Reply to interactive comments on "Constrained two-stream algorithm for calculating aerosol light absorption coefficient from the Particle Soot Absorption Photometer" by T. Müller et al.

J. S. Henzing (Referee)

Authors' comment: The authors would like to thank the referee for commenting and correcting the article. We thank for pointing to text passages, which were misleading or difficult to understand. We followed the suggestion of the reviewer and added some more discussion where needed.
Since the word processor obviously suppressed line breaks in the text of the review, the authors have broken the text into smaller pieces. We hope that we have done it right and can give satisfying answers.

General comments:
The paper deals with an important subject. Long-term datasets on aerosol light absorption have been obtained with filter-based methods that suffer from measurements artefacts. The most notable artefacts such as 1) enhanced absorption by multiple scattering and b) the reduction of this enhancement with increasing filter loading and 3) light extinction cause by scattering particles that abusively is interpreted as absorption (apparent absorption) are handled in the manuscript. The authors present a correction method that is new and has a better performance when compared to secondary reference absorption. The correction method is applicable to a larger range of different aerosol types that are present in the atmosphere. How well the method performs for small particles, e.g. traffic related combustion aerosol, that may penetrate the collecting filter substrate deeper than assumed in the model is yet to be determined.
Reply:
The effect of particle penetration depth was not incorporated in the model. This deficit was discussed on page 11106 lines 24ff and page 11122 lines 10ff.

Comment:
A drawback of the correction method may be that it seems not so straight forward for other PSAP owners to apply the suggested correction method. Making the model/method more readily available would be of great benefit.
Reply:
The authors are not refusing to make computer codes public available. It is up to potential users to contact the authors to get a copy prior to publication. After publication, the code will be publicly available on the web pages of the authors.

Comment:
The essential difference (less loss of information) between the constrained radiation transfer model developed here and a (complex) single formula to obtain the absorption coefficient from filter-based light transmission change measurements remains hidden until page 11112 top-para. I would like to see a paragraph at the end of section 2 that describes why correction formulas can never compete with the

model developed in this manuscript.
Reply (page 11111, top paragraph):
Right, the essential differences are that conventional correction functions lose some information. From equation (25) it becomes clear that CTS overcomes this problem by including the scattering optical depth and asymmetry parameter. We couldn't bring this information before equation (25), otherwise it would have been an anticipation without any proof. For the same reason we can not bring any statement on the performance of the CTS and conventional corrections at the end of section 2, which only deals with the basic technical principle of absorption photometers.

Specific comments:
Comment:
Abstract: "For high ssa the CTS correction significantly reduces error"…followed by "reduced to 30%." At first reading there may seem to be a contradiction. I suggest to consider reformulation.
Reply:
The sentences will be reformulated to: "For high single scattering
albedos the CTS correction significantly reduces the errors. For example, at a single scattering albedo of about 0.98 the CTS error amounts to about 30%, whereas errors using the Bond correction (Bond et al., 1999) are about 100%. "

Comment:
Section 2 page 11096, line 4.: Replace "calculated from the Beer Lambert law by" by "defined". Beer-Lambert refers to single scatter.
Reply:
The authors prefer to avoid the term "defined". The reason simply is that, as the reviewer stated, that the Beer-Lambert law strictly is valid for single scattering. The attenuation of particles deposited in a filter matrix violates again this main principle. Therefore the attenuation coefficient is calculated using an equation that is similar to the Beer-Lambert law. This violation is corrected afterwards by introducing correction functions. We will make this point clear and change the sentence to: "Inside the system of filter and deposited particles, multiple scattering of light occurs. Nevertheless, the filter attenuation coefficient is calculated using an equation of the form of the Beer-Lambert law, which is not strictly valid if multiple scattering is non-negligible.

Comment:
Section 2 page 11097, second para on MAAP.
Although MAAP is designed so that "No simultaneous measurements of aerosol scattering is needed" please add "additional" before "measurements" and make the remark that remaining cross sensitivity to scattering (apparent absorption) is observed to be within the range 0-3% (Petzold et al., 2005, Mueller et al. 2011). To make the last para of this page in line with this you may consider to higher the remaining cross sensitivity to scattering for PSAP to 2% (650nm) instead of 1%, which is more valid for the shortest PSAP wavelength.
Reply:

We will give the apparent absorption for all different wavelengths to avoid confusion.

