Response to RC1

We appreciate the referee comments and ideas that helped to improve the manuscript. Our responses are presented below (in black the original comments from the referee and our responses in green).

The paper presents an analysis of multiwavelength absorption data collected at an Amazonian site. Aerosol optical properties were measured with an aethalometer, MAAP and a nephelometer. The MAAP filter spots were later analyzed with an offline method, the MWAA that was considered here as the absorption standard. The MWAA is based on the same principle as the MAAP since it measures both transmitted and scattered light and a radiative transfer algorithm similar to that in the MAAP is applied to calculate the absorption coefficient. The good point is that it has several wavelengths, the weak points are that it is still a filter-based method with related artifacts and that its time resolution is not as good as that of online instruments.
The aethalometer data were processed with two methods, the Schmid et al. (2006) and the Collaud Coen et al. (2010) algorithm and the important Cref value was retrieved for both methods. The analysis shows that there are large sources of uncertainty of Cref and the wavelength dependency of absorption (AAE). One of the algorithms seems to be better in one respect, the other in another way. Absolute truth is still not found.

Although absolute truth is not found, our study presents new evidence that helps to evaluate the efficiency of the different AE correction algorithms to retrieve the absorption wavelength dependence. The main advantage of this work is that AE corrected data was compared to a multi-wavelength measurement that is compensated for multiple scattering effects. Given that the filter loading effect was not significant, the uncertainties in the reference multi-wavelength method are minimal, although still a filter-based method.

1. My first question is related to Eq. (11). On line 256 you write that you use five different AAEs to calculate SSA and further Cref. Does this not result in five different Crefs? Do you give the average as the final Cref? Why not using the calculated AAEs instead of five fixed values? The whole procedure is not clear enough and unambiguously explained so that I would try to apply it to my own data.

A reasonable range of AAE is used to calculate wavelength-dependent *C* values, which are then used to fit ln(*C*) vs. ln (λ). The obtained fit parameters are fitted vs. AAE in order to parameterize *C* as a function of λ and AAE. Being AAE unknown in the beginning and giving the fact that C depends on AAE, the parameterization was the optimal approach Schmid proposed to solve this issue.

The five different AAEs are only used for this parameterization and the calculated AAEs are used later in the algorithm to obtain the final absorption coefficients.

Our scripts are available in the following link:

https://dx.doi.org/10.6084/m9.figshare.c.3501153.v3

In order to make the procedure clearer we modified the manuscript as follows:

Changes to Section 2.3.1:

"Attenuation coefficients at 590 nm were interpolated to 637 nm assuming a power-law relationship as,

$$\sigma_{\mathrm{ATN}}(637\,\mathrm{nm}) = \sigma_{\mathrm{ATN}}(590\,\mathrm{nm}) \cdot \left(\frac{637\,\mathrm{nm}}{590\,\mathrm{nm}}\right)^{-å_{\mathrm{ATN}}} \qquad (7)”$$

“The compensated absorption coefficients, σ_{ap}, are calculated from attenuation coefficients, σ_{ATN}, by accounting for the different artifacts,

$$\sigma_{\mathrm{ap}} = \frac{\sigma_{\mathrm{ATN}}}{(C_{\mathrm{ref}} + C_{\mathrm{sca}}) \cdot R} = \frac{\sigma_{\mathrm{ATN}}}{\left(C_{\mathrm{ref}} + m_{\mathrm{s}} \frac{\omega_0}{1-\omega_0}\right)\left[\left(\frac{1}{f} - 1\right)\left(\frac{\ln \mathrm{ATN} - \ln 10}{\ln 50 - \ln 10}\right) + 1\right]} \qquad (5)$$

where C_{ref} compensates for the scattering effects in comparison with a reference absorption measurement, C_{sca} accounts for the scattering effect of non-absorbing aerosol particles and R, for the filter-loading effect. The Schmid formulation uses the scattering factor m_s and ω_0 to calculate C_{sca} and the filter loading correction proposed by Weingartner et al. (2003), which takes ATN = 10 % as a reference point and includes the shadowing factor parameter, f, which describes the slope between σ_{ATN} and ln(ATN).”

“The slope of this relationship was given by the shadowing factor parameter, f. By applying a linear fit to the R_{meas} values obtained from Eq. (9) and the attenuation data, as shown in Eq. (10), the term ($1/f - 1$) can be obtained from the slope.

$$R_{\mathrm{meas}} = \left(\frac{1}{f} - 1\right)\left(\frac{\ln \mathrm{ATN} - \ln 10}{\ln 50 - \ln 10}\right) + 1 \qquad (10)$$

Assuming f is wavelength independent, the averaged f is the used to calculate R at different wavelengths.”

