 155-165.

617 Cai, J., Yan, B., Kinney, P.L., Perzanowski, M.S., Jung, K.H., Li, T., Xiu, G., Zhang, D., Olivo, C., Ross, J., Miller,
618 R.L. and Chillrud, S.N. (2013). Optimization approaches to ameliorate humidity and vibration related issues
619 using the microaeth black carbon monitor for personal exposure measurement. *Aerosol Sci Tech* 47: 1196-
620 1204.

621 Cheng, Y.-H. and Lin, M.-H. (2013). Real-time performance of the microaeth® ae51 and the effects of aerosol
622 loading on its measurement results at a traffic site. *Aerosol and Air Quality Research* 13: 1853-1863.

623 Costabile, F., Alas, H., Aufderheide, M., Avino, P., Amato, F., Argentini, S., Barnaba, F., Berico, M., Bernardoni, V.,
624 Biondi, R., Casasanta, G., Ciampichetti, S., Calzolari, G., Canepari, S., Conidi, A., Cordelli, E., Di Ianni, A.,
625 Di Liberto, L., Facchini, M., Facci, A., Frasca, D., Gilardoni, S., Grollino, M., Gualtieri, M., Lucarelli, F.,
626 Malaguti, A., Manigrasso, M., Montagnoli, M., Nava, S., Perrino, C., Padoan, E., Petenko, I., Querol, X.,
627 Simonetti, G., Tranfo, G., Ubertini, S., Valli, G., Valentini, S., Vecchi, R., Volpi, F., Weinhold, K.,
628 Wiedensohler, A., Zanini, G., Gobbi, G. and Petralia, E. (2017). First results of the “carbonaceous aerosol
629 in rome and environs (care)” experiment: Beyond current standards for pm10. *Atmosphere* 8.

630 Dons, E., Temmerman, P., Van Poppel, M., Bellemans, T., Wets, G. and Int Panis, L. (2013). Street characteristics
631 and traffic factors determining road users' exposure to black carbon. *Sci Total Environ* 447: 72-79.

632 Drinovec, L., Gregorič, A., Zotter, P., Wolf, R., Bruns, E.A., Prévôt, A.S.H., Petit, J.-E., Favez, O., Sciare, J., Arnold,
633 I.J., Chakrabarty, R.K., Moosmüller, H., Filep, A. and Močnik, G. (2017). The filter-loading effect by
634 ambient aerosols in filter absorption photometers depends on the coating of the sampled particles.
635 *Atmospheric Measurement Techniques* 10: 1043-1059.

636 Drinovec, L., Močnik, G., Zotter, P., Prévôt, A.S.H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T.,
637 Wiedensohler, A. and Hansen, A.D.A. (2015). The "dual-spot" aethalometer: An improved measurement of

638 aerosol black carbon with real-time loading compensation. *Atmospheric Measurement Techniques* 8: 1965-
639 1979.

640 Düsing, S., Wehner, B., Müller, T., Stöcker, A. and Wiedensohler, A. (2019). The effect of rapid relative humidity
641 changes on fast filter-based aerosol-particle light-absorption measurements: Uncertainties and correction
642 schemes. *Atmospheric Measurement Techniques* 12: 5879-5895.

643 Glojek, K., Gregorič, A. and Ogrin, M. (2018). Black carbon air pollution - case study of loski potok. *Dela*: 25-43.

644 Good, N., Molter, A., Peel, J.L. and Volckens, J. (2017). An accurate filter loading correction is essential for
645 assessing personal exposure to black carbon using an aethalometer. *J Expo Sci Environ Epidemiol* 27: 409-
646 416.

647 Kecorius, S., Madueno, L., Londahl, J., Vallar, E., Galvez, M.C., Idolor, L.F., Gonzaga-Cayetano, M., Muller, T.,
648 Birmili, W. and Wiedensohler, A. (2019). Respiratory tract deposition of inhaled roadside ultrafine
649 refractory particles in a polluted megacity of south-east asia. *Sci Total Environ* 663: 265-274.

650 Kecorius, S., Madueño, L., Vallar, E., Alas, H., Betito, G., Birmili, W., Cambaliza, M.O., Catipay, G., Gonzaga-
651 Cayetano, M., Galvez, M.C., Lorenzo, G., Müller, T., Simpas, J.B., Tamayo, E.G. and Wiedensohler, A.
652 (2017). Aerosol particle mixing state, refractory particle number size distributions and emission factors in a
653 polluted urban environment: Case study of metro manila, philippines. *Atmospheric Environment* 170: 169-
654 183.

655 Kecorius, S., Tamayo, E.G., Galvez, M.C., Madueño, L., Betito, G., Gonzaga-Cayetano, M., Vallar, E. and
656 Wiedensohler, A. (2018). Activity pattern of school/university tenants and their family members in metro
657 manila – philippines. *Aerosol and Air Quality Research* 18: 2412-2419.

658 Legendre, P. (2018). *Lmodel2: Model ii regression*, 1.7-3 ed.

659 Madueño, Kecorius, Birmili, Müller, Simpas, Vallar, Galvez, Cayetano and Wiedensohler (2019). Aerosol particle
660 and black carbon emission factors of vehicular fleet in manila, philippines. *Atmosphere* 10.

661 Masey, N., Ezani, E., Gillespie, J., Sutherland, F., Lin, C., Hamilton, S., Heal, M.R. and Beverland, I.J. (2020).
662 Consistency of urban background black carbon concentration measurements by portable ae51 and reference
663 ae22 aethalometers: Effect of corrections for filter loading. *Aerosol and Air Quality Research* 20: 329-340.

664 Park, S.S., Hansen, A.D.A. and Cho, S.Y. (2010). Measurement of real time black carbon for investigating spot
665 loading effects of aethalometer data. *Atmospheric Environment* 44: 1449-1455.

666 Peters, J., Van den Bossche, J., Reggente, M., Van Poppel, M., De Baets, B. and Theunis, J. (2014a). Cyclist
667 exposure to ufp and bc on urban routes in antwerp, belgium. *Atmos Environ* 92: 31-43.

668 Peters, J., Van den Bossche, J., Reggente, M., Van Poppel, M., De Baets, B. and Theunis, J. (2014b). Cyclist
669 exposure to ufp and bc on urban routes in antwerp, belgium. *Atmospheric Environment* 92: 31-43.

670 Petzold, A., Ogren, J.A., Fiebig, M., Laj, P., Li, S.M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G.,
671 Sugimoto, N., Wehrli, C., Wiedensohler, A. and Zhang, X.Y. (2013). Recommendations for reporting
672 "black carbon" measurements. *Atmos Chem Phys* 13: 8365-8379.

673 Petzold, A., Schloesser, H., Sheridan, P.J., Arnott, W.P., Ogren, J.A. and Virkkula, A. (2005). Evaluation of
674 multiangle absorption photometry for measuring aerosol light absorption. *Aerosol Science and Technology*
675 39: 40-51.

676 R Core Team (2019). R: A language and environment for statistical computing.

677 Rakowska, A., Wong, K.C., Townsend, T., Chan, K.L., Westerdahl, D., Ng, S., Močnik, G., Drinovec, L. and Ning,
678 Z. (2014). Impact of traffic volume and composition on the air quality and pedestrian exposure in urban
679 street canyon. *Atmos Environ* 98: 260-270.

680 Segura, S., Estellés, V., Titos, G., Lyamani, H., Utrillas, M.P., Zotter, P., Prévôt, A.S.H., Močnik, G., Alados-
681 Arboledas, L. and Martínez-Lozano, J.A. (2014). Determination and analysis of in situ spectral aerosol
682 optical properties by a multi-instrumental approach. *Atmospheric Measurement Techniques* 7: 2373-2387.

683 Van den Bossche, J., Peters, J., Verwaeren, J., Botteldooren, D., Theunis, J. and De Baets, B. (2015). Mobile
684 monitoring for mapping spatial variation in urban air quality: Development and validation of a methodology
685 based on an extensive dataset. *Atmospheric Environment* 105: 148-161.

686 Van den Bossche, J., Theunis, J., Elen, B., Peters, J., Botteldooren, D. and De Baets, B. (2016). Opportunistic mobile
687 air pollution monitoring: A case study with city wardens in antwerp. *Atmospheric Environment* 141: 408-
688 421.