Comment:

Section 2, Last para

It is described that solemnly scattering particles lead to apparent absorption and that the apparent absorption becomes a smaller fraction of aerosol light scattering when relative transmittance goes down. Moreover apparent absorption was higher at larger wavelengths. This finding remains unexplained, whereas this may not be so obvious for the reader.

Reply:

Right, purely scattering particles lead to apparent absorption. That was defined few lines before (page 11097 line 6). The new findings (dependence on loading and asymmetry parameter) are the result of the calibration experiments.

Again, chapter 2 describes the basic principle and observations, and is not intended bring explanations for all artefacts. A discussion of the apparent absorption follows on page 11115. We will include that a sentence concerning the wavelength dependence (page 11115, line 2): … pronounced dependence on the particle asymmetry parameter. This dependence implicitly includes the observed wavelength dependences of apparent absorption (c.f. Mueller et al. 2011). A parameterization…"

Comment:

Please consider to include a short explanation with moral of story:. "Scattering aerosol, cloud droplets or filter fibers scatter light. Even when the preferred scattering direction is forward, the transmission of light through a volume with increasing amount of them goes down. In the multiple scattering regime a cloud looks very white from the top a greyish from below (even though every single droplet scatters forward). The higher the optical depth the lower the transmission and for a hypothetical not absorbing cloud the reflection goes up. For the particle collecting filter the same is true. Here I present some calculations to give an impression on how scattering particles are misinterpreted as absorption and how this leads to wavelength dependent apparent absorption. In Arnott et al., 2005 the filter transmission of a PSAP filter is about 22% at 550 nm. The aethalometer filter is physically and optically thicker with pristine filter transmission about 10%. A simple formula is given by equation 14 in the Arnott paper. The equation is valid for hypothetical non absorbing material.

Reply:

In fact Eq. (17) [=eq 18 in Arnott et al.] is the more fundamental equation. For non absorbing particles (ω=1) it can be simplified to be $\lim_{\omega \to 1} T(\delta_e) = \frac{1}{1+\left(\frac{1-g}{2}\right)\delta_e / \mu_1}$.

The difference from Eq. (14) in Arnott et al. (2005) is the constant μ_1 with a value between 1 and 1/√3. μ_1 accounts for the elongated light path in a multiple scattering medium. We have chosen the value of μ_1=1/√3, which is advised for multiple scattering environments (c.f. reply to Reviewer 2). A value of unity can explain a ping

pong effect (parallel to direction of incident light) but not the enhancement of the light path in direction perpendicular to the incident light. It is essential to use this constant throughout all equations used for deriving the filter optical properties and for simulating the radiative transfer for particle loading. The choice of μ_1 might explain that the derived filter scattering optical thicknesses differs from Arnott et al. (2005).
Also Eq. (7) is easy to use and more general. Thus using the simplified equation for non absorbing particles is not helpful and can obscure the influence of even small absorption.

Comment:
From that formula the filter optical depth for PSAP filters is calculated to be about 25, for asymmetry parameter of 0.72. (25 is much higher than the values in this manuscript under review) If now scattering fibers are added on top of the PSAP filter the transmission goes down, the same would be true for aerosol particles. If I use the scattering coefficient shown in Arnott Fig2. (525 Mmˆ-1), I find for the PSAP 0.525 aerosol optical depth (scatter), this leads to an apparent absorption. The same aerosol on an aethalometer filter has obviously less effect as the fractional increase in total optical depth is smaller. Looking at the wavelength dependence of the PSAP filter transmission (pristine), the wavelength dependence is also explained in similar manner. Here, I included a simple figure (fig.1.). Relative transmission is 100% for pristine and 70% for filter change. Filter spotsize and lpm flow taken from Mueller et al, 2011. Asymmetry parameter aerosol and filter same: 0.72. Sensitivity to asymmetry parameter is small if calculated according to equation 4 in this manuscript. For this calculation the apparent absorption is about 3% of scattering coefficient at pristine filter. The 3% of scattering coefficient is larger than 1-2% and the finding that apparent absorption for aethalometers is smaller than for PSAP (because the change in transmission is relatively smaller as the filter is already optically thick), seems not to be in agreement with the findings of Mueller et al. (2011) but there apparent absorption was already partly corrected for.
Reply:
The authors couldn't follow the discussion of the reviewer. How are the data shown in the attached figure calculated? It is unclear what we shall learn from it, and why that contradicts to the findings of this manuscript?
The thought experiment of the reviewer does not consider the particle penetration depth at all. We admit that we couldn't determine the penetration depth independently, but this was the only free parameter and was determined by adjusting the model to fit to experiments. In the chapter on error consideration this limitation was considered by assuming a high uncertainty for the penetration depth.

