Development of a Universal Correction Algorithm for use with any Filter-Based Absorption Photometers (amt-2019-336)

Reply to Anonymous Referee #2:

We thank the reviewer for the careful reading of our manuscript and for providing a set of very thoughtful comments and critiques, which will be addressed in turn below. Below we restate your editorial comments in italics, and then indicate our response and revision. Please note that all the revised text and references in this response document have been included in the revised manuscript.

> *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.*

Response #1: We first would like to kindly clarify that our intent in calling this a "universal" algorithm is due to its potential application on different filter-based absorption photometers (including PSAP, CLAP, TAP and Aethalometer), rather than its applicability to a wide range of AAE and SSA. The reviewer appears to have capture this intent with her/his final review comment: "*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,*" but this appears to be a different point entirely.

We did try to emphasize our intent for a "universal" algorithm in our original submission, e.g.:

Introduction:

Line 114: In addition, we propose "universal" correction algorithms that are applicable to any filter-based absorption photometer (e.g., CLAP, TAP, PSAP, and AETH) across multiple wavelengths by combining observed filter-based B_{abs} with B_{scat} (e.g., from a co-located nephelometer (NEPH)) and reference B_{abs} (e.g., from a co-located photoacoustic instrument).

Line 136: Our algorithms were then extended to the AETH data from the FIREX laboratory campaign and the PSAP data collected at the SGP site to verify the "universal" nature of the algorithms.

Discussion:

Line 660: Overall, the derived properties using the new correction are consistent across all instruments, suggesting its universality.

Conclusion:

Line 728: We also developed "universal" correction algorithms that are applicable to any filter-based absorption photometer (e.g., PSAP, CLAP, TAP, AETH), which will have utility for any historic or future filter-based absorption measurements and which have the potential to standardize absorption coefficients across all filter-based instruments.

However, this critical disconnect between our intent and the reviewer's interpretation of our intent exists. To clarify for the reviewer and any other reader of this work, we have added and/or revised text as follows:

We revised the title of our manuscript to "Development of a Universal Correction Algorithm **for Use with any** Filter-Based Absorption Photometer".

Line 118 (new text): "We emphasize that the "universal" feature of our algorithm is based on its applicability to different filter-based absorption photometers, rather than the ranges of aerosol parameter tested in this work."

line 337 (new text): "…, but we have the additional aim of developing a model that is applicable to any filter-based absorption photometer."

Line 562 (revised text, bolded here for emphasis): "**To explore the universal behavior of the new algorithms across different instruments**, we next apply our algorithms to the other filter-based absorption photometers operated during the FIREX study (TAP and AETH)."

> *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 Virkkual2005 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.*

Response #2: As we stated above, the so-called "universality" is not based on aerosol properties, so we again apologize for this confusion. Regardless, we do agree with the reviewer that our correction factors derived by B1999 and V2005 are only valid for the aerosols tested in this work. Therefore, we added the following sentence to emphasize this in Line 422: "It is important to keep in mind that the updated coefficient values of B1999 and V2005 (Table S7) are only valid for the aerosols investigated in this study. Future experiments are needed to systematically determine how the coefficients in B1999 and V2005 may change for different aerosol types."

Moreover, we added a new section (section 2.4.3) describing how we updated the coefficients in B1999 and V2005.

Line 316: "2.4.3 Refitting the coefficients in B1999 and V2005

With the reference measurements of B_{abs} from the photoacoustic instruments, we are able to refit the coefficients in the B1999 and V2005 corrections (C_2 to C_7 in Eq. (5) and Eq. (6)) using our data. Specifically, we use the Levenberg-Marquardt algorithm (1944) to iteratively fit the coefficients until the chi-square of the coefficients are minimized. The fitting is implemented using the "Curvefit" function in Igor Pro. It is worth noting that the derived coefficients may only be valid for the SGP and FIREX data. For aerosol properties different from our study, the optimal coefficients are likely to be different from the ones reported here. Hereafter, the B1999 and V2005 results with refitted coefficients are referred to as “updated B1999” and “updated V2005”, respectively.”

> *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.*

Response #3: We first would also like to correct an error in Eq. 9 which the reviewer highlights by the calculation of $g < 0$ in a later comment: the parameters after G_5 and G_6 ($\ln(\lambda) \times$ AAE and SSA $(\lambda) \times$ AAE, respectively) should be exchanged. Eq. 9 should be:

$$g(Tr(\lambda), SSA(\lambda), AAE) = G_0 + G_1 \times ln(Tr(\lambda)) + G_2 \times SSA(\lambda) + G_3 \times AAE + G_4 \times ln(Tr(\lambda)) \times SSA(\lambda) +$$

$$\boldsymbol{G_5} \times \boldsymbol{SSA(\lambda)} \times \boldsymbol{AAE} + \boldsymbol{G_6} \times \boldsymbol{ln(Tr)} \times \boldsymbol{AAE} + G_7 \times SSA(\lambda) \times AAE \times ln(Tr(\lambda))$$

The coefficient values in Table 4 and in our supplemental computer code were derived using this correct form of the equation, and there was simply an error in this equation in the manuscript text. We apologize to the reviewer for causing this confusion and are very grateful to her/him for bringing this to our attention. (This explains the negative “g values” that the reviewer discusses in a later comment).

Towards this comment, in the original manuscript, the development and interpretation of Eq. 9 were included in the supplementary materials (pages 2-6), as subsections of “model development”. In the main text, we briefly interpreted how these parameters relate to SSA, AAE, and Tr (Line 350). To further relate these parameters with aerosol properties, we add the following sentences:

Line 352: “Although Eq. (9) is developed based on statistical approaches, we attempt to relate this statistical model to physical effects. The coefficients $G_1 – G_3$ are fairly straightforward, as these account for the influence of filter loading (G_1), relative light scattering by the aerosols (G_2), and the brownness of the aerosols (G_3). The interaction terms ($G_4 – G_7$) are more difficult in assigning a physical meaning; however, the interaction between filter loading and relative light scattering (G_4) is might be interpreted as an absolute light scattering by the aerosols on the filter, while the interaction between filter loading and aerosol brownness (G_6) is somewhat analogous to G_4. The three-way interaction between filter loading, scattering and brownness of aerosols (G_7) is required because of the three two-way interaction terms.

