Correspondence to Anonymous Referee #1

The authors of the manuscript entitled "Intercomparison and characterization of 23 Aethalometers under laboratory and ambient air conditions: Procedures and unit-to-unit variabilities", thank the valuable comments and inputs from the Anonymous Referee #1. All the points expressed were addressed. Below, we answer to each one of your comments.

Major comments

Comment 1: The authors have mentioned the scattering effect and its compensation constant, C. Not much is detailed in the manuscript about this artifact and they claim this aspect is out of the scope of the study. However, this seems to be a relevant issue for Aethalometers. It would be nice if the authors could provide an estimate of how sensitive the AE is to the different artifacts; filter-loading, scattering by deposited particles, scattering by filter fibers.

Response: The Aethalometers were "calibrated" using the measurement of transmission of light (the determination of attenuation, ATN) and a measurement of carbon content using Soxhlet extracted filters. The assumption was made that the chemically refractory fraction of the sample (remaining on the filter after Soxhlet extraction) is also the light absorbing fraction – this was the "definition" of black carbon (Gundel et al., 1984). The parameter relating ATN and eBC mass concentration is the mass attenuation cross-section, that is the product of the multiple-scattering parameter *C* and the mass absorption cross-section (Drinovec et al., 2015).

The value of the multiple-scattering parameter *C* (as parametrized in Weingartner et al, 2003, and Drinovec et al., 2015) is crucial for the determination of the aerosol absorption coefficient. The separation of the loading effect and the multiple-scattering effect in the filter is arbitrary, to a degree. The quantification of the *C* value requires a comparison between the Aethalometer and a reference instrument. This has so far proved to be challenging, especially at high single-scattering albedos. Even when the loading effect is corrected for, the comparison slope, often interpreted as the parameter *C*, is influenced by the cross-sensitivity to scattering of all filter photometers, the particle size effects and their penetration depth into the filter. All of the effects above depend on the filter properties and the sample properties.

This manuscript is focused on the comparability, repeatability and noise of the measurements of the BC mass concentration, the so called eBC. Therefore, we prefer to discuss the measurement in terms of the sensitivity and any potential loss of sensitivity due to the properties of the sample. The potential change of the sensitivity when measuring a sample

different from what has been used for the characterization of the instruments is a major source of uncertainty of the Aethalometer measurement. Bernardoni et al (2020) have shown that the wavelength dependence of *C* is not very large. They have conducted their field campaign in the period of considerable uncertainty in the filter properties and have "played it safe" by using only the published filter with known properties (T60A20, also referred to as M8020). The unpublished results from the manufacturer (as documented in the filter box) seem to indicate that the multiple-scattering parameter for the new filter (M8060) C=1.39.

To assess the variation between the instruments, we have used well-defined samples. CAST soot and nigrosin, and then used the instruments to measure ambient air in urban background conditions. This serves well the comparison purpose. For the study of the changes in sensitivity when measuring different samples, focusing especially on BC with different size distributions, we have already carried out some laboratory experiments in the framework of the EMPIR Black Carbon project, using different types of reference instruments which measure the absorption without the interferences on the filter, for example the so-called "extinction minus scattering" (see below, reply to Comment 3), photoacoustic instruments and two different photothermal interferometers (one of which is described in Visser et al., 2020).

We have explicitly addressed the use of different tapes in the manuscript. The change of tape from one (T60A20, also referred to as M8020) type to the current one (M8060) requires the user to change the multiple-scattering parameter *C* and the leakage factor Zeta. In our experience, this is the most important systematic error in the measurements that the user can make in a field or laboratory measurement campaign. We have included this in the recommendations.

Comment 2: The CAST soot particles seem to have a particle number mean diameter at around 50 nm according to figure 4. This size might be too small for Aethalometer measurements given the particle penetration in the filter. Can the authors comment on that?

Response: We have used a well-defined sample to characterize and compare the Aethalometers. The size is representative of fresh BC, for example such as would be measured next to a busy street. One of the major considerations was the stable operation of the CAST BC source. We have verified the comparison using ambient measurements. The determination of the sensitivity of Aethalometers to BC of different sizes due to the different penetration into the filter or other artifacts is beyond this work. Please see above (reply to Comment 1) for the

range of experiments planned or carried out to assess the influence of the size distribution of the measured BC on the sensitivity of the filter photometers.

Comment 3: The authors used an AE33 as a reference but it is not clear why this instrument is the reference and how it was calibrated. I encourage the authors to provide details on this.