A comparison with Aethalometers would be interesting. However, we would like to avoid speculating on numbers for Aethalometer filters, e.g. the filter optical depth, apparent absorption, since we haven't measured the filter optical properties and we don't have such detailed characterization of the scattering artefact like we have for PSAP. Furthermore, we present a new correction method and we could demonstrate the performance for one type of absorption photometers, the PSAP. An application to other instruments is a logical consequence but it is out of the focus of this study.

Furthermore high quality data were missing at the time when writing this manuscript.

Comment: Section 3 page 11098 line 1-17

Here it becomes some-what confusing for me. I try to explain: Line 5 "a model (1) is developed". Line 6 this model includes another model (2) (two-stream RTM) and parametrisations for apparent absorption (this work) and absorption enhancement (Bond and Virkkula). Line 10 "A new correction method was derived from the model for particle-laden filters" I assume derived from model (1)? But the next line the method is given the name CTS thus it is model (2) that is constrained. So I'm confused. Maybe a drawing or scheme may be helpful?

Reply:

The authors agree. The passage from line 5 to the end of the section will be changed to:

"……In order to explain the observations a model for particle-loaded filters was developed. The model includes a two-stream radiative transfer model and parameterizations for the apparent absorption and the absorption enhancement. Since the two-stream model is constrained by the parameterizations, the overall model for particle-loaded filters is called Constrained Two-Stream (CTS) model. The CTS model is a forward calculation to calculate optical properties of the particle-filter system from a known loading with particles. An algorithm is presented for calculating the particle absorption coefficient from measurements of transmittances with a filter based absorption photometer. The core of the algorithm is the CTS model. Consequently the algorithm is called CTS algorithm. Parameterizations were derived from calibration experiments. Experiments with non-absorbing particles led to a new parameterization of the apparent absorption. Parameterizations of the enhancement effect for absorbing particles were taken from Bond et al. (1999) and Virkkula et al. (2005). The CTS correction was compared with the widely used corrections given in Bond et al. (1999) and Virkkula et al. (2005), which are referred as B1999 and V2005 corrections throughout the rest of the manuscript. The CTS algorithm was originally developed for the PSAP. However, one can adopt the correction to other types of filter-based absorption photometers. "

Comment:

Finally CTS, constrained on one side by Bond and Virkkula, is compared to Bond and Virkkula. Final line "one can adopt the correction" what specifically is to be adopted and is "one" the reader or do the authors have plans themselves?. If it is up to the reader, I suggest to include more guidance about Initialisation, constrainments and calibrations.

Reply:

In the summary (p 11121 lines 19 to 21) we wrote concerning a customization for other photometers: "However, the method can be implemented for any other filter-based absorption photometer that measures light transmittance, e.g. the aethalometer, after measuring the optical properties of the filter and the responses to non-absorbing particles." We agree that this description is not sufficient. We will

include a figure to summarize the workflow from model initialization to retrieval of absorption coefficients at the end of chapter 3. Nevertheless we will instruct the reader at the beginning of chapter 3 to have that overview in mind while reading the derivations in chapter 3. The figure is attached as supplementary file.

Comment:
E.g. page 11102 line 6, "illuminated with diffuse light", is that important when adopting.
Reply:
The authors think that this question can't sufficiently be answered. Lines 10 to 12: " If the filter is illuminated with collimated light, radiative transfer models that account for both diffuse and collimated light should be used."
We can't give more rigorous statement ending with "… must be used." since that would require the implementation and adoption of a four stream model, followed by an investigation of differences between the two stream and four stream models with varying illuminations. Such an investigation is out of the scope of this manuscript.