689 Viana, M., Rivas, I., Reche, C., Fonseca, A.S., Pérez, N., Querol, X., Alastuey, A., Álvarez-Pedrerol, M. and Sunyer,
690 J. (2015). Field comparison of portable and stationary instruments for outdoor urban air exposure
691 assessments. *Atmospheric Environment* 123: 220-228.

692 Virkkula, A., Makela, T., Hillamo, R., Yli-Tuomi, T., Hirsikko, A., Hameri, K. and Koponen, I.K. (2007). A simple
693 procedure for correcting loading effects of aethalometer data. *J Air Waste Manag Assoc* 57: 1214-1222.

694 Wickham, H. (2016). *Ggplot2: Elegant graphics for data analysis*, Springer-Verlag New York.

695 Wickham, H., François, R., Henry, L. and Müller, K. (2018). *Dplyr: A grammar of data manipulation*. , R package
696 version
697 0.8.3 ed.
698
699

700 **Table 1.** Ambient conditions during each mobile measurement campaign. Mean meteorological
 701 parameters are given with standard deviation in parenthesis.

Campaign conditions					Meteorological conditions		
Study area	Description	Altitude [m]	Period	Sources	T [°C]	RH [%]	P [hPa]
<i>Manila, Philippines^a</i>	Highly urbanized, megacity	5	Summer	Traffic	29.9 (2.8)	66.0 (12.2)	1012.2 (2.6)
<i>Rome, Italy^b</i>	Highly touristic and urbanized	21	Winter	Traffic	11.3 (3.4)	75 (14)	1016 (780)
<i>Loški Potok, Slovenia^c</i>	Rural	715 - 815	Winter	Wood burning	0.7 (4.1)	89.2 (7.5)	924.4 (11.03)

702 ^a MACE-2015; Alas et al., 2018

703 ^b CARE-2017; Costabile et al., 2017, Alas et al., 2019

704 ^c LP-2017; Glojek, Gregorič, Ogrin, 2018; meteorological information during the campaign
 705 obtained from a station at 775 masl

706

707 **Table 2.** Intercomparison parameters for each campaign

Study area and routes	Instruments		Intercomparison parameters		
	Mobile	Reference	Location	Time ^a	Duration (min)
<i>Manila, Philippines</i>					
Katipunan Route	AE51	MAAP	Urban street	nn & pm	< 5
Taft Route	AE51	MAAP	Urban background	nn & pm	< 5
Taft Route	AE51	MAAP	Urban street canyon	nn & pm	< 5
<i>Rome, Italy</i>					
Rome city route	AE51(2x)	MAAP	Urban background	am, nn, & pm	30
<i>Loški Potok, Slovenia</i>					
Village route	AE51(2x)	AE33	Rural village	am, nn, &	20
	MA200 (2x)	AE33	Rural background	pm	10

708 ^a “am” – morning; “nn” – noon to afternoon; “pm” – evening

709

710 **Table 3.** Channels (wavelengths) used to compare MA200 (5 wavelengths) measurements with
711 AE33 (7 wavelengths) with the loading parameter values derived for each wavelength of the
712 MA200 denoted as k_{MA200} .

	MA200 (nm)	AE33 (nm)	k_{MA200}
UV	375	370	0.03
Blue	470	470	0.024
Green	528	520	0.0215
Red	625	660	0.0156
IR	880	880	0.015

713

714

ACCEPTED MANUSCRIPT

715 Table A1. Descriptive summary of the instruments used in this study.

716

Instrument	Platform	Operating principle	Light source wavelength	Time resolution
AE51	Mobile	Attenuation of light by particle loaded filter	880 nm	10 s
MA200	Mobile	Attenuation of light by particle loaded filter	375nm, 470 nm, 528 nm, 625 nm, 880 nm	10 s
MAAP	Fixed	Absorption of light by particle loaded filter. Multiangle absorption photometers allows for the use of the radiative transfer scheme to remove scattering effects	637 nm	60 s
AE33	Fixed	Attenuation of light by particle loaded filter	370, 470, 520, 590, 660, 880 and 950 nm	60 s

717

718

719 Table A2. Summary of the IC periods for each route.

IC locations	# of IC periods	Total IC period in minutes	# of filter changes
<i>Manila campaign</i>			
Katipunan Route			
(urban street)	32	222	
(urban background)	73	128	77
Taft Route	86	383	34
<i>Rome campaign</i>			
Rome city route	41	1116	77
<i>Loški Potok campaign</i>			
Village route			
(rural village)	102	2287	
(rural background)	107	1166	107

ACCEPTED MANUSCRIPT

720 Table B1. Regression results for all AE51 correlations

Instrument	Study Area	IC Location	FLE Correction	Time of IC	Duration of IC	R ²	Slope	Time base	N (no. of IC points)
S5 vs S6	Rome		No			0.821	0.952 ± 0.003	10s	38909
	Loski Potok		No			0.972	1.003 ± 0.001	10s	27521
S5 vs Reference	Manila					0.367	0.879 ± 0.031	60s	1420
	Rome					0.985	1.017 ± 0.005	60s	772
	Loski Potok					0.985	0.808 ± 0.003	60s	1390
S6 vs Reference	Manila					NA	NA	60s	NA
	Rome					0.982	1.013 ± 0.004	60s	1157
	Loski Potok					0.973	0.841 ± 0.003	60s	3006
AE51 vs Reference	Loski Potok	Rural background				0.962	0.876 ± 0.005	60s	2888
		Rural village				0.978	0.826 ± 0.002	60s	1508
	Rome	Urban background				0.983	1.015 ± 0.003	60s	1929
	Manila	Urban background				0.845	0.871 ± 0.013	60s	815
		Urban street				0.545	1.55 ± 0.095	60s	222
	Urban street canyon				0.318	0.746 ± 0.056	60s	383	
S5 vs Reference	Loski Potok	Rural background	No			0.965	0.876 ± 0.008	60s	475
			Yes			0.962	0.916 ± 0.008	60s	475
		Rural village	No			0.986	0.806 ± 0.003	60s	915
	Yes				0.99	0.951 ± 0.003	60s	915	
S6 vs Reference		Rural background	No			0.96	0.876 ± 0.006	60s	1033
	Yes				0.959	0.934 ± 0.006	60s	1033	
		Rural village	No			0.973	0.840 ± 0.003	60s	1973
			Yes				60s	1973	
						0.979	0.962 ± 0.003		

721 Table B1 continued.

Instrument	Study Area	IC Location	FLE Correction	Time of IC	Duration of IC	R ²	Slope	Time base	N (no. of IC points)
AE51 vs Reference	Manila	Urban background	No	NN		0.726	0.905 ± 0.031	60s	325
		Urban street	No			0.409	1.518 ± 0.179	60s	105
		Urban street canyon	No			0.389	0.841 ± 0.078	60s	184
		Urban background	No	PM		0.888	0.862 ± 0.014	60s	490
		Urban street	No			0.709	1.573 ± 0.093	60s	117
		Urban street canyon	No			0.249	0.647 ± 0.081	60s	199
	Rome	Urban background	No	AM		0.988	1.022 ± 0.005	60s	718
			No	NN		0.939	0.941 ± 0.009	60s	747
			No	PM		0.975	1.006 ± 0.009	60s	464
	Loski Potok	Rural background	yes	AM		0.939	0.917 ± 0.010	60s	607
		Rural Village	yes			0.978	0.926 ± 0.004	60s	1202
		Rural background	yes	NN		0.978	0.894 ± 0.007	60s	353
		Rural Village	yes			0.972	0.917 ± 0.006	60s	692
		Rural background	yes	PM		0.954	0.975 ± 0.009	60s	549
Rural Village		yes			0.989	0.976 ± 0.004	60s	994	
S5 vs Reference			No		< 5 minutes	0.845	0.871 ± 0.013	60s	815
			yes		10 minutes	0.962	0.916 ± 0.008	60s	475
			yes		20 minutes	0.991	0.951 ± 0.003	60s	915
			No		30 minutes	0.985	1.017 ± 0.005	60s	772
S6 vs Reference			No		< 5 minutes	NA	NA	NA	NA
			yes		10 minutes	0.959	0.934 ± 0.006	60s	1033
			yes		20 minutes	0.979	0.962 ± 0.003	60s	1973
			No		30 minutes	0.982	1.013 ± 0.004	60s	1157