“In the next step, C, understood as the overall multiple scattering correction factor (C_{ref} + C_{sca}), is parameterized as a function of λ. The single scattering albedo, ω_0, at 637 nm is used in the following equation to calculate C as

$$C = C^{*} + m_s \frac{\omega_0}{1-\omega_0} \qquad (11)$$

where C^* corresponds to the multiple scattering effect by filter fibers and m_s to the aerosol scattering factor found by Arnott et al. (2005) (see Table S1). The implemented approach is useful to examine any wavelength dependence on C. The values of ω_0 are interpolated to the different Aethalometer wavelengths by using the Eq. (12), assuming that absorption and scattering coefficients follow a power-law wavelength dependence described by $å_{ABS}$ and $å_{SCA}$, respectively.

$$\omega_0(\lambda) = \frac{\sigma_{\mathrm{sp}}}{\sigma_{\mathrm{sp}} + \sigma_{\mathrm{ap}}} = \frac{\omega_{0,\mathrm{ref}}\left(\frac{\lambda}{\lambda_{\mathrm{ref}}}\right)^{-å_{\mathrm{SCA}}}}{\omega_{0,\mathrm{ref}}\left(\frac{\lambda}{\lambda_{\mathrm{ref}}}\right)^{-å_{\mathrm{SCA}}} + (1-\omega_{0,\mathrm{ref}})\left(\frac{\lambda}{\lambda_{\mathrm{ref}}}\right)^{-å_{\mathrm{ABS}}}} \qquad (12)$$

Different $å_{ABS}$ values (1; 1.25; 1.5; 1.75; 2) are then used to generate different correlation factors between ln(C) vs. ln(λ). The coefficients resulting from a quadratic fit are used to parameterize C as a function of $å_{ABS}$ (see Fig. 4 in Schmid et al. (2006)). An iteration procedure is used to force the convergence of $å_{ABS}$. In our calculations, the data converged after seven iterations.”

Changes to section 2.3.2:

“In this study we implemented the Collaud Coen correction algorithm that resembles the

Schmid correction (see eq. 14b in Collaud Coen et al. (2010)). This algorithm is different from the original Schmid algorithm in the calculations of the filter-loading effect and the multiple scattering correction factor. As shown in Eq. (6), the Schmid algorithm filters the data for ATN < 10 % in order to account only for the scattering by filter fibers in the C_{ref} calculation. On the other hand, Collaud Coen algorithm applies a prior filter-loading correction and then, by dividing the reference absorption data (MAAP) by the Aethalometer attenuation coefficients, they obtain C_{ref}, which accounts for both, scattering by filter fibers and scattering by embedded aerosol particles."

"Finally, the corrected absorption coefficients are calculated in a similar way to Eq. (5) but using m_s from Eq. (15) and averaging C_{ref}, m_s, ω_0 and R over a filter spot period; i.e., from a filter change time to the subsequent one."

Section 2.3.3 was merged with 2.3.2.

2. Line 256. "Using different AAE (å_abs = 1,..." Why do you use the symbols AAE and å_abs for the same thing? Be consistent throughout the text.

All "AAE" acronyms were replaced by the symbol "$\mathring{a}_{ABS}$".

3. L384-389 "A scatter plot of both corrections' outputs vs. MAAP measurements is shown in Fig. 4. We found that corrected AE data fitted very well the MAAP measurements in the case of the Schmid correction, with a slope of 1.04 (1.02 – 1.05); i.e, the Schmid correction overestimates the absorption coefficient by only 2– 5 %. In the case of the Collaud Coen correction, it was found that the AE corrected absorption coefficients were underestimated by 19 – 21 %." I don't understand. My first thought is that the Crefs are just wrong. As far as I have understood the whole thing of getting Cref is based on forcing the aethalometer-derived absorption to match the MAAP-derived absorption. Should not the slope should be very close to if not exactly 1 regardless of the algorithm selected? Please explain.

We found an error in our algorithms that affected the C_{sca} term of the Collaud Coen correction and scaled down the absorption coefficient obtained with this correction. After fixing the error, we updated Fig. 4, Fig 3c, and Fig. 8. Now both corrections show a good comparison to the MAAP at 637 nm. The update in the algorithms did not affect the wavelength dependence of the absorption. All updated figures and discussion are attached to this document.