Response: This is an important remark. To a degree, when comparing instruments of the same design, the choice of the instrument, to which others are compared, is arbitrary. However, the instrument needs to be well characterized and party to a very strict quality control process. The reference AE33 belongs to the WMO-GAW World Calibration Centre for Aerosol Physics (WCCAP); the instrument receives adequate maintenance and is operated with the correct accessories. The flow of this AE33 is calibrated with an externally calibrated flowmeter model 4140 F, TSI Inc. Additionally, the absorption coefficients reported by this reference aethalometer, have been compared with the absorption estimated by a reference set-up from the WCCAP, consisting in one nephelometer Aurora 3000, EcoTech, measuring the aerosols light scattering coefficients, and one CAPS PMex Monitor, Aerodyne Research, Inc, measuring the aerosols optical extinction. The absorption from the reference set up is calculated as *absorption = extinction – scattering*.

We have included complementary information about the performance of the reference aethalometer in the supplementary material (Figure S1), and in the section "2.3 Experimental set-up", which contains now the next paragraph:

"*The aethalometer AE33 used as reference belongs to the WCCAP; it receives frequent maintenance, and is operated with the correct accessories (filter tape M8060). The flow of this AE33 is calibrated with an externally calibrated flowmeter model 4140 F, TSI Inc. The reported absorption coefficients of this reference AE33, have been compared with the absorption calculated by a reference set up from the WCCAP, consisting in one nephelometer Aurora 3000, EcoTech, measuring the aerosols light scattering coefficients, and one CAPS PMex Monitor, Aerodyne Research, Inc, which measures the aerosols optical extinction. The absorption from the reference set up is calculated as absorption = extinction – scattering, at 450, 525 and 635 nm; the measurements at 635 nm are extrapolated to 660 nm. The results from this comparison are shown in Figure S1 from the supplementary material*".

Comment 4: When discussing the wavelength-dependency of the unit-to-unit variability the authors should comment on the effects of different artifacts that affect Aethalometer measurements. Since the unit-to-unit variability is only based on a comparison to a reference Aethalometer, the whole wavelength-dependency seem to fit quite well but it is well known that, for example, scattering artifacts will be different at the different wavelengths. The way it is presented might lead to the reader to understand that Aethalometers would be good for retrieving the absorption Angstrom coefficient. Is that the case? Can the authors comment on that? Lines 531-534 is a strong statement that cannot be supported with the evidence presented here.

Response: The largest wavelength-dependent uncertainty in the measurements is due to the loading effect – the attenuation (ATN) scales with the mass attenuation cross-section and the loading effect is higher at lower wavelengths. When determining the absorption Ångström exponent from the Aethalometer data, the loading effect needs to be corrected first. Any potential dependence of the multiple-scattering parameter *C* is taken care of in the second step. The wavelength dependence of *C* is a smaller artifact than the loading effect (using values from Bernardoni et al., 2020, and scaling them to the new tape M8060 (Yus-Díez et al., in preparation); or carrying the experiments with proper reference instruments, see answer to Comment 1). However, the assumption of a universal *C* used to determine the wavelength dependence of the absorption coefficient is only an assumption, the dependence needs to be determined with reference measurements (Drinovec et al., in preparation) and in different environments and different single-scattering albedo values (Yus-Díez et al., in preparation). There might not be a universal effective *C* values, but the cross-sensitivity to scattering (Arnot et al., 2005) and its wavelength dependence (see above) will need to be determined depending on the sample properties using additional measurements.

To quantify further the wavelength dependent unit-to-unit variability, we introduced a new subsection named "3.4 Wavelength-dependency of the light absorption"; it describes the results from the estimation of the AAE using power law fitting for the three aerosol sources measured during the intercomparison.

The section 3.4 includes the following:

3.4 Wavelength-dependency of the light absorption

The absorption Ångström exponents (α) were calculated for soot and ambient air measurements, by applying a power law fitting describing the wavelength (λ) dependency of the aerosol light absorption (b_{abs}):

$$b_{abs} = \lambda^{-\alpha}, \qquad (9)$$

The absorption coefficients b_{abs} were first determined from Eq. 5, using the 5 minute-averaged eBC mass concentrations, and the default values of the mass absorption cross sections (σ_{air}) used by the AE33 for each wavelength (370 nm: 18.47 $m^2 \cdot g^{-1}$, 470 nm: 14.54 $m^2 \cdot g^{-1}$, 520: 13.14 $m^2 \cdot g^{-1}$, 590: 11.58 $m^2 \cdot g^{-1}$, 660: 10.35 $m^2 \cdot g^{-1}$, 880: 7.77 $m^2 \cdot g^{-1}$, 950: 7.19 $m^2 \cdot g^{-1}$) (Magee Scientific, 2018).