722

723 Table B2. Regression results for all MA200 correlations.

Instrument	IC Location	FLE Correction	Wavelength	R ²	Slope	Time base	N(no. of IC points)
MA200_75 vs 69			UV	0.876	1.083 ± 0.002	10s	27474
			Blue	0.574	1.121 ± 0.004	10s	
			Green	0.917	1.059 ± 0.002	10s	
			Red	0.929	1.051 ± 0.002	10s	
			IR	0.935	1.034 ± 0.002	10s	
MA200_75 vs AE51_S5			IR	0.917	0.965 ± 0.002	10s	30236
MA200_69 vs AE51_S6			IR	0.911	1.019 ± 0.001	10s	63495
MA200_69 vs Reference	Rural background	no	UV	0.887	0.681 ± 0.008	60s	930
			Blue	0.827	0.794 ± 0.012	60s	
			Green	0.931	0.830 ± 0.008	60s	
			Red	0.943	0.981 ± 0.008	60s	
			IR	0.952	1.026 ± 0.008	60s	
	Rural background	yes	UV	0.900	0.995 ± 0.001	60s	
			Blue	0.829	0.978 ± 0.015	60s	
			Green	0.938	0.976 ± 0.008	60s	
			Red	0.946	1.084 ± 0.009	60s	
			IR	0.952	1.097 ± 0.008	60s	
Rural village	no	UV	0.798	0.369 ± 0.005	60s	1825	
		Blue	0.670	0.480 ± 0.008	60s		
		Green	0.893	0.580 ± 0.005	60s		
		Red	0.922	0.747 ± 0.005	60s		
		IR	0.954	0.837 ± 0.005	60s		
	Rural village	yes	UV	0.935	0.854 ± 0.008		60s
			Blue	0.810	0.842 ± 0.010		60s
			Green	0.964	0.894 ± 0.005		60s
			Red	0.967	0.999 ± 0.005		60s
			IR	0.975	1.017 ± 0.004		60s

724

725

726 Table B2 continued.

Instrument	IC Location	FLE Correction	Wavelength	R ²	Slope	Time base	N(no. of IC)	
MA200_75 vs Reference	Rural background	no	UV	0.947	0.742 ± 0.008	60s	445	
			Blue	0.957	0.814 ± 0.008	60s		
			Green	0.961	0.881 ± 0.008	60s		
			Red	0.963	1.014 ± 0.010	60s		
			IR	0.965	1.013 ± 0.009	60s		
	Rural background	yes	UV	0.941	1.087 ± 0.013	60s		
			Blue	0.957	1.022 ± 0.010	60s		
			Green	0.962	1.044 ± 0.010	60s		
			Red	0.964	1.117 ± 0.010	60s		
			IR	0.964	1.076 ± 0.010	60s		
	Rural village	no	UV	0.828	0.289 ± 0.005	60s	843	
			Blue	0.876	0.401 ± 0.005	60s		
			Green	0.899	0.471 ± 0.006	60s		
			Red	0.925	0.625 ± 0.006	60s		
			IR	0.957	0.743 ± 0.006	60s		
		Rural village	yes	UV	0.936	0.782 ± 0.007		60s
				Blue	0.956	0.821 ± 0.006		60s
				Green	0.963	0.851 ± 0.006		60s
Red				0.968	0.924 ± 0.006	60s		
			IR	0.978	0.971 ± 0.006	60s		

727

728 **Figure Captions**

729 **Fig. 1.** Intercomparison for eBC mass concentrations (10-second median) measured by the AE51
730 S6 and AE51 S5 during the collocated MM in (a) Rome, Italy and (b) Loški Potok, Slovenia.
731 Data were taken from two backpacks with identical instrumentation running simultaneously, side
732 by side for each run, throughout the campaign. RMA regression was used to fit the two
733 measurements. For IC details, see Tables 2, A1, A2, B1, and B2.

734 **Fig. 2.** Intercomparison between the AE51 units against the reference eBC mass concentration
735 measurements at three different study areas: (a) Manila, Philippines, (b) Rome, Italy, and (c)
736 Loški Potok, Slovenia. The time resolution is 1 minute and OLS method forced through the
737 origin was used for fitting. Data were taken from IC done during the mobile measurement runs
738 when the runners were passing by the vicinity of the aerosol container and the backpacks were
739 placed near it. For this, IC periods in all sites were combined into their respective cities. For IC
740 details, see Tables 2, A1, A2, B1, and B2.

741 **Fig. 3.** Intercomparison between the AE51 units against the reference eBC mass concentration
742 measurements as a function of location of IC ((I) rural background, (II) rural village, (III) urban
743 background, (IV) urban street and (V) urban street canyon) per study area ((a) Manila, (b) Rome,
744 and (c) Loški Potok). This is basically the same as in Fig. 2, but now also segregated into
745 different IC locations. For IC details, see Tables 2, A1, A2, B1, and B2.

746 **Fig. 4.** Intercomparison of eBC measurements of AE51 against AE33 at the Loški Potok
747 campaign before (red: uncorrected) and after (blue: corrected) filter-loading effect correction.
748 Data were taken from IC done during the mobile measurement runs when the runners were
749 passing by the vicinity of the aerosol container and the backpacks were placed near it. For IC
750 details, see Tables 2, A1, A2, B1, and B2.

751 **Fig. 5.** Intercomparison between AE51 and reference instrument as a function of IC location
752 (colors) and time of IC: (I) morning, (II) afternoon, and (III) evening. The columns correspond to
753 study area: (a) Manila, (b) Rome, (c) Loški Potok. For IC details, see Tables 2, A1, A2, B1, and
754 B2.

755 **Fig. 6.** Intercomparison between AE51 and reference instrument as a function of duration of IC:
756 (a) <5 minutes, (b) 10 minutes, (c) 20 minutes, and (d) 30 minutes. The colors correspond to the
757 models of AE51. For IC details, see Tables 2, A1, A2, B1, and B2.

758 **Fig. 7.** Intercomparison between the measurements (10-second median, uncorrected) from the
759 MA200_75 and MA200_69 for all wavelengths (a-e) during the collocated MM in Loški Potok,
760 Slovenia. RMA regression was used for fitting. For IC details, see Tables 2, A1, A2, B1, and B2.

761 **Fig. 8.** Intercomparison between the eBC mass concentrations (10-second median; uncorrected)
762 measured by the MA200 units (at 880 nm; uncorrected) against the AE51 units during the

763 collocated MM in Loški Potok, Slovenia. RMA regression was used for fitting. For IC details,
764 see Tables 2, A1, A2, B1, and B2.

765 **Fig. 9.** Intercomparison of the measurements from the each MA200 ((I) and (II) for MA69, (III)
766 and (IV) for MA75, for each IC location (rural village and rural background). The red and blue
767 dots represent uncorrected and corrected measurements. For IC details, see Tables 2, A1, A2, B1,
768 and B2.

769 **Fig. A1.** Binned raw measurements from the AE51 plotted against the attenuation (ATN) for a)
770 Katipunan Route, b) Taft route, and c) Rome city route. Data were taken from the raw AE51
771 measurements (1-s resolution) from all the runs performed in each location (see Table A2),
772 wherein a new filter was used for each run. The duration of a run is 1 hour for the Katipunan and
773 Taft Route, and 2.5 hours for the Rome route. The blue and red dots represent the median and
774 mean eBC mass concentration per bin, respectively, with the error bars as standard deviation.
775 The solid lines are the linear fit for each statistic. The whole ATN range was used for linear
776 fitting.

777 **Fig. A2.** Frequency distributions of the measurements per ATN bin for the a) Katipunan Route, b)
778 Taft route, and c) Rome city route.

779 **Fig. A3.** Same as Fig. A1 but this time the fit was only done on the data below the 95th
780 percentile.

781 **Fig. A4.** Scatter plots of the deviation between the AE51 and reference instruments expressed in
782 ratios (left panels) and differences (right panels) for (a and b) the Katipunan ($n = 222$), (c and d)
783 Taft ($n = 383$), and (e and f) Rome ($n = 1116$) datasets.

