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.

Referee 2:

Authors' comment: We thank the referee for reviewing this article and commenting it. We would particularly thank for careful reading and pointing to errors and inconsistencies in the equation.

General comments:
This paper proposed an interesting way to obtain the particle absorption coefficient from filter based measurements such as PSAP. This is an important topic in many aerosol related fields. Due to the simultaneous presence of absorption and scattering, it is challenging to accurately separate absorption from scattering in this type of measurements. The authors developed a novel correction method aiming at obtaining accurate aerosol absorption coefficient from filter based measurements. The development of the novel correction method seems rigorous. However, two key equations, Eqs. (31) and (33), were found to contain errors, which casts doubt on the correctness of some the results presented in this paper. Details will be given later.

Reply to reviewer:
Thank you for your very careful review of our paper, and for the comments. In the derivations of equations some errors occurred. The final equations we used are correct and the results do not change. To make it easier to follow the derivations in the Appendix and because of some typos in the equations, a revised and corrected Appendix is attached as a supplementary file.

Comment:
In Eq. (5) on page 8, the summation is over all particles. For individual particles the concept of scattering coefficient does not apply. The average g should be weighted by particle scattering cross section.
Reply:
We agree with the reviewer. We will replace the scattering coefficient by the scattering cross section in equation 5. To introduce the cross section we will change the following sentence to: "… where *i* denotes the *i*th particle with scattering cross section C^i_{sca}. The scattering coefficient and scattering coefficient of a potpourri of particles are related by $\sigma = C_{sca}\,N$, where *N* is the number concentrations of particles.

Comment:
Below Eq. (5), there is a typo in the transmittance, $T = I_t/I_r$.
Reply:
Will be corrected.

Comment:
At the beginning of page 10, it is commented that Eq. (11) is symmetric. Eq. (11) possesses the symmetry only if $R_1 = 1-T_1$, i.e., both filter layers are nonabsorbing. This point should be made explicitly.
Reply:
We thank the reviewer for this correction.
We will change the first sentence of the paragraph to: "For nonabsorbing layers with R=1-T Eq. (11) is symmetric with respect to the order of the layers. In section 3.2 we show that the PSAP filter is only slightly absorbing, and Eq. (11) can be though to be symmetric. That

implies that it doesn't matter from which side the filter is illuminated…"

Comment:

In this paper $\boldsymbol{\mu_1}$ *is assigned a value of 1/ 3 (Table 2) without any justification. The relative optical depth of a particle-loaded two-layer system given in Eq. (12) is a very important quantity in this paper. It is dependent on the particle and the filter properties as well as the particle concentration profile through the relative particle penetration depth* $\boldsymbol{\eta_f}$*. Near the end of page 14, a value of 0.2 was chosen in this paper to match the enhancement factor of Bond et al. (1999). Although it is not clearly indicated in the paper, I assume that* $\boldsymbol{\eta_f} = 0.2$ *was assumed throughout this paper. By fixing the value of* $\boldsymbol{\eta_f}$ *it is effectively assumed that particles do not penetrate further into the filter, see Fig. 2. Is this realistic? In reality should* $\boldsymbol{\eta_f}$ *vary with time?*

Reply:

μ_1 is indeed a not motivated parameter. The value varies in literature by a factor of about 2. A study of the literature finally led a to a sufficient explanation, why we have chosen a value of $1/\sqrt{3}$. The text following to Equation (8) will be modified to:

"… μ_1 typically is between unity and $1/\sqrt{3}$ for diffuse light propagation (Chandrasekhar, 1950; Sagan and Pollak, 1967; Liou, 2002). The value of μ_1 accounts for the elongation of the path length in the medium because of multiple scattering. In Equation 6 and 7 one can see that μ_1 scales the extinction optical depth. Values near unity are limiting cases where multiple scattering is negligible and does not contribute to a path length elongation. It easily can be derived that for this case ($\mu_1 \rightarrow 1$) and negligible backscattering ($g \rightarrow 1$) the equation for the transmittance simplifies to Lambert Beer law with only having the absorption optical depth in the exponent ($T = e^{-\delta_a}$). A value of $\mu_1=1/\sqrt{3}$ was proven to be a good value for cases with considerable multiple scattering (Sagan and Pollak, 1967, Lyzenga, 1973), supporting the value of $1/\sqrt{3}$ used here, and which also was used in Arnott et al. (2005). "

The dependence of μ_1 on particles and the assumption of a constant step function for the concentration profile require some more discussion. A paragraph at the end of section 3.1 will be included:

"The two stream model is based on few assumptions, since a rigorous solution of the general radiative transfer equation is impossible. The parameter μ_1 was motivated by the elongation of the light path in a multiple scattering environment. In our model μ_1 is used for describing the optical properties of the blank filter. Effects of particle loading on μ_1 are desirable and would be coupled to the relative penetration depth η_f. Unfortunately loading effects on μ_1 and η_f can not handled by the model. The assumption of a step function of the particle concentration across the filter is not realistic. From sampling theory the particle profile should be exponential decreasing in regions of constant collection efficiencies. Furthermore the collections efficiency will change with time because of previously collected particles. A compensation of these model weaknesses by matching the model to experimental calibration functions is shown in section 3.3."

A Further paragraph at end of section 3.3 will be:

"It is assumed that the sensitivity functions from calibration experiments for absorbing and scattering particles implicitly include filter sampling artifacts. The radiative transfer model is not able to handle these artifacts, but the CTS algorithm inherently compensates for sampling artifacts from the experimental calibrations correction."