Comment: Section 3.1.
It is mentioned that Arnott et al. (2005) used a two stream model. Page 11099 (line 13-14) it is mentioned that in this manuscript "the simpler two-layer model is used" Thus the same modelling approach is chosen and many of the equations following are identical. However what is the difference only becomes clear if the Arnott paper is studied. I would like to read a few lines where this work continues where the work of Arnott et al. stopped.
Reply:
It was convenient to lean on the two layers two stream method as described by Arnott et al. But the main idea is different compared to Arnott et al. This is that the full loading state is important for radiative transfer and must be preserved until the last step of the correction. As a consequence the model is set up in terms of optical thicknesses and not in terms of absorption and scattering coefficients. As a consequence, we do not end up with a parameterization (which must be solved iteratively) but with a model, which has the radiative transfer model still in its core. Furthermore the matching of model and calibration is completely different.
So we oppose to see the manuscript as a continuation of the Arnott paper. However, we appreciate the work of Arnott et al. since it inspired us and provided a lot of helpful information.

We considered the possibility of including a short comparison of CTS and the method presented in Arnott et al. (2005). But that would necessarily involve a detailed overview chapter on all papers (Bond et al., 1999, Virkkula et al., 2005, Collaud Coen et al. 2010, Schmid et al. 2006) dealing with photometer calibrations. We fear that overview would be lengthy but would not bring new insights. If CTS will be adopted for Aethalometer in the future, then a discussion alongside with the Arnott correction is a must.

Comment: Page 11100

Reasons for the inequality expressed in (3) are given: 1. No independent light scattering.2. Multiple scattering medium. 3.Particle interference. The authors argue that the missing theoretical solutions are solved by empirical parameterizations of apparent absorption and absorption enhancement. To me it is clear how Multiple scattering (2) and absorption enhancement are linked. But I do not see directly how apparent absorption links to the other two. Please comment or just leave it to the conclusion that filter based measurement techniques have artefacts (explained earlier and elsewhere) and that in this manuscript a correction method is developed.

Reply:

The reviewer is right. The sentence "… the interactions are implicitly included in empirical parameterizations. " could be misleading in the context and is too short to explain the basic idea. We will change the paragraph to:

"This means that particle scattering and absorption optical depths in a multiple scattering environment differ from the optical depths for the same particle population in an airborne state because of interference effects. First, there is an interference because the particles are deposited on fibers and do not scatter light independently. Second, the particles are deposited in a multiple scattering environment because of the high number of light scattering fibers in the vicinity of the individual particles. Then the path length of photons passing the filter becomes larger and the probability to be absorbed increases. Furthermore the particle concentration in the filter increases with time and particles may interfere among each other. There is no theoretical solution describing such interference interactions that we can include in the radiative transfer model. Fortunately, this model uncertainty is implicitly compensated by the CTS model. The calibration experiments are subject to these interferences, which means that the parameterization of the apparent absorption and absorption enhancement implicitly contain the interference effects. The CTS corrections thereby inherit a compensation for the interference effects."

Comment:

Equation 4 and introducing text.

Am I confused? The asymmetry parameter can be used to calculate the fraction of light that is scattered forwardly and backwardly. In the single scattering regime this is very convenient. But how does it work with a cloud. The phasefunction of every droplet can properly be integrated to obtain the asymmetry parameter of that drop. However, for many droplets thus a cloud, all individual droplets with large forward scatter and g close to 1, the cloud becomes highly reflective and the transmission goes down. A dense PSAP filter resembles a cloud and thus although single fibers scatter very strongly in forward direction, g filter is only 0.72 (this work). I wonder what happens if the top filter pack, the borosilicate/glass, is doubled. Obviously the transmission goes down and the asymmetry parameter is getting smaller? So what happens if the filter is loaded with particles can we simply weigh the g filter and g aerosol with the optical depth. I just don't know if equation 4 is applicable here. I do not say it isn't, I cannot judge. Can I calculate g pristine filter form numbers in Figures 5?