Figure 14 shows the histograms of the α estimated for each instrument in group D and the reference aethalometer, before and after maintenance. During soot measurements (Fig. 14a), the median absorption Ångström exponents before maintenance, ranged from 1.19 to 1.3; after maintenance, the median values of α fluctuated from 1.21 to 1.29. For ambient air (Fig. 14b), the median α before maintenance varied from 1.43 to 1.77; after maintenance, the median α ranged from 1.34 to 1.4. For both aerosol sources, the variability of the absorption Ångström exponents were reduced after maintenance (soot: Interquartile range IQR before maintenance = 0.08, IQR after maintenance = 0.05; ambient air: IQR before maintenance = 0.1, IQR after maintenance = 0.07). Values of α larger than 1, may indicate the presence of organic compounds in the aerosol particles of soot and ambient air.

The values of α shown in Fig. 14 were calculated using the absorption from channels 1 to 7 (370 nm to 950 nm); some studies suggest the omission of $b_{abs,\,370\,nm}$ reduces the uncertainty in the estimation of the absorption Ångström exponent and their use in source apportion models (Zotter et al., 2017). We have revised the impact of calculating α with measurements from six channels (470 nm to 950 nm), but no significant advantage or improvement was found from the omission of $b_{abs,\,370\,nm}$ while calculating α from the samples measured in this study.

Minor comments

Comment 5: l. 87 "When optical methods are used, black carbon is called equivalent black carbon (eBC), because the mass concentration is indirectly retrieved from measurements of light attenuation" Please detail more, why BC measured by FBAP is called equivalent BC?

Response: In agreement with this comment we have complemented this description from the introduction as follows:

"*When optical methods are used, the mass concentration of black carbon is indirectly retrieved from optical measurements of light attenuation caused by the aerosol particles – the determined quantity is equivalent to the mass concentration and therefore called equivalent black carbon (eBC; see Petzold et al., 2013). This method employs a conversion factor known as the Mass Absorption Cross Section (MAC), to estimate the eBC mass concentrations*".

Comment 6: l. 146 "In the end, it is provided a series of recommendations for operation and maintenance." Please rephrase.

Response: The last paragraph of the introduction was modified:

"*In this investigation, the authors present the results from the largest intercomparison of aethalometers model AE33, where 23 instruments were characterized and measured BC mass concentrations from three different aerosol sources. The main goal is to determine the unit-to-unit variabilities and their tendencies throughout the spectral range covered by the AE33. Also, we studied the influence of the maintenance activities and accessories used by the instruments, in the reported eBC concentrations. In the end, we provide a series of recommendations for the instrument operation and maintenance*".

Comment 7: l. 247 Remove the n from "ration".

Response: Thanks, this typo was corrected.

Comment 8: l. 257 Please avoid starting sentences with an acronym or abbreviation.

Response: Thanks for this observation, we have checked the document and fixed this.

Comment 9: l. 278 I encourage the authors to mention the R version they used instead of the IDE version (Rstudio).

Response: Agree. In the subsection "2.5 Data processing and analysis", we have modified the sentence as follows:

"*The processes of data cleaning and analysis were performed in the software R version 4.0.0*".

Comment 10: l. 305 This filter tape was used when? Before the 2016-2017 tape (M8050)?

Response: The filter T60A20 was in effect used before the filter M8050, from 2014 to 2016. We have clarified this in the text of the manuscript, now the paragraph reads as:

"*The aethalometer D03 used the T60A20 filter tape (also known as M8020 or AE33-FT), made from TFE-coated glass fibers; this was the first filter used in the AE33 (Drinovec et al., 2015), and was available from 2014 to 2016, it has been since discontinued*".

Comment 11: l. 392 What about D02 and D05 in the beginning of the comparison? It looks like the deviation is >10%.

Response: Thank you for pointing this out. We have modified the paragraph to mention these initial deviations and its causes. Now the paragraph reads as follows:

"*The final comparison was performed during two days. As observed in Figure 8a, the deviations among the eBC mass concentrations reported by the instruments reduced significantly for all aerosol sources (<10 %), in comparison with the initial and intermediate comparisons performed in group D. Significant deviations (>10%) were observed only at the beginning of this final stage, after the first tape advance while the instruments measured BC mass from soot; these higher deviations are associated with the initial adjustment required by the compensation algorithm to a new aerosol source and the effect of a filter tape advance, as mentioned earlier*".