Comment:

It is not clear to me how to understand Eq. (22). To obtain F_{fmod} *from Eq. (22) it is necessary to know* $\boldsymbol{\delta}$ *(the denominator). My understanding is that* $\boldsymbol{\delta}$ *in the denominator of Eq. (22) is also calculated or modeled using Eq. (12). If this is indeed the case,* $\boldsymbol{\delta}$ *in Eq. (22) should be* *written as* $\boldsymbol{\delta}_{mod}$.

Reply:

The reviewer is right, it should be the modeled relative δ^{mod}. We will correct Eq (22).

Comment:

The derivation of Eq. (31) given in Appendix A contains errors. By following the derivation given in Appendix A I arrived at

$$\delta(\delta_{ap}) = \ln\left(\frac{(c_1 + c_2)e^{c_2\delta_{ap}} - c_1}{c_2}\right) \qquad (31)$$

Reply:

The equation derived by the reviewer (equation above) and Eq. (31) of the manuscript

$$\delta(\delta_{\mathrm{ap}}) = \ln\left(\frac{e^{c_2\delta_{\mathrm{ap}} + \ln(c_1 + c_2)} - c_1}{c_2}\right)$$

are equivalent. Using the product rule the term (c_1+c_2) can be written as a multiplicative factor to the exponential term or as summand in the exponent.

Comment:

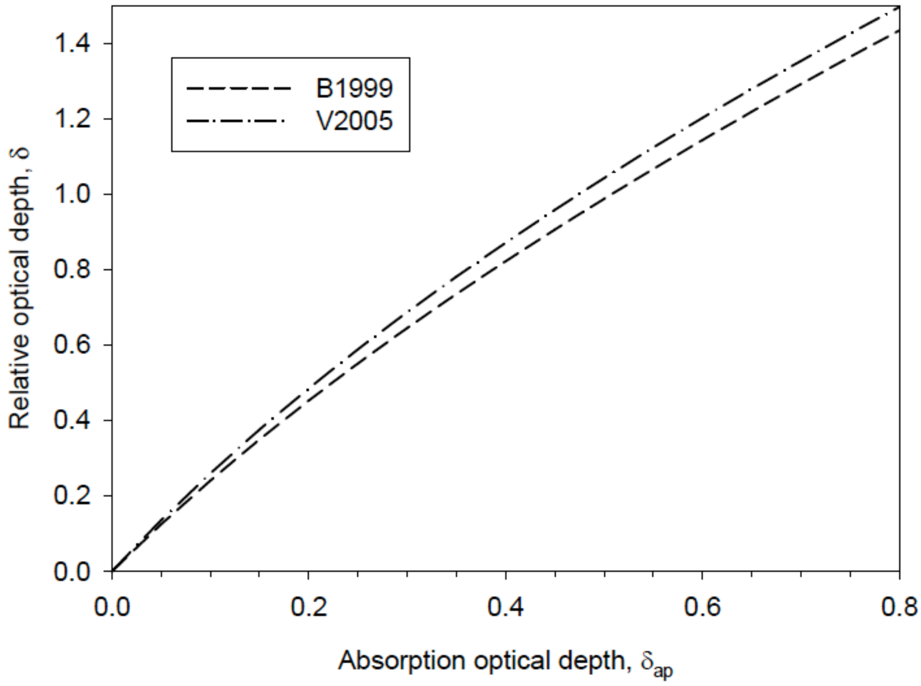
The derivation of Eq. (33) from Eq. (32) for black particles is also incorrect. For black particles Eq. (33) should be

$$f_{tr,V2005} = c_1 - c_2(h_0 + h_1)\ln(\tau) \qquad (33)$$

It is therefore expected that Eq. (34) is also incorrect. From following the steps of derivation given in Appendix B, but starting from Eq. (33) given above, I arrived at the following expression for Eq. (34)

$$\delta(\delta_{ap}) = \sqrt{\frac{2\delta_{ap}}{-c_2(h_0 + h_1)} + \left(\frac{c_1}{c_2(h_0 + h_1)}\right)^2} + \frac{c_1}{c_2(h_0 + h_1)} \qquad (34)$$

By using Eq. (31) and Eq. (34) given above for B1999 and V2005, respectively, and the parameters given in the paper Fig. 8 is revised as follows



The B1999 curve does not change too much from that given in Fig. 8 of the paper. However, the V2005 curve changes significantly from that shown in Fig. 8 of the paper. In addition, the two curves are very close to each other. Since the errors in Eqs. (31) and (34) of the paper are critical to the rest of the paper, some of the results should be re-calculated using the correct expressions of these equations.

Reply:
There is an error in eq. (32). The correct equation from Virkkula 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 occurred. c_2 must be replaced by $-c_2$. The corrected equations are:

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

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

The sign error only occurs in the manuscript. The used computer code was not affected, and results do not need to be recalculated.

Further Literature:

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

Further corrections:
Appendix A:

In equation (A3), (A4), (A5) the variable of integration will be corrected ($\frac{d}{dl} \rightarrow dl$).

Furthermore: " Substituting $x(l) = c_1 + c_2^{\delta(l)}$ …" will be corrected to $x(l) = c_1 + c_2 e^{\delta(l)}$

To make it easier to follow the derivation we included one more step in (A5)

More steps were included between (A6) and A(7).

Appendix B:

Typos were corrected (h_2 is replaced by h_0)

Attached supplementary file:

Revised Appendices A and B

Figure illustrating the principle of CTS (c.f. comments of reviewer 3)