To further this physical interpretation of our statistical model (Eq. 9), we explore the relationship between $\frac{B_{abs}}{B_{ATN}}$ and ln(Tr), which essentially follows a “y = m·x + b” form, where y is $\frac{B_{abs}}{B_{ATN}}$ and x is ln(Tr). The slope (m) is defined as $G_1 + G_4 \times SSA + G_6 \times AAE + G_7 \times SSA \times AAE$, and the intercept (b) is defined as $G_2 \times SSA + G_3 \times AAE + G_5 \times SSA \times AAE$. Therefore, different combinations of SSA and AAE modulate this relationship between $\frac{B_{abs}}{B_{ATN}}$ and ln(Tr). For example, loading “black” particles on the filter (e.g., AAE ~1 and SSA ~0.3) tends to produce larger values of $\frac{B_{abs}}{B_{ATN}}$, while loading “white” particles on the filter (e.g., AAE ~3 and SSA ~0.9) tends to produce smaller values of $\frac{\mathrm{B_{abs}}}{\mathrm{B_{ATN}}}$ (see Fig. S3 of this work and Fig. 4 in Virkkula et al. (2005)). This relationship becomes more complex when considering, e.g., mixed sulfate and black carbon particles; SSA can be high while AAE is low, and the corresponding $\frac{\mathrm{B_{abs}}}{\mathrm{B_{ATN}}}$ can be variable (also see Fig. S3). Therefore, in order to properly compensate for the effects of loading and aerosol optical properties, a multiple linear regression with interaction terms is introduced in Eq. (9)."

> *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.*

Response #4: Thank you for this discerning comment. It was our hope that the use of a broader range of SSA and AAE could make this more widely applicable, but as the reviewer has pointed out elsewhere, this will not be strictly true (and we agree!). Regardless, to address this comment, we generated Figure R1 for the SGP data using similar "AAE-SAE space" as that in Cappa et al. (2016) and Schmeisser et al. (2017). In panel a), our data are colored by SSA and the SGP results from Schmeisser et al. (2017) is illustrated by the brown marker and error bars. In panel b), the AAE and SAE results of all NOAA/ESRL sites from Schmeisser et al. (2017) overlap with our SGP data.

Here, we first emphasize that the SGP data used in our work are subsamples of the campaign reported in Schmeisser et al. (2017), but in general, the AAE and SAE values calculated for the SGP site in our work agree well with the values reported for SGP in Schmeisser et al. (2017). Moreover, our results of AAE and SAE overlap with most of the values from the NOAA/ESRL sites, except when marine aerosols or dust contribute to the local aerosol populations. Though none of the NOAA/ESRL sites fall into the clusters of BC or BC/BrC, some of our data can represent the optical properties of aerosols from these clusters. Therefore, we highlight that our algorithm developed by the SGP data *may* have the potential to be generalized to a variety of environmental conditions, but we would need to validate this using observations from more studies. We again emphasize that our intent was to develop an algorithm that is universal across all filter-based absorption photometers, and not universal across all aerosol types, so such a validation is outside the scope of this work.

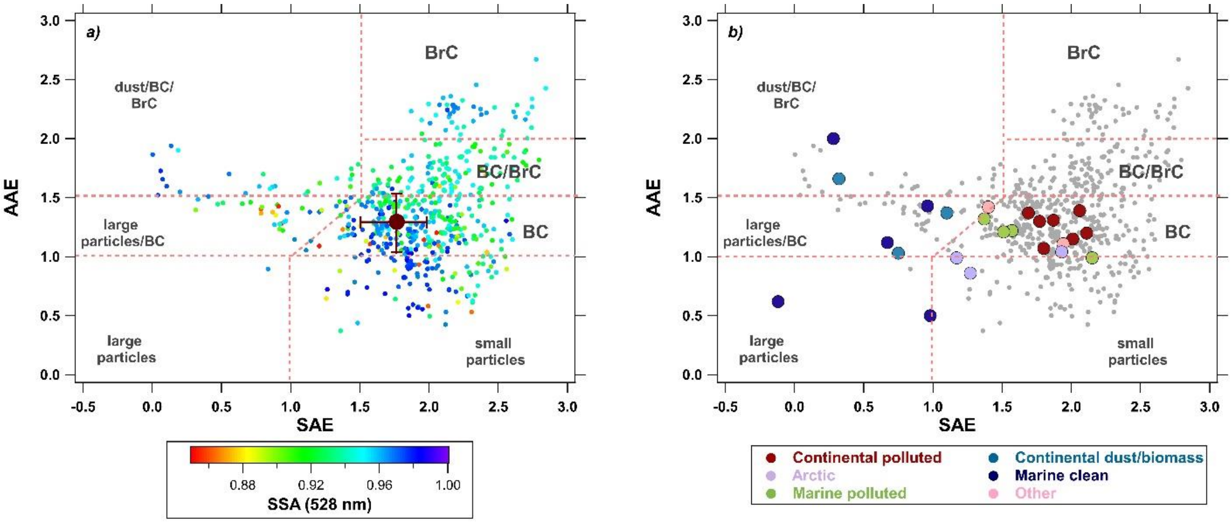

Figure R1. AAE vs. SAE for the SGP data. The three parameters are calculated using photoacoustic B_{abs} and Nephelometer B_{scat}. The panels are overlaid with the classification scheme presented in Cappa et al. (2016) and Schmeisser et al. (2017). In panel a), the averaged values (and standard deviation) of AAE and SAE reported for the SGP site in Schmeisser et al. (2017) are illustrated by the brown marker and error bars. Our results are colored by the corresponding SSA. In panel b), the results in Schmeisser et al. (2017) are colored by the types of observational station and our results are colored in grey.

Figure R1 has been included in the revised supplementary material (Figure S15) and revision was made accordingly:

Line 252: "The scatterplot of AAE and SAE for the SGP data can be found in Fig. S15. Our results of AAE and SAE are compared to the values reported for different NOAA Earth System Research Laboratory (ESRL) observational sites in Schmeisser et al. (2017)".

Line 608: "To investigate if our algorithms are suitable to correct B_{abs} obtained from different ambient environments, the aerosol properties from the SGP site are compared to those from the other NOAA/ESRL observational sites. We use the similar "AAE-SAE space" as that in Cappa et al. (2016) and Schmeisser et al. (2017) to infer dominant aerosol types (e.g., BC, BrC, dust, mixed dust/BC/BrC, see Fig. S15). ..."

