

Interactive comment on “Development of a Universal Correction Algorithm for Filter-Based Absorption Photometers” by Hanyang Li et al.

Anonymous Referee #2

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The manuscript describes a new correction for filter-based absorption photometers. The method is compared with known correction methods. The authors explain that the method is "universal" due to the wide range of AAEs and SSAs. However, important factors known in the literature are not considered. Details will come later in the review.

The naming of this method in the title as "universal" method is wrong. It remains hidden to the reader that this method has only been developed and tested for specific aerosol types. The limitations are given in the conclusion. Furthermore, it is important to emphasise that the corrected factors for the Bond1999 and Virkkua2005 corrections are only valid for the specific aerosols investigated in this study. An application as a 'standardised' method, as suggested by the authors, is only possible after successful testing with different aerosol types.

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Known correction methods are closely related to physical models. Terms of the corrections are associated with physical effects (e.g. load correction, scattering correction). The model presented here is based more on mathematical optimisation methods. A discussion of the derived parameters (G0 to G7) and how these parameters are related to aerosol properties is missing.

Detailed Review

Line 130: The authors point out the wide range of SSAs and AAEs. However, it is not shown to what extent these variables are correlated in the available data sets. Scatterplots of AAE, SSA and also SAE would provide valuable information. On this basis, an aerosol classification, as shown for example in Schmeisser et al. (2017), could give valuable information to what extent the data set is sufficient for a universal correction.

Chapter 2: Information on relative humidity or aerosol drying is missing. For developing correction methods, a chapter on instrument calibrations and description of corrections is required.

A consideration of the particle size effect is missing. It is known that smaller particles can penetrate deeper into the filter and absorb more light there by multiple scattering by the filter material. This effect was theoretically illustrated in Moteki et al (2010) and Nakayama et al (2010) and has been shown in laboratory measurements. According to Nakayama et al., this effect would explain differences of a factor of 2 and greater for mono disperse aerosol. This effect is unfortunately the worst quantified effect for complex aerosols. For the present manuscript this means that without a particle number size distribution of the particles containing absorbing material, the magnitude of the effect cannot be estimated. A correction is only possible with a complex aerosol physical characterization. Nevertheless, possible artefacts due to this effect should always be discussed.

The cross-sensitivity to scattering is important for high SSAs. The Magnitude depends

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on particle size and wavelength (wavelength effect can be seen in Müller et al., 2011a). It was shown with radiative transfer calculations (Mueller et al., 2014) that this effect is linked to the scattering asymmetry parameter. In Mueller et al. (2014) it was also shown that for high SSAs ($SSA > 0.95$) the corrections become strongly nonlinear and are only insufficiently described by current correction methods. This can also be found in equation 8 of the present manuscript. The second term in 'g' is $(1-ssa)/(1-(1-c1)*ssa)$. In what range of SSAs equation 9 is a good approximation of equation 8?

The approach in equation 9 is a linear equation with interaction terms. According to radiative transfer calculations, this approach makes sense only in a limited range of parameter values. Why AAE is used as independent parameter is not quite obvious. Light scattering and absorption are independent for each wavelength. An indirect dependence on the aerosol type (e.g. composition) is possible. Unfortunately, the parameters and constant are only treated mathematically. A link to aerosol properties would be welcome. This could result in further boundary conditions for the mathematical fit and a more robust determination of the values of the constants might be possible.

In Li et al (2019) the FIREX data set has already been discussed under a different aspect. It was shown that PAX generally provides lower values than other instruments (TAP, CLAP, Offline, EC/OC, SP2). The authors wrote: "This could imply that most instruments over-estimate BC concentrations, but it could also imply that the PAX-870 measurement is incorrect. However, in the absence of a "ground truth" measurement, we cannot confirm or reject either claim. These discrepancies between instrument cannot be explained by measurement uncertainties alone." This gives rise to two possibilities: a) aerosol type or b) larger experimental uncertainties than expected from instrumental uncertainties

a) The special properties of the generated aerosol types are responsible for discrepancies during FIREX. Biomass burning is a challenge. Especially since the different types of instruments measure different properties! The PAX and CLAP/PSAP measure the light absorption and are therefore comparable in principle. It is correct to consider PAX

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as a reference instrument, because filter based methods require complex corrections. The available data for the FIREX campaign must therefore be considered as being very specific and cannot be used as a universal calibration.

b) The discrepancies between PAX and PSAP/CLAP (see Fig. 5 in Li et al. 2019) for "Black" cluster like particles are significantly higher for the FIREX experiment than values from the literature. These differences require a precise presentation of the calibration, data processing and possible cross-comparisons to underline the high quality of data. Differences due to artefact of measurement techniques should be discussed in detail.

Line 198: From which instrument was the scattering Angström exponent (SAE) calculated? Where scattering data corrected for truncation?

The results of the multidimensional fit must be checked for physical meaning. Example Table S7, B1999-SGP-CLAP: The parameters C2 and C3 add up to about 5 for all wavelengths. This represents the correction factor for a fresh filter ($Tr=1$) and black aerosol (negligible scattering). The strong wavelength dependence of C2 and C3, however, contradicts the literature. The results in Virkkula et al (2005) (see Figure 2) do not show such a strong wavelength dependence of calibration constants.

Line 306: Aim 1: The range of values of Babs is not important for any correction. Corrections are function of the optical depth of loaded particles (shown in Mueller et al (2014)). Concentration levels is merely important for performing experiments in an acceptable time and proper signal to noise.

Figure 3: Is the SSA given for the respective wavelength or for the wavelength of the reference device, the PAX?

Line 670: In Mueller et al. (2011a) no photoacoustic photometer was used. The "reference" instrument was a MAAP. In Arnott et al. (2005) a slope of 1.6 was determined. The measurements were, to the knowledge of the Reviewer, also performed with "SGP-

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aerosol". Thus, the aerosol could have similar properties to the SGP measurements performed in this study and would not be a true independent measurement (speculation of the reviewer). Davis et al (2019) compared the B1999, V2005 and M2014 (Mueller et al. 2014) methods. Corrected values with the B1999 and V2005 methods were 40 to 50% higher than values of a PAS. The M2014 method could almost halve the deviations, although the M2014 method is identical to the B1999 and V2005 methods for low SSAs. The improvements are therefore clearly due to the improved scattering correction and probably also to the introduction of a mixed term between scattering and absorption. It would be very interesting to see how the performance of the M2014 method would be for the present FIREX measurements.

Line 701: The authors wrote "It is unclear how the algorithms will work for other absorbing aerosols (e.g., dominated by fossil fuel emissions or mineral dust)." There is a simple test. For an unloaded filter ($\ln(\text{TR})=0$) and assuming an aerosol with $\text{SSA}=0.9$ and $\text{AAE}=1.1$, a not atypical aerosol, g can be calculated using the parameters listed in Table 4. Calculated values are: $g=0.19$ for FIREX/CLAP_green and $g=-0.3$ for SGP/CLAP_green. Negative values are not possible. What has the reviewer done wrong?

As already indicated, a deeper discussion of the results with regard to aerosol physical properties would be welcome. This could lead to an outlook for further correction models that include the advantages of the individual models.

The reviewer has not addressed the application of the new correction to Aethalometers. The reason is that the reviewer is in complete agreement with the authors that there should be a standard method, since the physics in PSAP, CLAP and Aethalometers is the same.

Literature

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