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Atmospheric Measurement Techniques Discussions

Interactive comment on "Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms" *by* M. Collaud Coen et al.

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Answers to W. P. Arnott:

The authors thank W. P. Arnott for his valuable comments. He pointed out specificities of the filter-based absorption measurements and particularly of the Aethalometer, that can be more precisely described in the paper. Most of these points have been analyzed during this study, but were not always extensively described in the paper.

1. First, the authors ignore one very important fact of the Aethalometer: It has a variable starting point for filter transmittance because the instrument conditions the

new filter spot using ambient air (discussed in the Arnott 05 paper).

REPLY: The Aethalometer has a filter-preconditioning cycle after each tape change that exposes the filter to sampled air before the measurement start. In rack mount Aethalometers (AE16, AE21, AE22, AE31) the sample flows through the filter tape for a part of the preconditioning cycle only (about 3 minutes), and the flow is diverted through the by-pass cartridge filter during most of the preconditioning cycle (G. Mocnik, Aerosol d.o.o., Magee Scientific, personal communication). This preconditioning cycle can modify the zero point of the light intensity I0 (Eq. 1) and consequently the attenuation ATN. As also stated in Arnott et al. (2005), this is normally not an issue for ambient measurements as reported in this study, but it could be an issue when sampling from highly polluted sources. A different starting point for light intensity can affect the filter loading correction 1/R for the Weingartner correction and for the new correction, which directly uses the attenuation (ATN), and the determination of the calibration constant Cref, since the absorption coefficient corrected for the filter-loading artifact is used to calculate Cref. The shift of the starting filter transmittance will induce an increase of the 1/R values that is greater for low attenuation in the case of the Weingartner filter-loading correction and for high attenuation in the case of the new filter-loading correction. As stated in the paper (§3.3), the filter-loading correction only slightly contributes to the total correction (2-4 % of the total correction), so that the variable starting point for filter transmittance has a very low impact on the total correction and cannot explain the large variations in the slope between AE and MAAP absorption coefficients. However, this higher filter loading correction also implies a higher calibration constant Cref. A first estimate leads to a mean Cref increase of 1-2 % for the Weingartner filter-loading correction and an increase of 2-5% for the new filter loading correction. A larger increase is only probable for very high attenuation (> 50%) and very low single scattering albedo (< 0.5). A better quantification of this effect is however presently not possible since the shift of the zero point of the filter transmittance is presently not known for any of the stations. This is however an important point to better understand the Aethalometer operation mode and the necessary corrections.

The filter preconditioning cycle will therefore be mentioned in the experimental part of the revised manuscript and the resulting consequences will be discussed in the result section.

2. The Arnott 05 paper gives a prediction for a scattering correction based on aerosol scattering and asymmetry coefficients (see Eq. 15). Scattering corrections depend on scattering optical depth and asymmetry parameter, so comparisons with TSI neph estimated asymmetry parameters should also be considered.

REPLY: I compared the obtained new and the slope between AE and MAAP absorption coefficient with the asymmetry parameter. No correlation was however observable for any of the parameters and for any of the stations as reported under §3.3. I also tried to include the asymmetry parameter to weight the scattering correction as well, but the obtained results were not satisfactory at all. Since an effect of the asymmetry parameter on the scattering correction seems obvious, I will add in the revised manuscript some more information on the unsuccessful attempts to include the asymmetry parameter in the Aethalometer correction. 3. The authors do not mention relative humidity. Were all measurements performed at low RH? RH is known to cause serious issues with PSAP absorption measurements. REPLY: Information on relative humidity is given in Table 1, where all the details on the measurement sites are summarized. JFJ and HOP have heated inlet that ensure low RH in the instruments. CAB has also a heated inlet since April 2008. The humidity percentage can have a great impact on the multiple scattering constant Cref determination as explained under 3.1.2. I studied the relation between the relative humidity measured by the Nephelometer and Cref in order to explain the different Cref values encountered at the different stations and their seasonal cycle, particularly at JFJ and CAB. However the RH is not the key parameter explaining the variability in Cref values, so that I did not mention it explicitly in the paper. Further clarifying information on this topic will however be given in the revised manuscript.

4. With a plethora and proliferation of choices for corrections to the filter based methods, is it not better to simply divide ATN by one number for each wavelength and call it

good? All the corrections have advantages and disadvantages, and require a lot more analysis. I find that when averaged over sufficiently long periods to allow for aerosol uniformity and at least on full cycle of aerosol measurement between new and loaded filter, the averages from the Aethalometer compare well with reference methods. The Aethalometer does not do well with plumes and very low single scattering albedos (below 0.5).

REPLY: To find a simple correction that accounts for the main AE artifacts as proposed seems to be a very good idea. Anyhow, as long as the difference between all the measured Cref cannot be explained, the determination of these numbers for each wavelength will remain a problem. Furthermore much care has to be taken in order not to falsify the real wavelength dependence of the absorption coefficient by choosing wrong wavelength dependencies for these constants. However, this study tries to apply theoretically based corrections to the known AE artifacts and allows an assessment of the performance of already published AE corrections.

5. The MAAP is taken as a reference method, and the Aeth. is interpolated to its wavelength (which Aeth. measurements were used – which wavelengths?). Is the MAAP a reference method? As a filter based method, it suffers from RH effects and from unknown particle penetration depth in the filter media.

REPLY: As also stated by the first anonymous referee, the MAAP is not an absolute reference method. No absolute reference method measuring the aerosol absorption coefficient at relatively low concentration levels is however presently available. All the filter based methods (which have typically a very low measurement detection limit) suffer from several artifacts linked to the use of the filter, but the other absorption measuring methods suffer from other artifacts. Similarly each of the methods also has some limitations in their applications. However, MAAP has already resolved some of the filter based artifacts and leads therefore to a more reliable measurement than the Aethalometers. Its fairly widespread use over the past several years also permits the authors to find 4 datasets allowing the comparison for sufficiently long measuring peri-

ods of both measuring methods for ambient aerosol. However, the authors completely agree with the fact that MAAP is not yet an absolute reference method and this will be explicitly cited in the revised paper.

6. In the Arnott 2005 paper, it was acknowledged that the fit parameters were site dependent and filter starting point dependent. The intention of that paper was to develop a formalism based on multiple scattering theory. The authors, and others, are encouraged to explore their own choices for the empirical fitting parameters.

REPLY: It is mentioned under §2.2.2 that the constants given in Arnott et al. (2005) are not applicable to all datasets and cannot be therefore employed without further analysis of the aerosol types.

7. The Aeth would be improved if it conditioned after filter change using filtered ambient air. It would also be improved if a thermal denuder was used up front to remove most of the scattering aerosol and particle bound organics and inorganics. Then many of these issues would be mitigated, assuming the primary absorber is elemental carbon. However, as is now becoming clear in the work of Subramanian and others, liquid phase aerosol light absorption is going to be a real challenge for filter based measurement methods.

REPLY: The reviewer is completely right, but the aim of this paper is not to modify the AE instrumental procedure, but to test all the proposed AE corrections on long operational datasets of ambient aerosols in order to take advantage of already existing long-term data sets.

Interactive comment on Atmos. Meas. Tech. Discuss., 2, 1725, 2009.