Comment 12: Fig. 9 It looks like D01 and D03 performed better before maintenance. Could you please comment on that?

Response: From Figure 9 the relative slopes of the aethalometer D01 before and after service, indicate deviations of 2% and 6%, respectively. For the aethalometer D03, the deviations before and after service were 2% and 7%, respectively. In both instruments, the deviations (how much the slopes differ from 1), are certainly higher after service, even if they are all relatively low (<10%). As mentioned in line 414, the correlations shown in Figures 9, 10 and 11 were calculated using measurements performed after one or two tape advances, to avoid the effects produced by the filter advance and the initial adjustment of the compensation algorithm. To evaluate the deviations, it is also fundamental to observe the complete time series before and after service (Figures 5, 6 and 8), which give a broader perspective of the performance of the instruments; from this analysis it is clear the offsets and deviations are significantly higher before maintenance, more remarkable in the case of the instrument D03.

We consider clarifying this analysis is relevant, therefore we extend the comments about the specific variabilities shown in the Figures 9, 10 and 11.

Comment 13: Fig. 9-11 Please use decimal points and not commas in the annotations.

Response: We have fixed this error in the revised version.

Comment 14: Table 4 The slope values in the table do not seem to be the same ones shown in Fig. 9. Am I missing something?

Response: Thanks for this observation, you are correct. The non-corresponding values in the Table 4 were corrected.

Comment: 15: Please define the acronyms EBAS, EMEP.

Response: We have asked Stephen Platt from NILU (Norwegian Institute for Air Research) taking care of the EBAS database, about the meaning of the acronym; in his response, Stephen explains the name of the database EBAS is no longer an acronym but only the name of the database.

EMEP → European Monitoring and Evaluation Programme. This was included in the revised version.

References

Arnott, W. P., Hamasha, K., Moosmüller, H., Sheridan, P. J., and Ogren, J. A.: Towards aerosol light-absorption measurements with a 7-wavelength aethalometer: Evaluation with a photoacoustic instrument and 3-wavelength nephelometer, Aerosol Science and Technology, 39, 17–29, https://doi.org/10.1080/027868290901972, 2005.

Bernardoni, V., Ferrero, L., Bolzacchini, E., Forello, A. C., Gregorič, A., Massabò, D., Močnik, G., Prati, P., Rigler, M., Santagostini, L., Soldan, F., Valentini, S., Valli, G., and Vecchi, R.: Determination of Aethalometer multiple-scattering enhancement parameters and impact on source apportionment during the winter 2017–2018 EMEP/ACTRIS/COLOSSAL campaign in Milan, Atmos. Meas. Tech. Discuss. [preprint], doi:10.5194/amt-2020-233, in review, 2020.

Drinovec, L., Močnik, G., Zotter, P., Prévôt, A. S. H., Ruckstuhl, C., Coz, E., Rupakheti, M., Sciare, J., Müller, T., Wiedensohler, A. and Hansen, A. D. A.: The “dual-spot” Aethalometer:

An improved measurement of aerosol black carbon with real-time loading compensation, Atmos. Meas. Tech., 8(5), 1965–1979, doi:10.5194/amt-8-1965-2015, 2015.

Gundel, L. A., Dod, R. L., Rosen, H. and Novakov, T.: The relationship between optical attenuation and black carbon, Sci. Total Environ., 36, 197–202, doi:10.1016/0048-9697(84)90266-3, 1984.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., Holzer-Popp, T., Kinne, S., Pappalardo, G., Sugimoto, N., Wehrli, C., Wiedensohler, A. and Zhang, X. Y.: Recommendations for reporting black carbon measurements, Atmos. Chem. Phys., 13(16), 8365–8379, doi:10.5194/acp-13-8365-2013, 2013.

Visser, B., Röhrbein, J., Steigmeier, P., Drinovec, L., Močnik, G., and Weingartner, E.: A single-beam photothermal interferometer for in-situ measurements of aerosol light absorption, Atmos. Meas. Tech. Discuss. [preprint], doi:10.5194/amt-2020-242, in review, 2020.

Weingartner, E., Saathoff, H., Schnaiter, M., Streit, N., Bitnar, B. and Baltensperger, U.: Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers, J. Aerosol Sci., 34(10), 1445–1463, doi:10.1016/S0021-8502(03)00359-8, 2003.