Reply:
The authors can't follow the analogy with droplets and doubling the filter pack. One easily can prove that strong forward scattering (g= 1) leads to following forms of equations (6) and (7) :

$$\lim_{g \to 1} R = 0$$

$$\lim_{g \to 1} T = \frac{1}{e^{(1-\omega)\delta_e/\mu_1}} = e^{\delta_a \mu_1}$$

The interpretation then is: no backscattering occurs and the transmittance is the Lambert Beer Law for absorbing only particles. Since all light is scattered in the forward direction the scattering does not contribute to the reduction of the transmittance. The factor μ_1 accounts for the elongation of the pathlength due to multiple scattering. In the case of g=1 it is obvious that the value of μ_1 must be close to unity for a collimated incident light beam. But if the incident light is diffuse, the situation is different. The effective light path in the medium becomes longer compared to a perpendicular incident light beam because of the inclined lightpath. Therefore, the factor μ_1 reflects properties of illumination and the internal scattering. This thought experiment shows that one can not argue with simple models, e.g. droplets with g=1. For an understanding of the complicated relation between g and μ_1 we refer to Literature (e.g. Lyzenga, D. R.: Note on the Modified Two-Stream Approximation of Sagan and Pollack, Icarus, 19, 240-243, 1973).

The asymmetry parameter can't be calculated from data shown in figure 5, since we already derived two parameters (scattering and absorption optical depth) from the two measured parameters. The asymmetry parameter is an estimation considering the value given in Moteki et al. and scattering calculations. The resulting value is 0.75 (value is given at page 11103 line 27). Details are given in section 3.2.

Certainly, weighting the asymmetry parameters instead of the phase function is a simplification. This is necessary since the phase function is unknown. We will add this to the text.

Comment: Page 11106 line 17.
Apparently, the factor "mu" is chosen 1/sqrt3 whereas in Moteki it was 1, it is not clear why this is chosen but effectively this difference implies almost a factor of 2 in the total optical depth in equations 6 and 7. "The transmittance calculated from Moteki is an order of magnitude smaller than observations for this study". Choice of the value of mu is suggested as reason, the impact of this choice is easily verified and should be done. Also the asymmetry parameter is suggested as explanation, but Moteki's asymmetry parameter is larger whereas the calculated transmission is smaller, right?
Reply:
The question for the factor μ_1 was answered in the context of previous questions and the extreme case of unity was rejected. Unfortunately we can not reproduce the values given in Moteki et al. (2010), as already stated on page 11103 lines 18ff. Furthermore is seems to us very unlikely that the filter consists of fibers of diameter

of 0.5 µm. Electron Microscope pictures as shown in fig. 2 in Submaranian et al. (2007) suggests that fibers can have much larger diameters than 0.5µm. Therefore we suppose that the filter properties derived from scattering calculations and the assumption of μ_1 =1 might result in high uncertainties of the filter optical depth. Deriving filter parameters from measured transmittance and reflectance seems to us to be the better way.

Comment: Page 11106 line 27.
"With this value the calculated enhancement factors are in good agreement with the enhancement factor for the PSAP given in Bond et al." For what experiment or situation? Bond enhancement parametrisation is used as model constrain. How independent is the observation of "good agreement"? Please elaborate a bit on this.
Reply:
η_f was estimated from matching the model to both Bond et al. (1999) and Virkkula et al. (2005). Furthermore we will refer to Fig. 8.
We will change the text to: "… With this value the calculated enhancement factors are in good agreement with enhancement factors from the PSAP given in Bond et al. (1999) and Virkkula (2005). The comparison is shown in in Fig. 8."

Comment:
Section 3.1 line 21 "such a model". Many similarities with CTS but what is new here as compared to Arnott 2005 (except applied aethalometer)
Reply:
This question was answered in the context of a previous question.