784 **Fig. A5.** Correlation between the uncorrected and corrected ($k = 0.005$) eBC mass concentrations
785 for the AE51 measurements along the a) Katipunan route, b) Taft route, and c) Rome route. The
786 color of the dots represents the ATN. The red dashed line represents the 1:1 line, while the solid
787 blue line represents the linear fit.

788

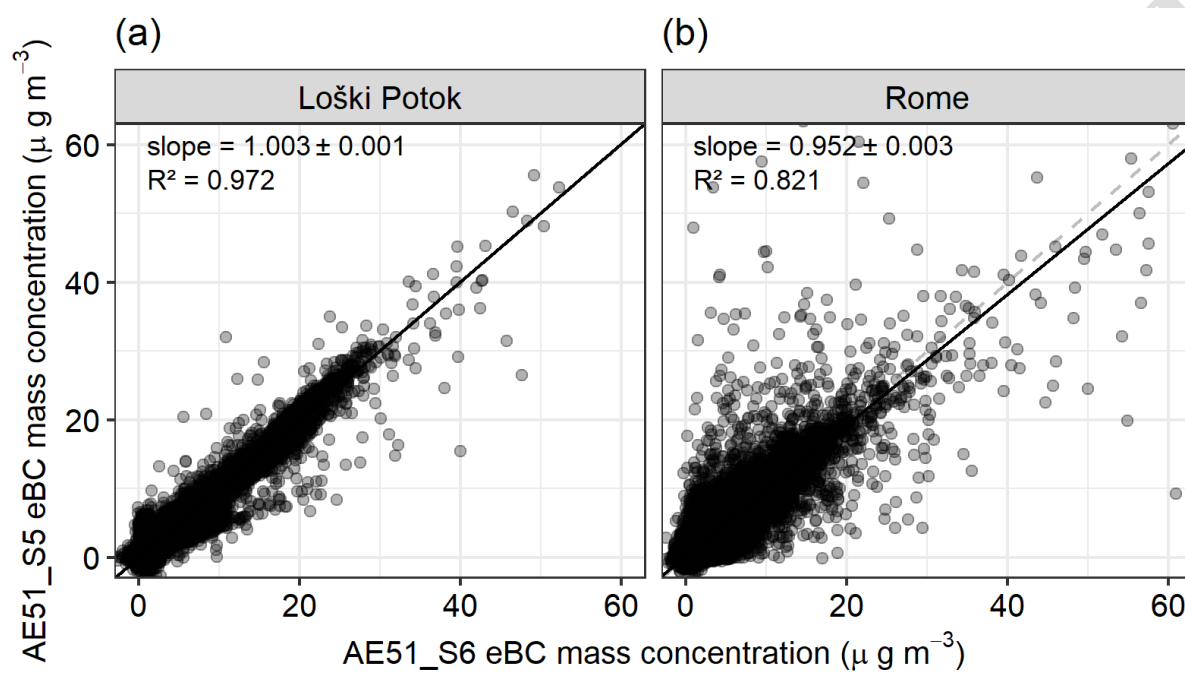
789

ACCEPTED MANUSCRIPT

790

791

792



793

794

795

796

797

798

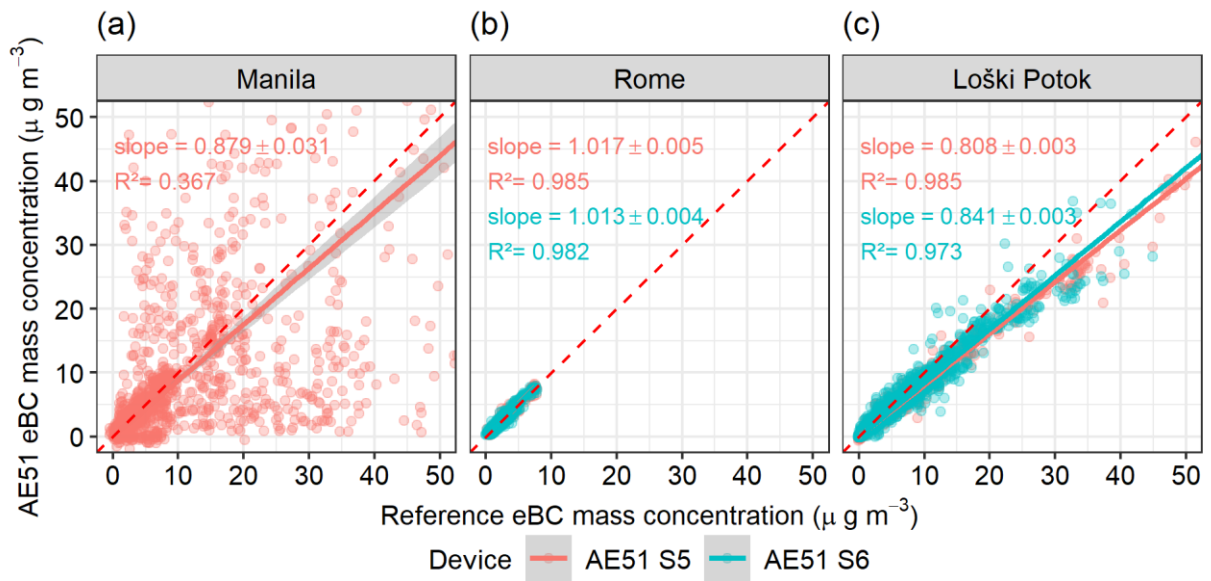
799

800

801

Fig. 1.