To further investigate how different algorithms apply to different aerosol properties, we generated Figures R2 and R3 (SGP and FIREX, respectively), in which the variable on y axis is B_{abs} ratio = corrected B_{abs} (different corrections) / B_{abs} from photoacoustic instruments (reference), and the parameters on x axis include relative humidity (RH), AAE, SAE, and SSA (528 nm). In general, an apparent association between the B_{abs} ratio and these parameters exists in the uncorrected data (raw B_{ATN}), and this association persists when using B1999 and V2005, especially for RH and SSA. However, these associations are reduced or eliminated when applying our algorithm on the filter-based absorption measurements. Although RH and SAE are not included in our algorithm, our algorithm appears to account for any influence that these parameters have on the measurements. This effect may be captured by one of the interaction terms in our statistical model, but as discussed in Response #3 above, it is difficult to assign physical meaning to these. These figures will also support our response to some of the reviewer's comments later in this file (Response #5 and Response #6).

Revision was made accordingly:

Line 375: "We evaluate the model by plotting $\frac{\mathrm{B_{abs}}}{\mathrm{B_{ATN}}}$ against aerosol properties not included in Eq. (9) (such as relative humidity and aerosol geometric mean diameter, which have been previously reported to bias corrections of filter-based $\mathrm{B_{abs}}$, (Moteki et al., 2010; Nakayama et al., 2010; Schmid et al., 2006)). The results are presented in Fig. S5-S7."

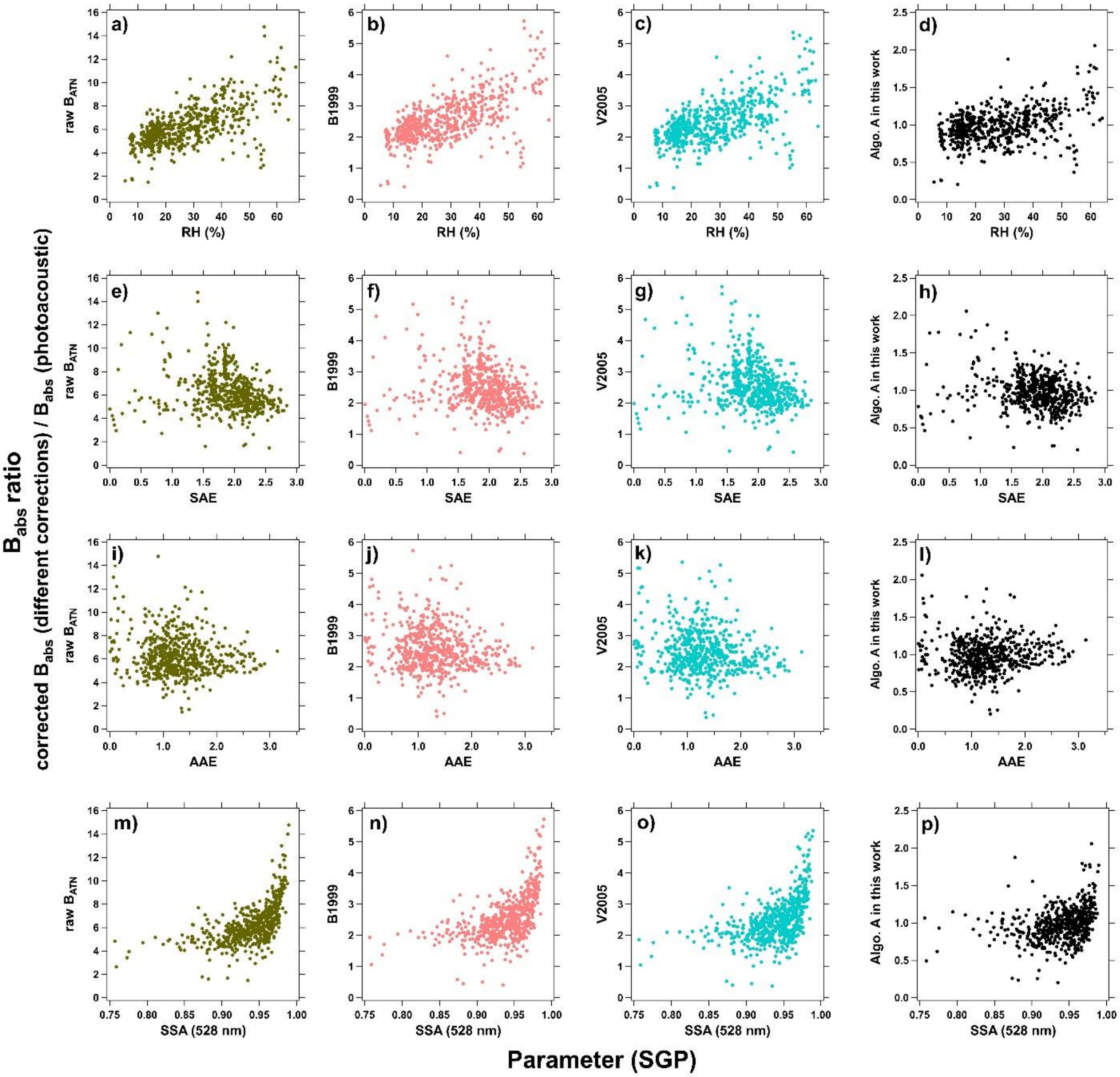

Figure R2. $\mathrm{B_{abs}}$ ratio vs. different parameters at the SGP site. In the first column, $\mathrm{B_{abs}}$ ratio = uncorrected $\mathrm{B_{ATN}}$ / $\mathrm{B_{abs}}$ (photoacoustic). In the other columns, $\mathrm{B_{abs}}$ ratio = corrected $\mathrm{B_{abs}}$ by different corrections / $\mathrm{B_{abs}}$ (photoacoustic).