4. L439-440. It is reminded to evaluate critically the corrected filter-based absorption data when using it to retrieve BrC / BC contributions. There are quite a few papers that discuss this same issue. Give some references to such papers, some more credit to earlier work could be given.

We rephrased as:
"A near-UV over- or underestimation of the data, will substantially affect brown carbon calculations, if apportionment algorithms based on the wavelength dependence of absorption are used. More details on the effects of inaccurate $\mathring{a}_{ABS}$ on the BrC/BC apportionment are discussed in Garg et al., 2016; Schuster et al., 2016a, 2016b; Wang et al., 2016 and references there in. A BrC estimation is beyond the scope of this paper."
The included references are detailed below.

5. Fig 3. Beta should have units. scattering coefficient = beta x lambdaˆ(-alpha)

The referee is right. We added the units to Fig. 3 (a).

6. Figures 6 and S4 are just the same with the exception that in S4 there is one more line. Why would you not show it simply in Fig 6 and omit S4?

We agree. Figure 6 was updated to include all data included in Fig. S4. Figure S4 was removed from the supplementary material.

 # End of referee comments and author responses #

References

Garg, S., Chandra, B. P., Sinha, V., Sarda-Esteve, R., Gros, V. and Sinha, B.: Limitation of the Use of the Absorption Angstrom Exponent for Source Apportionment of Equivalent Black Carbon: a Case Study from the North West Indo-Gangetic Plain, Environ. Sci. Technol., 50(2), 814–824, doi:10.1021/acs.est.5b03868, 2016.

Schuster, G. L., Dubovik, O. and Arola, A.: Remote sensing of soot carbon – Part 1: Distinguishing different absorbing aerosol species, Atmos. Chem. Phys., 16(3), 1565–1585, doi:10.5194/acp-16-1565-2016, 2016a.

Schuster, G. L., Dubovik, O., Arola, A., Eck, T. F. and Holben, B. N.: Remote sensing of soot carbon – Part 2: Understanding the absorption Ångström exponent, Atmos. Chem. Phys., 16(3), 1587–1602, doi:10.5194/acp-16-1587-2016, 2016b.

Wang, X., Heald, C. L., Sedlacek, A. J., de Sá, S. S., Martin, S. T., Alexander, M. L., Watson, T. B., Aiken, A. C., Springston, S. R. and Artaxo, P.: Deriving brown carbon from multiwavelength absorption measurements: method and application to AERONET and Aethalometer observations, Atmos. Chem. Phys., 16(19), 12733–12752, doi:10.5194/acp-16-12733-2016, 2016.

Changes to the manuscript

Updated equation

Equation 14 (15 in the new version)

$$m_s = \beta_{SCA}^{(d-1)} \cdot c \cdot \lambda^{(-å_{SCA}(d-1))}; \qquad (15)$$

$d = 0.564$;

$c = 0.00032910$ (σ_{sp} in Mm^{-1} units)

Updated figures

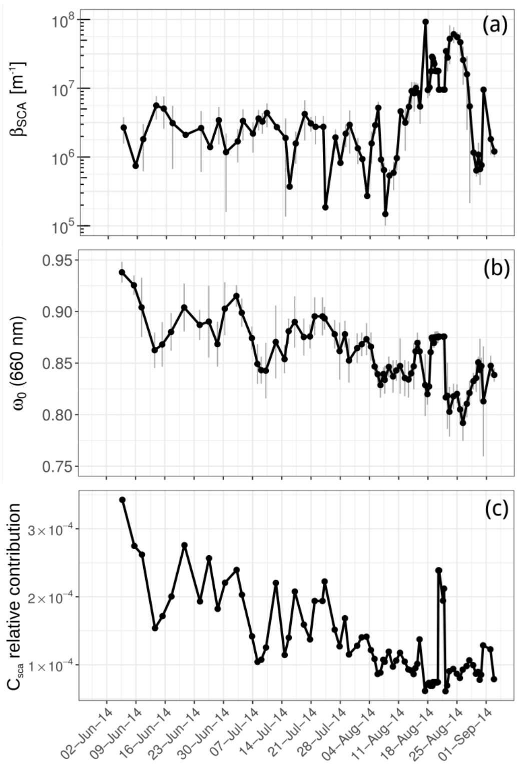

Figure 3. Filter cycle averaged data corresponding to (a) scattering proportionality constant, (b) single scattering albedo at 660 nm, and (c) relative contribution of C_{sca} to the total multiple scattering compensation ($C_{ref} + C_{sca}$) at 660 nm. Vertical bars in (a) and (b) correspond to one standard deviation.