802



803

804

805

806

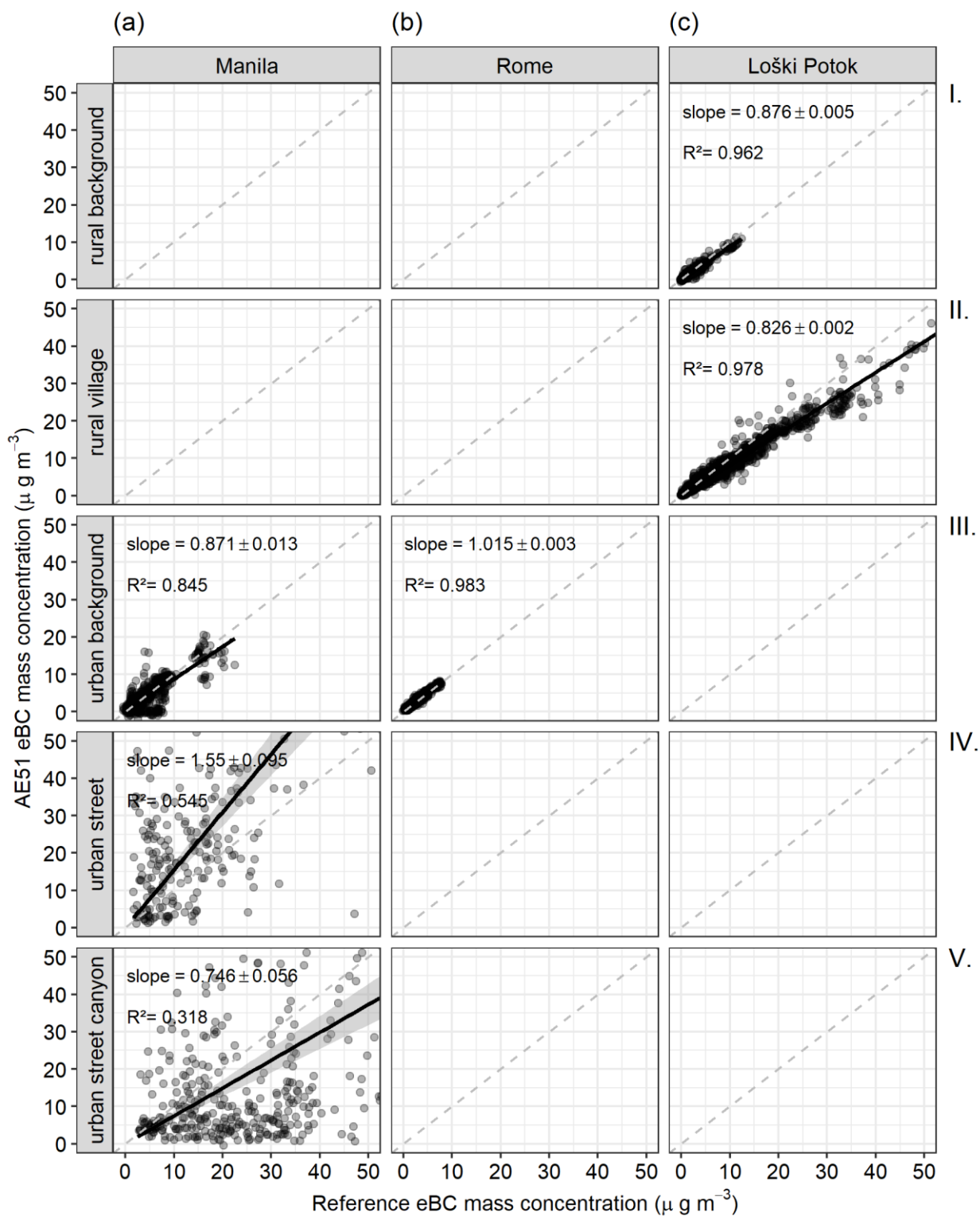
807

808

809

810

Fig. 2

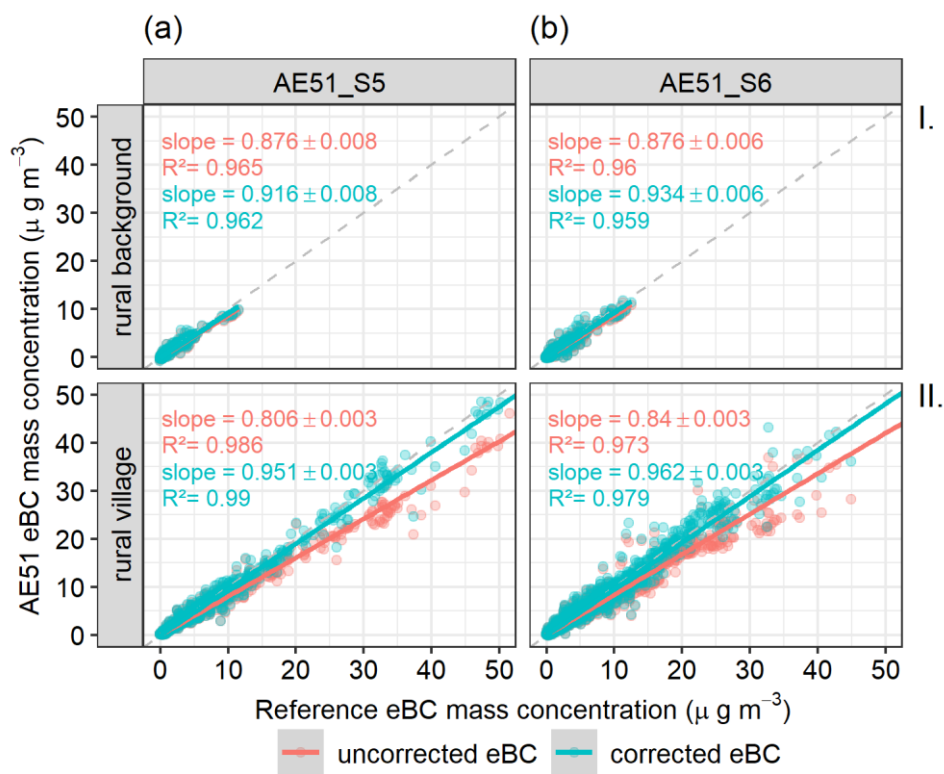


811

812

813

Fig. 3



814

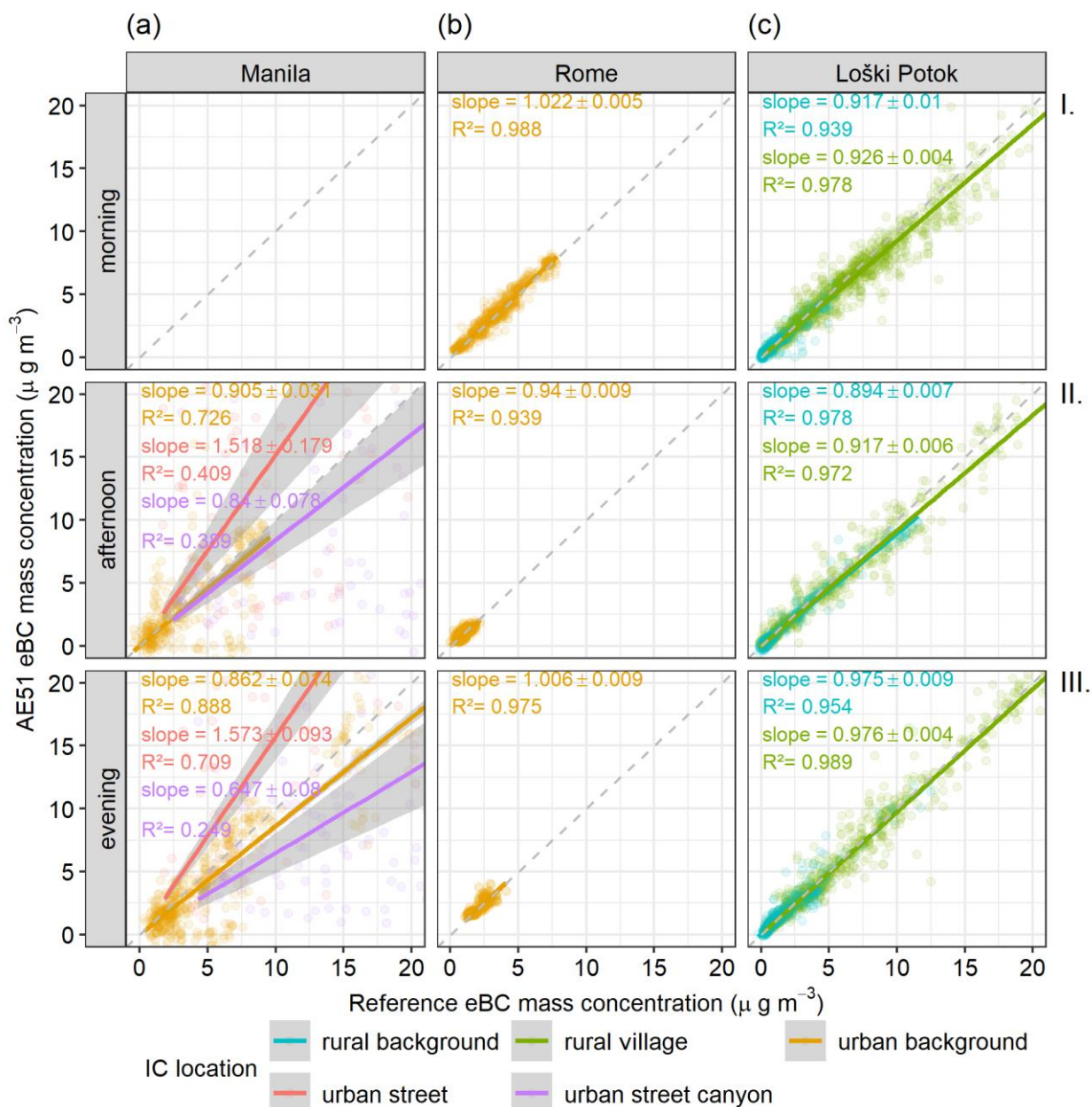
815

816

817

818