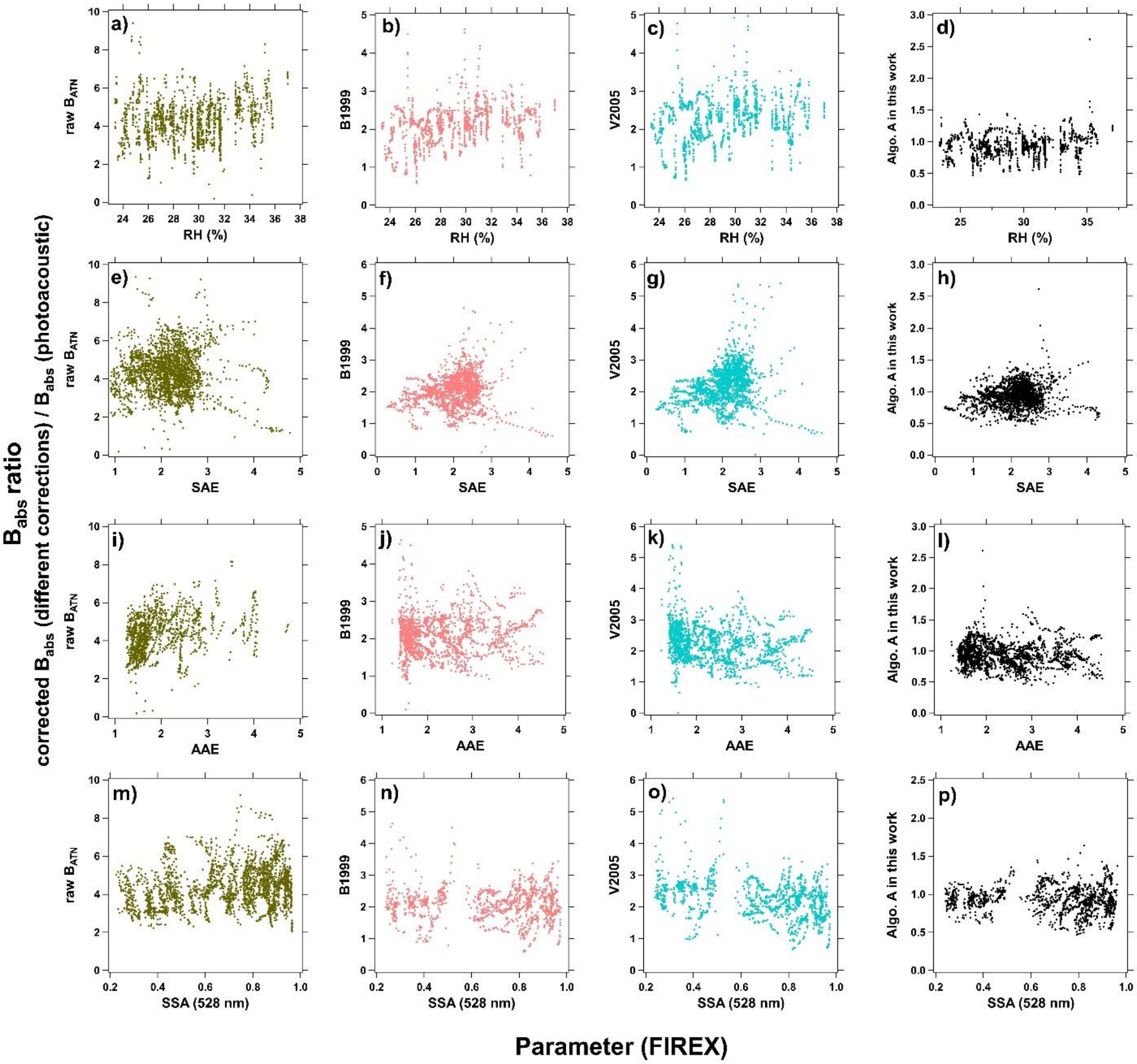

Figure R3. B_{abs} ratio vs. different aerosol properties from the FIREX campaign. In the first column, B_{abs} ratio = uncorrected B_{ATN} / B_{abs} (photoacoustic). In the other columns, B_{abs} ratio = corrected B_{abs} from different corrections / B_{abs} (photoacoustic).

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.

Response #5: Following the reviewer's suggestion, we added the information about RH for both datasets (FIREX: 29.3 ± 3.3, SGP: 26.1 ± 13.9) in a footnote to Table 2. As seen in Figures R2 and R3, our algorithm appears to be able to account for any RH effects. However, we admit that the values of RH for both datasets are relatively low, so extrapolating our algorithms to more humid environments may introduce uncertainties.

Regarding instrument calibration, we agree with the reviewer that it is important when developing correction methods. Thus, when estimating the overall uncertainty of our algorithm in Section 3.5 of the original version, we considered calibration uncertainties (such as the accuracy of the operating wavelengths and the properties of the calibration materials) as part of the measurement uncertainties.

We admit that the instrument calibration procedure was not clearly described in the method section of the original version. It is now revised as following:

Line 230: "Section 2.3. Calibrations

Following Bond et al. (1999) and Ogren et al. (2010), the filter-based instruments were calibrated and corrected for sample area, flow rate, and filter type (see Li et al. (2019) for the FIREX data and Sherman et al. (2015) for the SGP data). Other than that, we did not do any verification beyond the manufacturer's calibration for the filter-based instruments. The SGP nephelometer measurements were corrected for truncation effects (Sherman et al., 2015). The FIREX photoacoustic measurements were calibrated by ammonium sulfate aerosol and fullerene soot (Li et al., 2019)."

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.

Response #6: We agree with the reviewer that aerosols with different particle sizes may result in different penetrating depth in the filter. For the FIREX data, we had SMPS-derived size distribution (14.6 – 736.5 nm, mobility diameter), so we generated Figure R4 to investigate how number-based geometric mean diameter (d_{pg}) of aerosols affects the corrections' performance. Arguably, the pattern for the B1999 and V2005 data agrees with those reported in Moteki et al. (2010) and Nakayama et al. (2010), in that absorption tends to be over-estimated for smaller particles and that this effect is gradually reduced with increasing particle size. Unfortunately, the size distribution of SGP aerosols is unavailable during our target time period, so we cannot extend this analysis to those data.

Compared to B1999 and V2005, the B_{abs} ratio derived by our new correction is much close to unity when plotting against d_{pg}. Although d_{pg} was not considered when developing the algorithm, any effects related to particle size appear to be captured by our algorithm, potentially in one of the interaction terms (like RH and SAE were above). Unfortunately, we do not have a strong qualitative physical explanation for this.