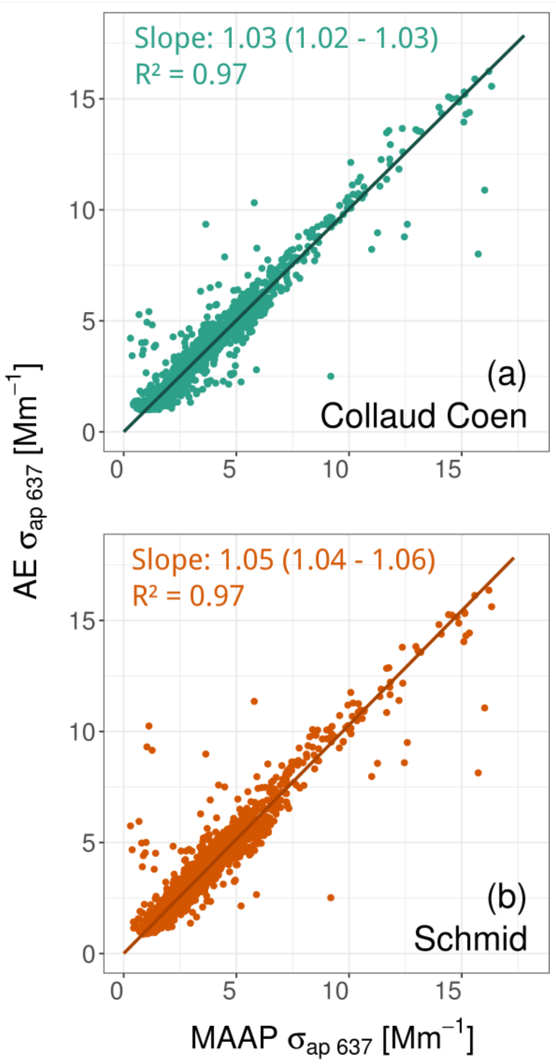

Figure 4. Scatter plot of (a) Collaud Coen and (b) Schmid corrections results vs. MAAP absorption coefficients (all data at 637 nm). The fit was obtained by applying a standardized major axis regression. The fit slopes include the limits to the 95% confidence intervals in brackets.

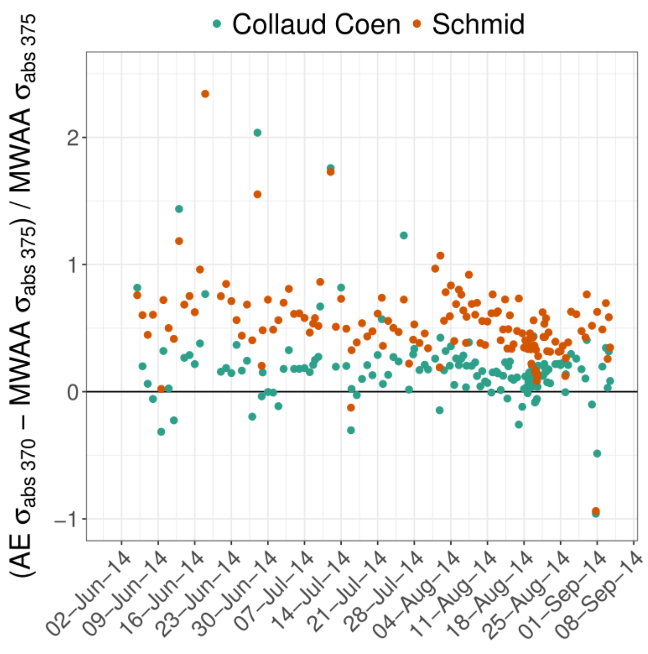

Figure 8. Overestimation of Aethalometer corrected absorption coefficients relative to MWAA at 370 nm. Values above zero are related to an overestimation of σ_{ap} and, below zero, to an underestimation of σ_{ap} at this given wavelength.

Updated discussion

Section 3.1:

"We observed that a lower ω_0 during the biomass burning period was related to a decrease in the scattering correction factor, C_{sca}. The relative contribution of C_{sca} was examined and it was found that the relative contribution from the scattering correction decreases with decreasing ω_0, and increasing β_{sca}, see Fig. 3."