Fig. 4



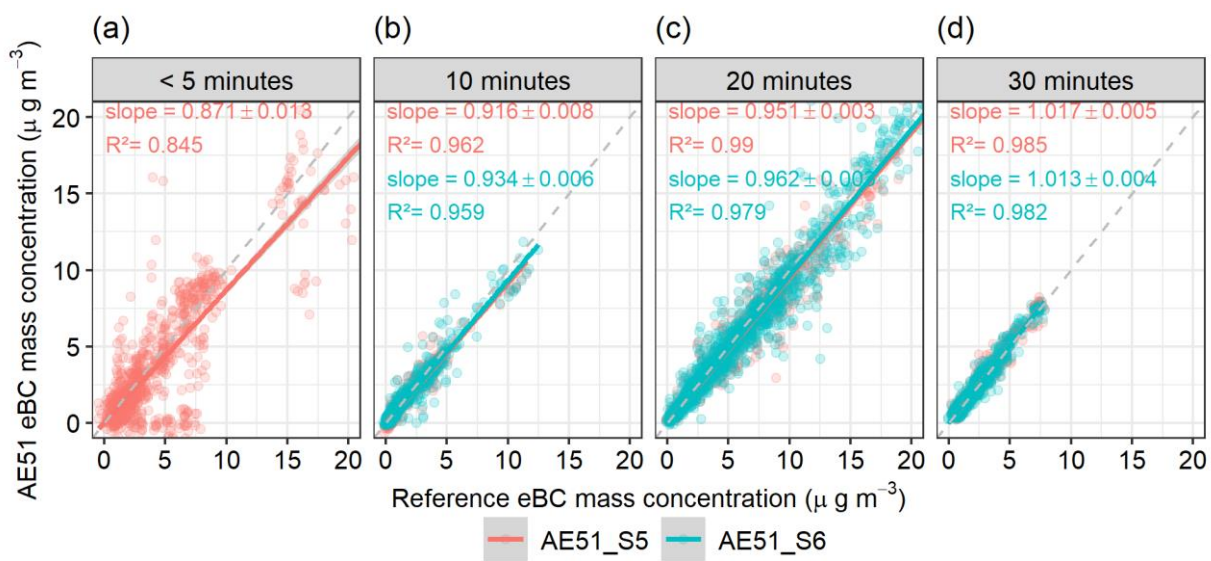
819

820

821

822

Fig. 5



823

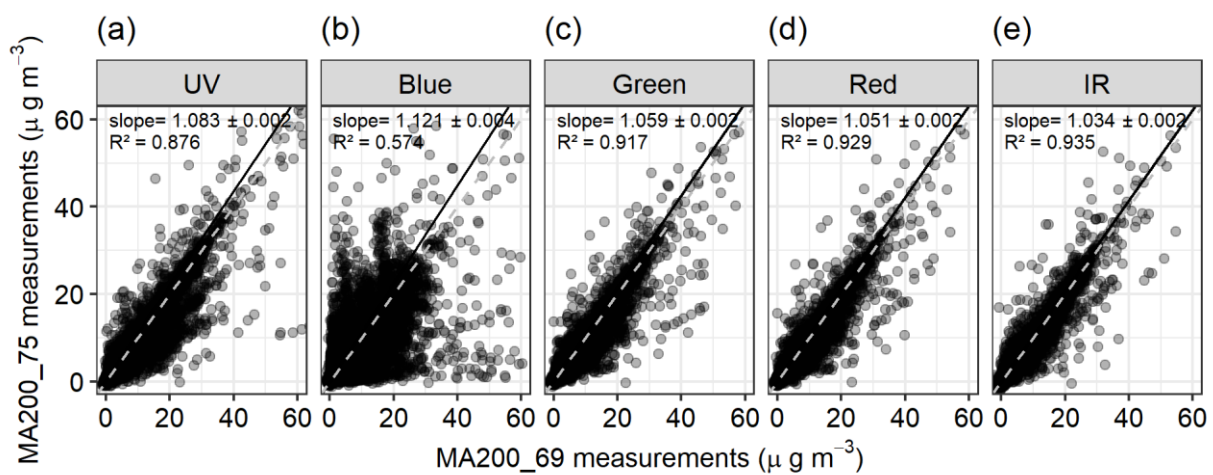
824

825

826

827

Fig. 6



828

829

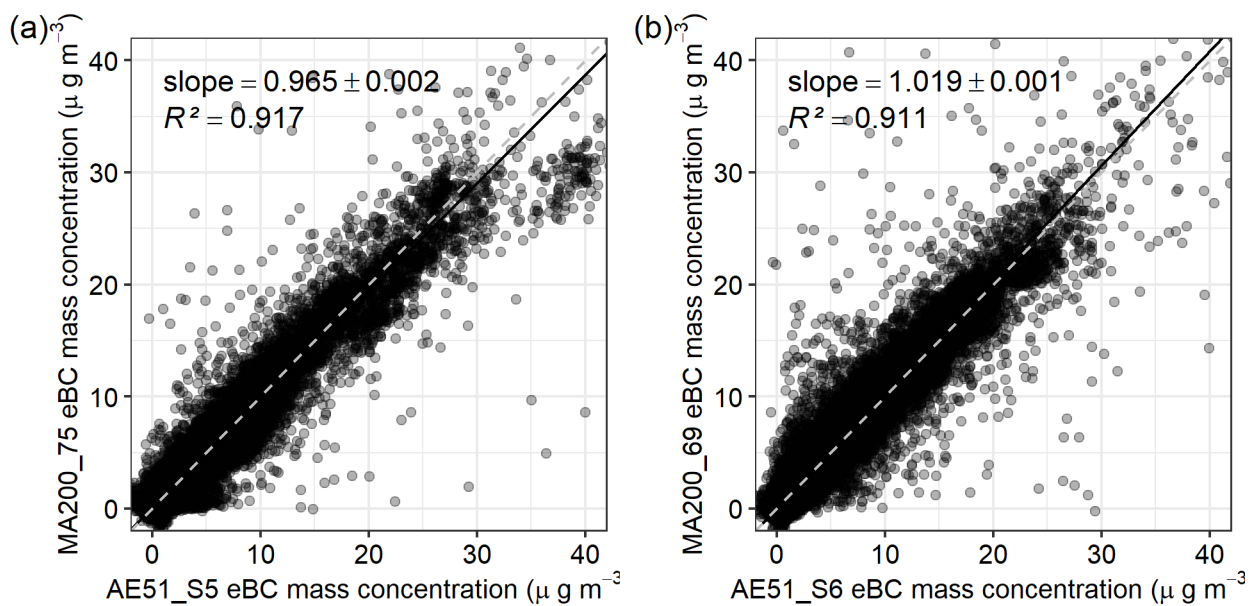
830

831

832

833

Fig. 7