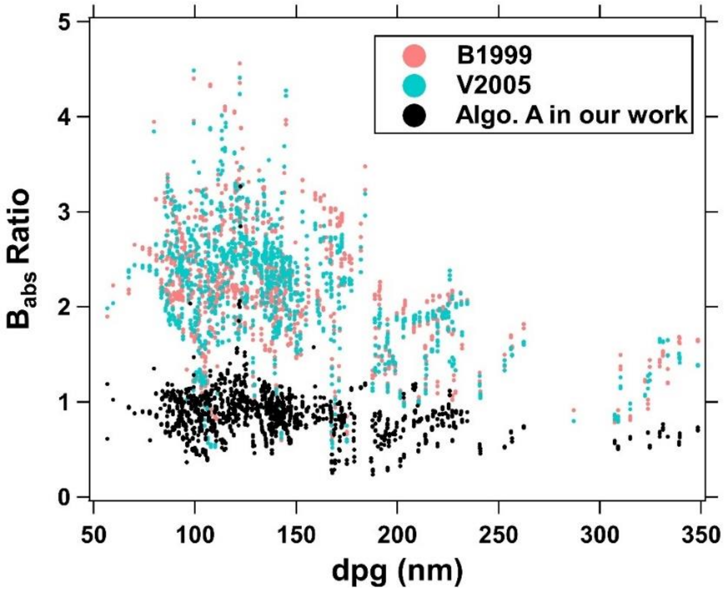

Figure R4. B_{abs} ratio vs. d_g from the FIREX campaign.

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

Response #7: This is an excellent comment. To compare Eq. 9 to Eq. 8, we produce the Figure R5 to compare the simulated "g" term from our correction and the previous corrections. In panels R5a and R5b, we use the original coefficients reported in B1999 and V2005 to simulate the "g" term in Eq. 8. In panels R5c and R5d, we use the updated B1999 and V2005 coefficients from our Table S7 (FIREX-CLAP). When simulating Eq. 9 (panel R5e), we estimate AAE as a function of SSA ($AAE = a + b\times SSA^c$), similar to the procedure of "Algorithm C" in our manuscript. Then, we plot the results of "g" derived by all corrections as a function of SSA (panels R5f – R5h: Tr = 0.9, 0.75, and 0.5).

In general, the values of "g" term from all corrections increase with decreasing Tr and SSA. However, the figure suggests that there are variations among the corrections for different combinations of Tr and SSA. For example, the original B1999 and V2005 corrections tend to yield greater values of "g" than the other corrections (eventually, insufficient correction), and the agreement between them gets worse as Tr and SSA decrease (panels R5f - R5h). Another observation from the figure is that our correction is in better agreement with the updated B1999 and V2005, but this agreement depends on both SSA and Tr. For example, when SSA > 0.95, our correction does not exhibit as strong of a non-linearity as the updated B1999 and V2005. Hence, Eq. 9 is a good approximation of Eq. 8 when SSA < ~0.95, and the non-linearity is dampened compared to Eq. 8 as SSA approaches unity. We have added this discussion to the updated SI.

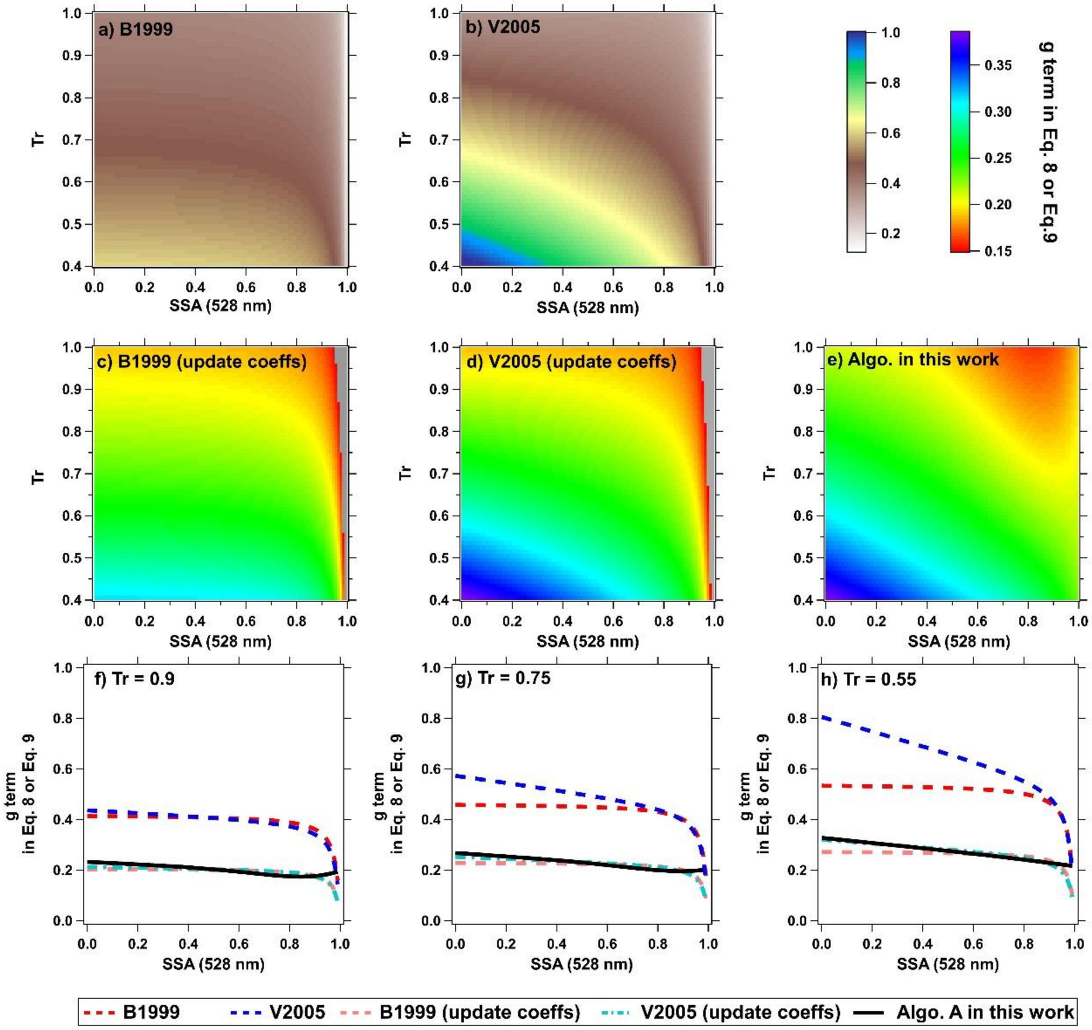

Figure R5. Simulated "g" term in Eq.8 or Eq. 9. In panel c) and d), the grey regions correspond to "g" values less than 0.15.