"A scatter plot of both corrections' outputs vs. MAAP measurements is shown in Fig. 4. We found that corrected AE data fitted very well the MAAP measurements for both correction algorithms. The slopes were 1.05 (1.04 – 1.06) and 1.03 (1.02 – 1.03) for the Schmid and Collaud Coen corrections, respectively, with significant correlation factors. The slight difference between both correction schemes in terms of the comparison to MAAP measurements can be related to the parameterization of *C* applied by Schmid et al., which is not implemented by Collaud Coen et al., and the way Collaud Coen et al. estimate C_{ref}."

Section 3.2:

"The original attenuation Ångström exponent (without applying any compensation) was also found to fit quite well the MWAA-retrieved $\mathring{a}_{ABS}$, (Slope IQR: 0.89 – 1.10 with $R^2 = 0.75$, not shown). This finding is in accordance with Ajtai et al., 2011 who found a good agreement between 4-λ PAS measurements and the Aethalometer raw wavelength dependence at a sub-urban site."

Section 3.4: Removed.

Conclusions

"We applied two different correction algorithms to compensate for the various Aethalometer absorption measurement artifacts. The compensated data was compared to an offline multi-wavelength reference absorption measurement technique. This comparison allowed studying the effects of the correction schemes on the absorption at lower wavelengths and showed how this affects the $\mathring{a}_{ABS}$ retrieval. We found that both analyzed algorithms efficiently reproduce the reference MAAP absorption coefficients from Aethalometer data. However, the Schmid algorithm overestimates the $\mathring{a}_{ABS}$ compared to that obtained by the multiple wavelength measurement (MWAA). On the other hand, the Collaud Coen algorithm as well as the "raw" Aethalometer attenuation spectral dependence reproduced quite well the $\mathring{a}_{ABS}$ values obtained from MWAA measurements. The under- or overestimation of short-wavelength absorption coefficients by compensation algorithms is a factor that has to be considered when using corrected Aethalometer data to apportion the black and brown carbon contributions to total absorption. When comparing the absorption coefficients obtained from the different correction algorithms to the reference measurement at 370 nm, we found that the Collaud Coen algorithm is more appropriate to achieve the best comparison at this wavelength, especially for data with $\sigma_{ap} > 5\ Mm^{-1}$. The Schmid algorithm resulted in high enhancements of the absorption coefficients at 370 nm over the sampling period."

Abstract

"Deriving absorption coefficients from Aethalometer attenuation data requires different corrections to compensate for artifacts related to filter-loading effects, scattering by filter fibers, and scattering by aerosol particles. In this study, two different correction schemes were applied to 7-wavelength Aethalometer data, using Multi-Angle Absorption Photometer (MAAP) data as a reference absorption measurement at 637 nm. The compensation algorithms were compared to 5-wavelength offline absorption measurements obtained with a Multi-Wavelength Absorbance Analyzer (MWAA), which serves as a multiple-wavelength reference measurement. The online measurements took place in the Amazon rainforest, from the wet-to-dry transition season to the dry season (June – September 2014). The mean absorption coefficient (at 637 nm) during this period was 1.8 ± 2.1 Mm^{-1}, with a maximum of 15.9 Mm^{-1}. Under these conditions, the filter-loading compensation was negligible. One of the correction schemes was found to artificially increase the short-wavelength absorption coefficients. It was found that accounting for the aerosol optical properties in the scattering compensation significantly affects the absorption Ångström exponent ($\mathring{a}_{ABS}$) retrievals. Proper Aethalometer data compensation schemes are crucial to retrieve the correct $\mathring{a}_{ABS}$, which is commonly implemented in brown carbon contribution calculations. Additionally, we found that the wavelength dependence of uncompensated Aethalometer attenuation data significantly correlates with the $\mathring{a}_{ABS}$ retrieved from offline MWAA measurements."

References

Ajtai, T., Filep, Á., Utry, N., Schnaiter, M., Linke, C., Bozóki, Z., Szabó, G. and Leisner, T.: Inter-comparison of optical absorption coefficients of atmospheric aerosols determined by a multi-wavelength photoacoustic spectrometer and an Aethalometer under sub-urban wintry conditions, J. Aerosol Sci., 42(12), 859–866, doi:10.1016/j.jaerosci.2011.07.008, 2011.