834

835

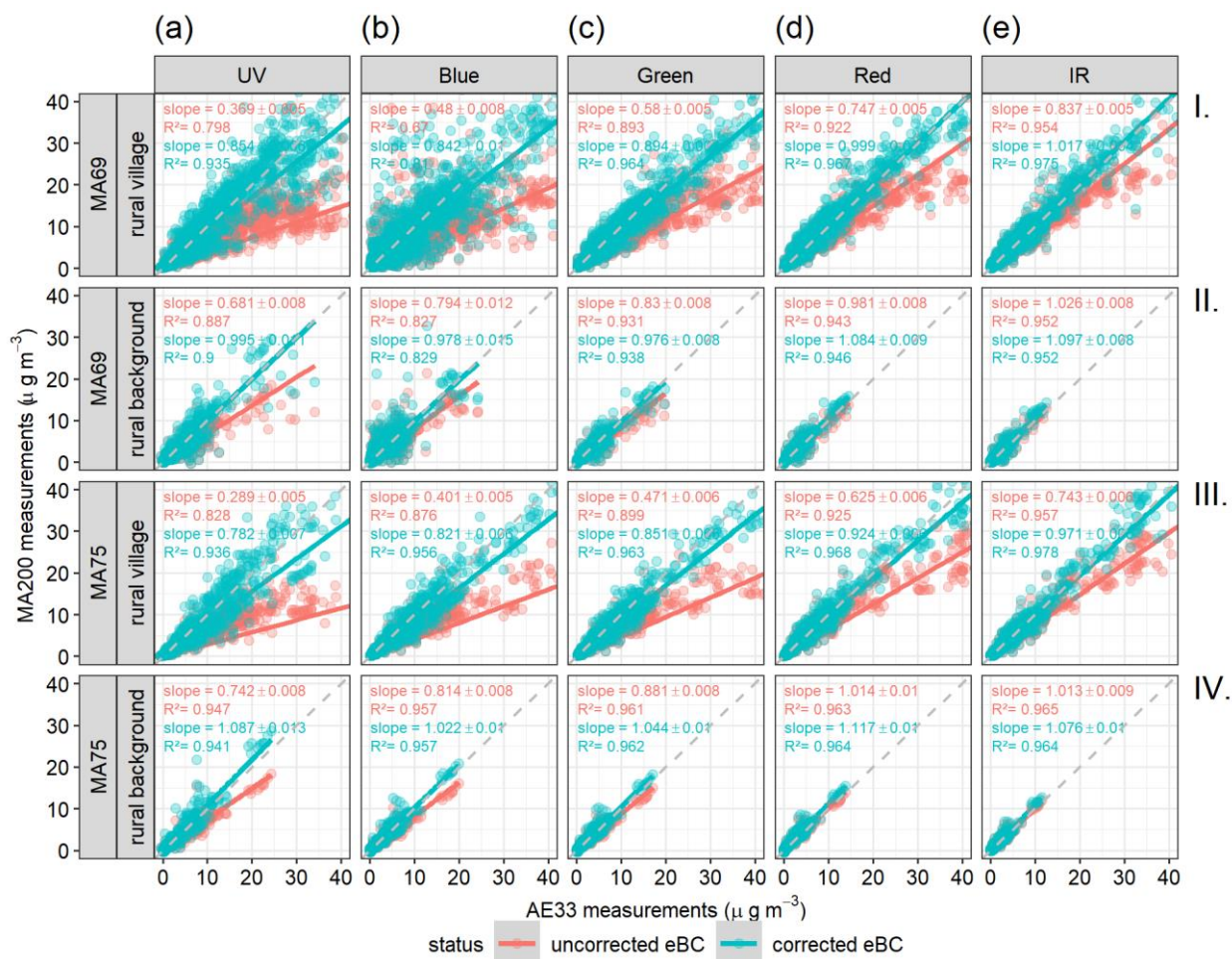
836

837

838

839

Fig. 8



840

841

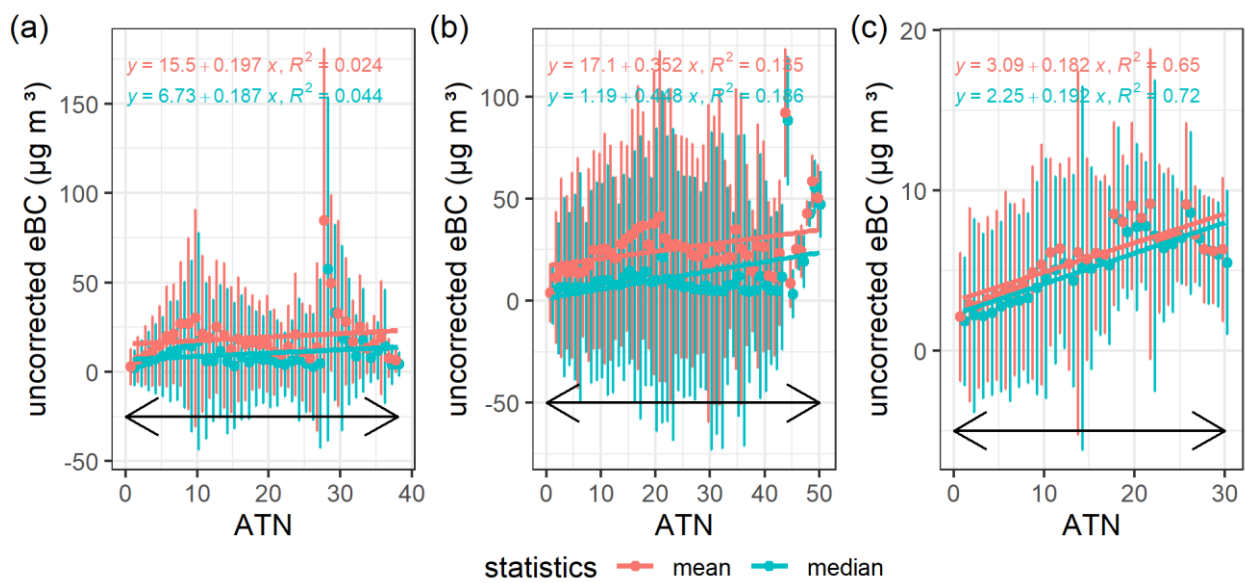
842

843

844

845

Fig. 9



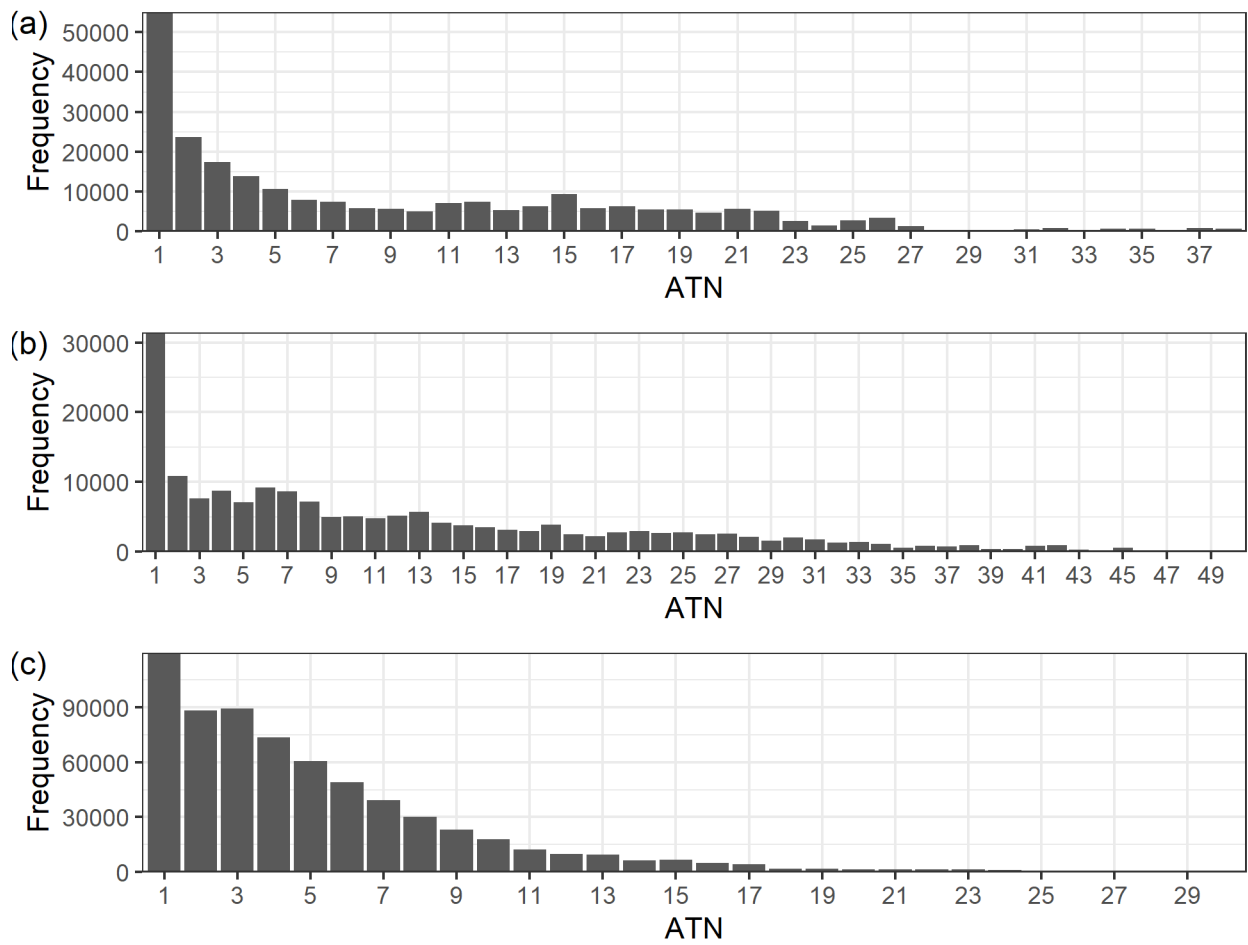
846

847

848

849

Fig. A1.