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.

Response #8: There are three reasons why we included AAE as an independent variable, which are summarized as follows, in order of importance:

1) We found that a model with only SSA and Tr could not well explain the variation of B_{abs} / B_{ATN} (discussed briefly in Response #3). As the reviewer indicated, aerosol type may play a role. To incorporate aerosol composition in our correction, we proposed to use AAE because AAE can provide information about aerosol brownness, and it can be inferred from the filter-based instrument (so no additional instruments are needed).
2) AAE allows for the implementation "Algorithm C" when there are no measurements of B_{scat} to compute SSA.
3) Incorporating AAE into our correction allows to correct B_{abs} at multiple wavelength simultaneously, while the previous corrections treat them separately.

To the reviewer's point, Figure R2 above does suggest that B_{abs} / B_{ATN} is independent of AAE. However, different combinations of SSA and AAE result in different values of this ratio, e.g., as in Figure S3b, reproduced and annotated here:

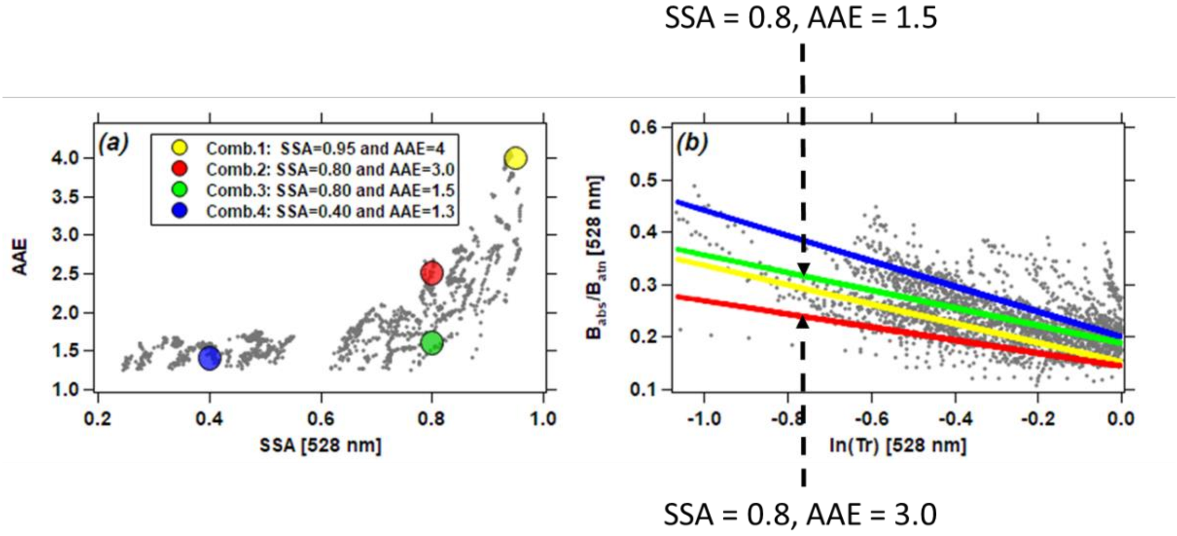

Figure S3. Simple slopes analysis of cross-level interaction of SSA and AAE in predicting $\frac{B_{abs}}{B_{ATN}}$ as a function of ln(Tr) at 528 nm.

Hence, both of these curves differ with a constant offset of ~0.1, with the only difference being the value of AAE used in the computation. We also note that Schmid et al. (2006) and Kim et al. (2019) demonstrate a dependency of AAE on one of the terms used in the Weingartner at al. (2003) correction algorithm for the aethalometer.

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 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.

Response #9:

We appreciate the reviewer's comment on this manuscript and our previous work. As stated earlier in this reply, the aerosol properties from the biomass burning are not used to demonstrate the "universal" characteristic. We utilize the FIREX aerosols as they are very important class of absorbing particles that can be examined in the context of the correction. To clarify that the set of coefficients from the FIREX campaign that we are suggesting is appropriate for biomass burning, we add a footnote below Table 4:

Footnote text: "The coefficients derived from FIREX may be more appropriate for biomass burning aerosols, and the coefficients derived by SGP may be more appropriate for rural background environments in the absence of marine aerosols and dust (see Fig. S15)".

To our knowledge, FIREX is the only biomass burning comparison between PAX and PSAP/CLAP that out there (Tasoglou et al. (2018) compared PAX/MAAP). Although the aerosols from the black cluster in Li et al. (2019) tends be more similar to "pure" BC compared to the other two clusters, these aerosols may be still different from the BC used in the previous studies (such as OA/eBC and AAE). Specifically, that comparison in Figure 5 from Li et al. (2019) includes aerosols from flame-generated soot (using, e.g., propane) and internal combustion engines.

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

Response #10: For the FIREX data, we calculated SAE using B_{scat} measured by the two Photoacoustic Extinctiometers (PAX): 405 and 870 nm. For the SGP data, we calculated SAE using B_{scat} measured by the Nephelometer (450, 550, and 700 nm).

The Nephelometer-B_{scat} at SGP was corrected for truncation effects (Sherman et al. 2015). During the FIREX campaign, there was a cyclone on the inlet of the sample barrel; thus, the sampled aerosol were generally sub-micron and truncation correction would be quite minor. Otherwise, we have no way to make a quantitative correction for those data.

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.

Response #11: Ogren (2010) stated that it was unclear how their correction worked for wavelengths other than 574 nm, as they only tested 1-λ PSAP. Moreover, while we agree that Virkkula et al. (2005) does show a weak wavelength dependence on correction coefficients, other literature does demonstrate this (e.g., (Drinovec et al., 2017; Kim et al., 2019; Schmid et al., 2006)), so a wavelength dependence of these coefficients is not unreasonable.

These differences between models (and spectral dependencies or lack thereof) largely exists because most of these (e.g., B1999, V2005, ours, along with the Weingartner, Schmid, and others that we touch on in our manuscript) are simply not process-based physical models; they are statistical models informed by data. (M2014 appears to be an exception).