Comment:
Comment: Page 11109 Do I understand correctly that the calibration experiment for black aerosol is assumed to be not scattering when it comes to sensitivity calculations? Would that mean that some scattering effects are "calibrated-in"?
Reply:
The parameterizations given in B1999 and V2005 were derived from slightly scattering aerosols (soot). But the corrections explicitly include a correction term for scattering. Setting the scattering coefficients and single scattering albedo to '0' the resulting filter transmission functions are for hypothetically purely absorption particles. So, there is no scattering effect 'calibrated in'

Comment:
Equations 33 and 34. I cannot rewrite equation 33 in terms of relative optical depth (34) appendix B. Possibly this is related to "that are not yet understood" line 5 page 11114 but I did not check.
Reply:
There are some errors in the equations.
There is an error in eq. (32). The correct equation from Virkkula et al. (2005) translated to the notation in this manuscript is:

$$f_{tr,V2005}(\tau,\omega_0) = c_1 + c_2\left(h_0 + h_1\omega_0\right)\ln(\tau) - s\,\frac{\sigma_{sp}}{\sigma} \qquad (32)$$

Equations (33) is correct, but in equations (34) and appendix (B5) a sign error occured. c2 must be replaced by -c2. The corrected equations are:

$$\delta(\delta_{ap}) = \sqrt{\left(\frac{c_1}{c_2 h_2}\right)^2 - \frac{2\delta_{ap}(\delta_{ap})}{c_2 h_0}} + \frac{c_1}{c_2 h_2} \quad (34)$$

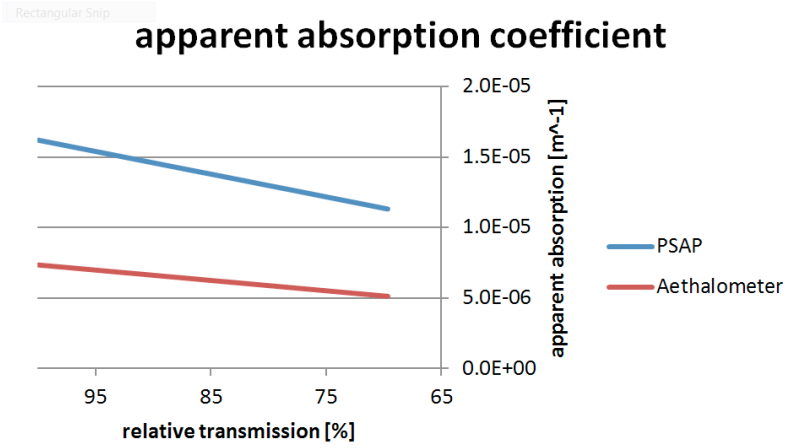
$$\delta(L) = \sqrt{\left(\frac{c_1}{c_2 h_2}\right)^2 - \frac{2\delta_{ap}(L)}{c_2 h_0}} + \frac{c_1}{c_2 h_2} \quad (B5)$$

The sign error only occurs in the manuscript. The used computer code was not affected, and results do not need to be recalculated.
A supplementary file with a complete revision of the appendix is attached. Some more steps are included to make it easier to follow the derivations.

Comment:
Page 11115 on the parameterization given. A five(!) parameter fit is compared with the data the fit is based on. The data in Fig 9b seems not to be spread nicely around zero. More importantly the cases with small asymmetry parameters, thus higher scattering exponents, thus small particles are not well described by the parameterization, i.e. measured 20% too high. A satisfactory explanation for this behavior was not found. Could deeper penetration partly explain the stronger enhancement for small particles?
Reply:
Unfortunately the authors did not find a simpler function for parameterization. Furthermore the choice of the function was little motivated by: 1) having a constant, 2) having a term with a linear dependence of the asymmetry parameter, and 3) having an term (exponential function in Eq. 36) for describing a dependence on loading. Finally, we haven't found a better function and the five parameters are really needed.
The reviewer is right. Beside systematic uncertainties of the asymmetry parameter a penetration depth effect is also possible. We will include that suggestion in the text.

Comments:
Typos
Start aethalometer with capital A?
Page 11094 line 18 "if" replaced by "whether"
Page 11108 line 14 "G" must not be capital.
Page 11108 limited by?
Reply:
Typos will be corrected.

Figure of the reviewer: The figure was commented in a paragraph before.



Further literature:
Lyzenga, D. R.: Note on the Modified Two-Stream Approximation of Sagan and Pollack, Icarus, 19, 240-243, 1973.

Attached supplementary file:
Revised Appendices A and B
Figure illustrating the principle of CTS (c.f. comment of reviewer 3).