850

851

852

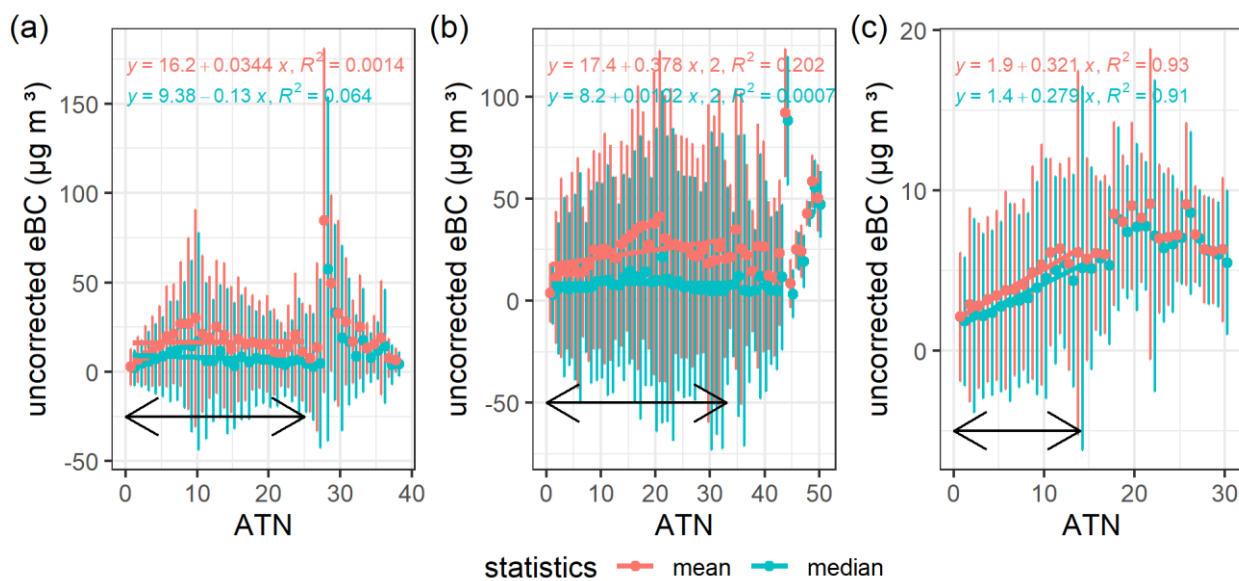
853

854

855

ACCEPTED

Fig. A2.



856

857

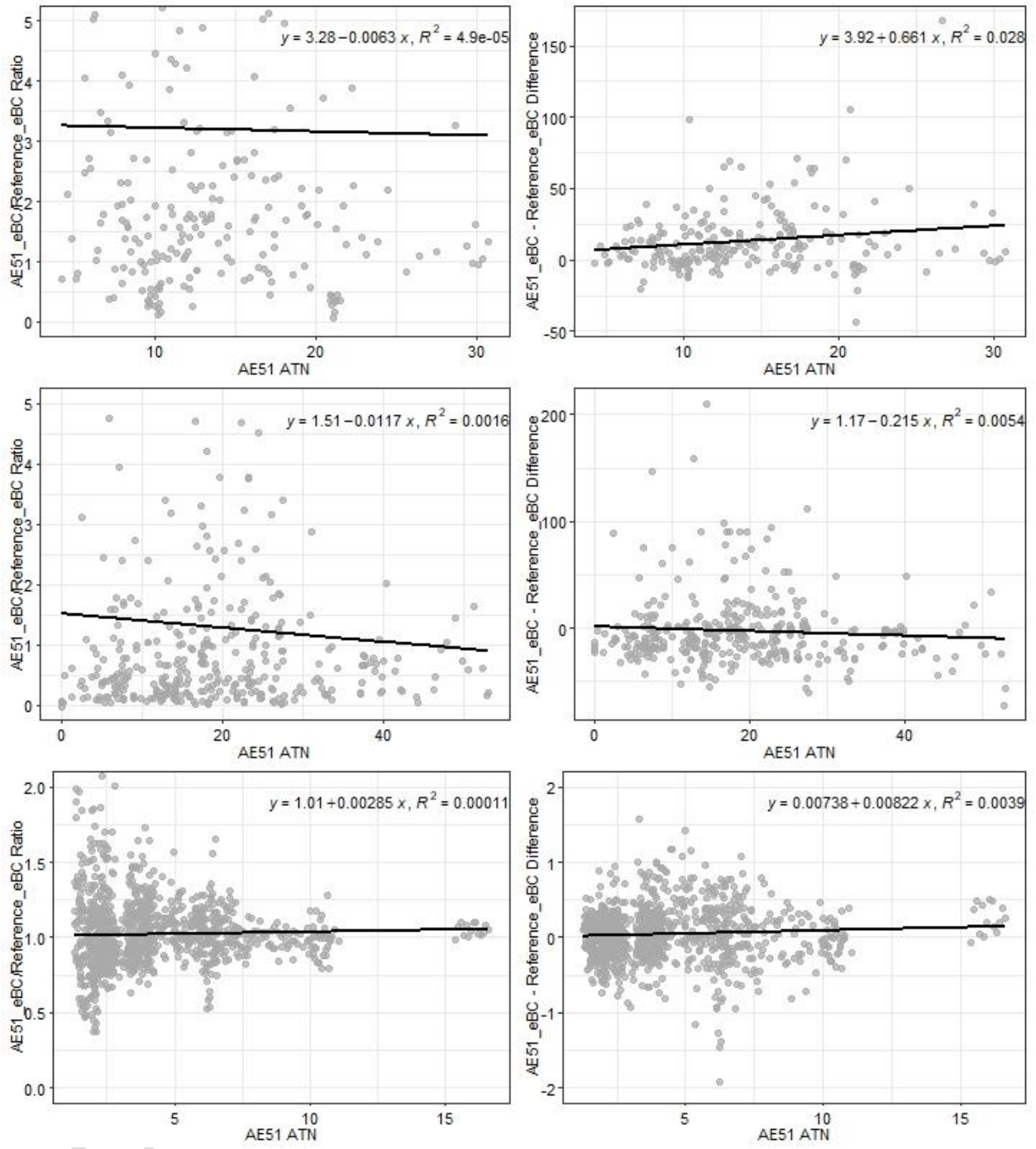
858

859

860

861

Fig. A3.



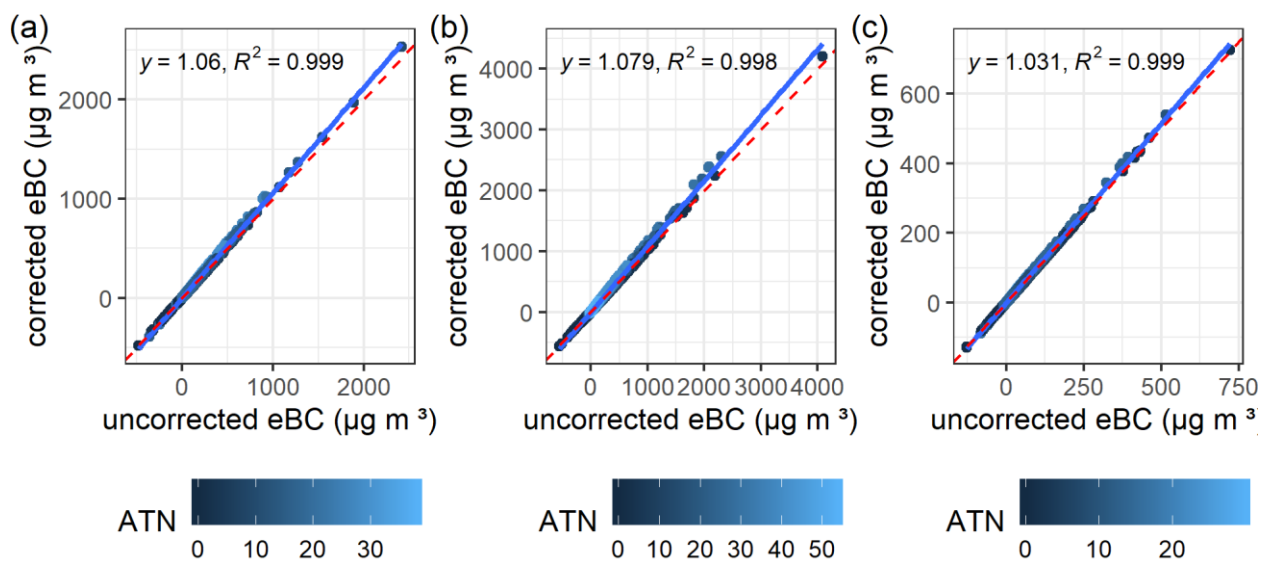
862

863

864

865

Fig. A4



866

867

868

869

870

Fig. A5