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.

Response #12: We thank the reviewer for this comment. Accordingly, we delete "extend the correction a wider range of B_{abs}" in Line 338.

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

Response #13: The SSA represents the respective wavelengths (652, 528, and 467 nm).

The caption is now revised as: Line 478 (revise text, bolded): "The data points are colored by the corresponding SSA **at the given wavelength**".

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-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.

Response #14: Unfortunately, we do not have backscattering measurements for the FIREX data (because we used PAX for B_{scat}, instead of Nephelometer); therefore, it appears to not be possible to apply M2014 on the FIREX data. However, to address the reviewer's comments, we tested M2014 using our SGP-CLAP data. The parameters used by us (e.g., δ_{af}, δ_{sf}, μ_1, χ) are the same as those in Müller et al. (2014) and Davies et al. (2019). We first regenerate Fig. 6 and Fig. 7 in Müller et al. (2014) to validate our coding (see Figure R6 below).

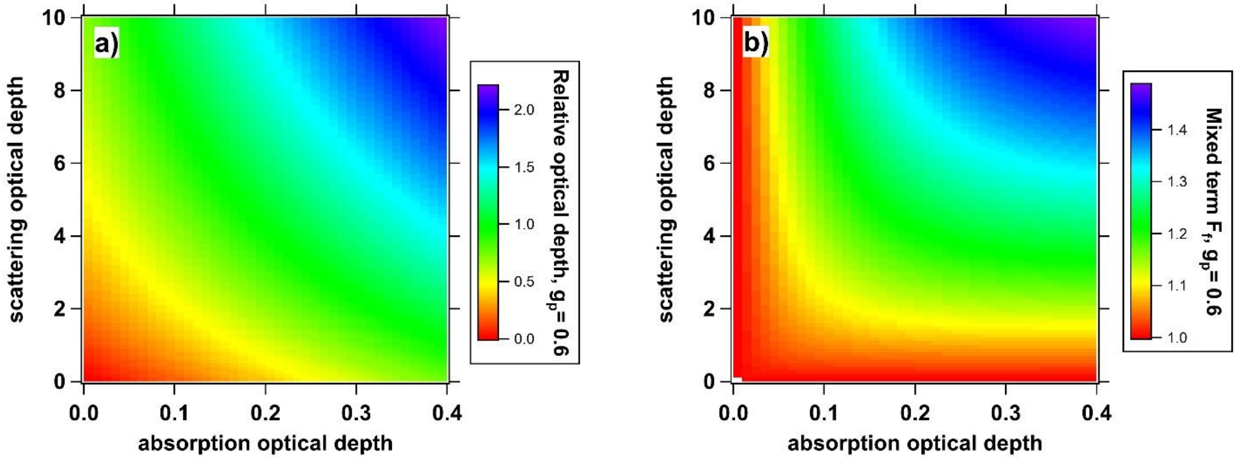

Figure R6. Simulated relative optical depth (panel a) and F_f (panel b) as a function of scattering and absorption optical depths.

As seen in Figure R7, B_{abs} corrected by the M2014 correction agrees fairly well with those derived by the original B1999 and V2005, but overestimates the photoacoustic measurements by factors ~2.5. Another observation is that the performance of M2014 increases as the wavelength decreases that (as seen by the R^2), which is consistent with the results for urban emissions in Davies et al. (2019). We will consider digging deeper to compare the performance of M2014 against the other corrections in the future.

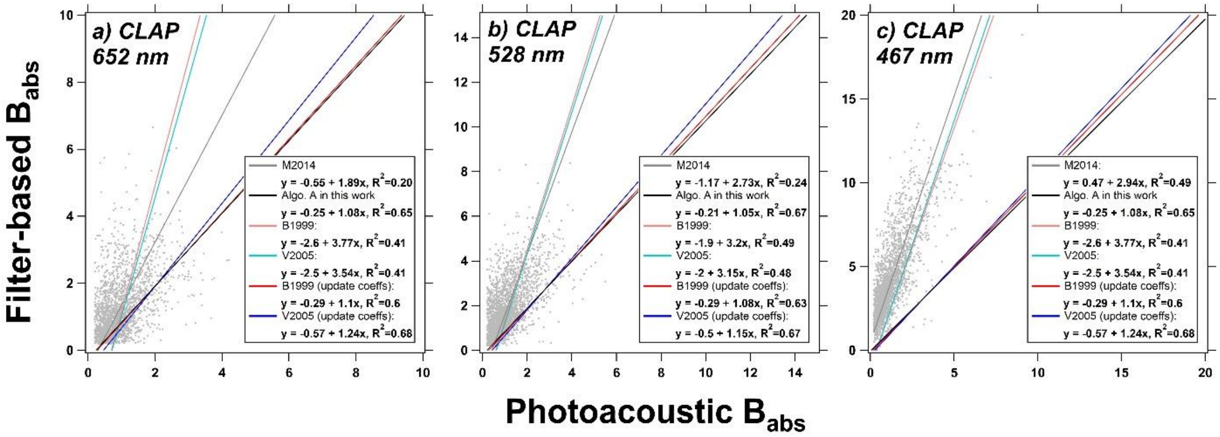

Figure R7. Inter-comparison between the CLAP-derived B_{abs} corrected by the M2014 correction and the reference B_{abs} at 652, 528, and 467 nm for the SGP data. The relationships derived by the other corrections discussed in our manuscript are given in the figure.

Regarding M2014, we make the following revision in our manuscript:

Line 275: “Besides the above mentioned B1999 and V2005 corrections, the constrained two-stream (CTS) correction proposed by Müller et al. (2014) can be also applied on PSAP-similar instruments. The CTS correction was developed based on the relationship between absorption optical depth and B_{abs}. However, it is not straightforward to reformulate the CTS correction as a function of Tr as those presented in Eq. (5) and Eq. (6). Thus, we exclude the B_{abs} results corrected by the CTS correction in the following analysis. For those who are interested in the B_{abs} results corrected by the CTS correction, we provide the correction results of our SGP-CLAP data in Fig. S9.”

Moreover, we agree with the reviewer that the introduction of a mixed term between scattering and absorption may improve the correction performance (**response #8**). This is also our motivation to include the interaction terms in our correction (**response #3**). For example, SSA × AAE may account for the mixing effects between B_{abs} and B_{scat}.

> *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(TR)=0) and assuming an aerosol with SSA=0.9 and 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?*

Response #15: We thank the reviewer for pointing this out. As mentioned in **response #3**, we realized that there was a mistake in Eq. 9 in the manuscript. Using the revised Eq. 9 and the coefficients in Table 4, we calculate the g term (528 nm) using the values assumed by the reviewer (ln(Tr)=0), SSA=0.9, and AAE=1.1): FIREX/CLAP: 0.16; FIREX/TAP: 0.47; SGP/CLAP: 0.11; SGP/PSAP: 0.12.

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.

Response #16: Thank you again for your helpful comments (and we are glad that you are in agreement that there should be some standard method!). We have tried to address the issues raised as best as possible, and we look forward to hearing from you on our manuscript.

References

Bond, T. C., Anderson, T. L. and Campbell, D.: Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols, Aerosol Sci. Technol., 30(6), 582–600, doi:10.1080/027868299304435, 1999.

Cappa, C. D., Kolesar, K. R., Zhang, X., Atkinson, D. B., Pekour, M. S., Zaveri, R. A., Zelenyuk, A. and Zhang, Q.: Understanding the optical properties of ambient sub- and supermicron particulate matter: results from the CARES 2010 field study in northern California, Atmos. Chem. Phys., 16(10), 6511–6535, doi:10.5194/acp-16-6511-2016, 2016.

Davies, N. W., Fox, C., Szpek, K., Cotterell, M. I., Taylor, J. W., Allan, J. D., Williams, P. I., Trembath, J., Haywood, J. M. and Langridge, J. M.: Evaluating biases in filter-based aerosol absorption measurements using photoacoustic spectroscopy, Atmos. Meas. Tech., 12(6), 3417–3434, doi:10.5194/amt-12-3417-2019, 2019.

Drinovec, L., Gregoric, A., Zotter, P., Wolf, R., Anne Bruns, E., Bruns, E. A., Prevot, A. S. H., Favez, O., Sciare, J., Arnold, I. J., Chakrabarty, R. K., Moosmüller, H., Filep, A. and Mocnik, G.: The filter-loading effect by ambient aerosols in filter absorption photometers depends on the coating of the sampled particles, Atmos. Meas. Tech., 10(3), 1043–1059, doi:10.5194/amt-10-1043-2017, 2017.

Kim, J.-H., Kim, S.-W., Ogren, J. A., Sheridan, P. J., Yoon, S.-C., Sharma, S. and Lin, N.-H.: Multiple scattering correction factor estimation for aethalometer aerosol absorption coefficient measurement, Aerosol Sci. Technol., 53(2), 160–171, doi:10.1080/02786826.2018.1555368, 2019.

Levenberg, K.: A method for the solution of certain non-linear problems in least squares, Q. Appl. Math., 2(2), 164–168, 1944.

Li, H., Lamb, K. D., Schwarz, J. P., Selimovic, V., Yokelson, R. J., McMeeking, G. R. and May, A. A.: Inter-comparison of black carbon measurement methods for simulated open biomass burning emissions, Atmos. Environ., 206, 156–169, doi:10.1016/j.atmosenv.2019.03.010, 2019.

Moteki, N., Kondo, Y. and Nakamura, S.: Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, J. Aerosol Sci., 41(5), 513–521, doi:10.1016/j.jaerosci.2010.02.013, 2010.

Müller, T., Virkkula, A. and Ogren, J. A.: Constrained two-stream algorithm for calculating aerosol light absorption coefficient from the Particle Soot Absorption Photometer, Atmos. Meas. Tech., 7(12), 4049–4070, doi:10.5194/amt-7-4049-2014, 2014.

Nakayama, T., Kondo, Y., Moteki, N., Sahu, L. K., Kinase, T., Kita, K. and Matsumi, Y.: Size-dependent correction factors for absorption measurements using filter-based photometers: PSAP and COSMOS, J. Aerosol Sci., 41(4), 333–343, doi:10.1016/j.jaerosci.2010.01.004, 2010.

Ogren, J. A.: Comment on “Calibration and Intercomparison of Filter-Based Measurements of Visible Light Absorption by Aerosols,” Aerosol Sci. Technol., 44(8), 589–591, doi:10.1080/02786826.2010.482111, 2010.

Schmeisser, L., Andrews, E., Ogren, J. A., Sheridan, P., Jefferson, A., Sharma, S., Kim, J. E., Sherman, J. P., Sorribas, M., Kalapov, I., Arsov, T., Angelov, C., Mayol-Bracero, O. L., Labuschagne, C., Kim, S.-W., Hoffer, A., Lin, N.-H., Chia, H.-P., Bergin, M., Sun, J., Liu, P. and Wu, H.: Classifying aerosol type using in situ surface spectral aerosol optical properties, Atmos.

Chem. Phys., 17(19), 12097–12120, doi:10.5194/acp-17-12097-2017, 2017.

Schmid, O., Artaxo, P., Arnott, W. P., Chand, D., Gatti, L. V., Frank, G. P., Hoffer, A., Schnaiter, M. and Andreae, M. O.: Spectral light absorption by ambient aerosols influenced by biomass burning in the Amazon Basin. I: Comparison and field calibration of absorption measurement techniques, Atmos. Chem. Phys., 6(11), 3443–3462, doi:10.5194/acp-6-3443-2006, 2006.

Sherman, J. P., Sheridan, P. J., Ogren, J. A., Andrews, E., Hageman, D., Schmeisser, L., Jefferson, A. and Sharma, S.: A multi-year study of lower tropospheric aerosol variability and systematic relationships from four North American regions, Atmos. Chem. Phys., 15(21), 12487–12517, doi:10.5194/acp-15-12487-2015, 2015.

Tasoglou, A., Subramanian, R. and Pandis, S. N.: An inter-comparison of black-carbon-related instruments in a laboratory study of biomass burning aerosol, Aerosol Sci. Technol., 52(11), 1320–1331, doi:10.1080/02786826.2018.1515473, 2018.

Virkkula, A., Ahlquist, N. C., Covert, D. S., Arnott, W. P., Sheridan, P. J., Quinn, P. K. and Coffman, D. J.: Modification, calibration and a field test of an instrument for measuring light absorption by particles, Aerosol Sci. Technol., 39(1), 68–83, doi:10.1080/027868290901963, 2005.

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.