Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2016-294-RC2, 2017

Atmospheric Measurement Techniques Discussions
Open Access
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Interactive comment on "On Aethalometer measurement uncertainties and multiple scattering enhancement in the Arctic" *by* J. Backman et al.

Anonymous Referee #1

Received and published: 29 January 2017

The work presented in the manuscript is very important as it introduces a new technique for post-processing filter photometer data, focusing on the Aethalometers in the Arctic. The novel approach synthesizes previous work on the reduction of instrumental noise and the associated detection limit. The manuscript is detailed and presents the post-processing approach in a way that will render the methodology useful for users of different filter photometers: Aethalometers, PSAPs, CLAPs... The approach is novel in that it allows averaging the data to obtain constant relative uncertainty by changing the averaging window. It also shows the very low uncertainties which can be obtained using this methodology. The presented work is an important contribution to the interpretation of Arctic and global measurements of aerosol absorption. In addition, the

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authors report the relative scaling factor between the raw Aethalometer measurements of the attenuation coefficient and the processed PSAP/CLAP/MAAP measurements, reporting the absorption coefficient. They report this as the scattering enhancement factor.

There are terminological and methodological issues that need to be accounted for prior to publication in AMT.

Once the major and minor issues are addressed, the manuscript would make a perfect addition to the compendium of filter photometer related literature in AMT.

The measurement in filter photometers such as Aethalometer, PSAP and CLAP is one of transmission of light and the determination of the change of attenuation (ATN). Then the attenuation coefficient is calculated and the eBC concentration is derived from this coefficient using the mass attenuation cross-section. While the authors use the proper term “mass attenuation cross-section” in the text, they use the term “uncorrected light absorption coefficient (sigma_0)” (starting on p. 5, line 20). This is inaccurate – the quantity is the “attenuation coefficient”, this quantity is then post-processed for loading effects and divided by the multiple scattering coefficient C to obtain the “absorption coefficient”. This procedure is based on assumptions which need to be tested to the greatest extent possible.

The authors use the PSAP, CPAL and MAAP as “reference” instruments. The claim of “reference” is not substantiated. The paper very clearly delineates the way to obtain the factor C, but this can be interpreted just as the relative normalization factor to harmonize the determination of the absorption coefficient from different filter photometers. And here lies the crux of the problem: all instruments which are being compared are filter photometers and the principle of operation for most of them is nearly identical. The claim of C being interpreted as the “multiple scattering enhancement factor” needs to be further substantiated. Since no non-filter method was available, the methodology needs to be proven at least internally consistent.

The determination of the absorption coefficient necessitates the determination of the multiple scattering parameter C (Weingartner et al., 2003). The parameter C is to a degree arbitrarily separated from the loading effects, which influence the determination of the absorption coefficient as well. If C is to be the parameter describing the multiple scattering effects in the filter matrix, it should not depend on ATN. This can be considered to be the "proof" of the separation of the multiple scattering from the loading effects (the Weingartner et al. parameter R). The authors show that C does depend on ATN (Fig. 8, p. 29). The authors correctly point out that the existing post-processing algorithms do not necessarily ensure the lack of dependence of C on ATN (Collaud Coen et al., 2010), however for background sites, the loading effects are most probably non-existent (Virkkula et al., 2015; Drinovec et al., 2016). The post-processing algorithm needs to be site specific, as the loading effects are a function of the entire loading of the sample spot and the physical and chemical properties of the entire sample deposit. This could be the reason of the difference between Summit and other Arctic sites.

The reason for the C dependence on ATN can be due to the non-compensation of the Aethalometer data (even though this is questionable for global background sites; Virkkula et al., 2015; Drinovec et al., 2016) or the loading effects in the so-called "reference" instruments, which are known to feature loading effects or saturation (Bond et al., 1999; Virkkula et al., 2005; Hyvärinen et al., 2013). The authors correctly identify this weakness of the presented work in the beginning of section 4.2 when they mention that the C they report is essentially a slope between attenuation and absorption coefficients determined with different filter photometers.

The authors need to present the criteria for the "goodness of evaluation" of loading effects in all filter photometers. They have already used the way to go about this (when discussing C): the aerosol absorption should not depend on ATN for measurements in all filter photometers. This analysis should be added to section 4.2 and discussed: the plot of sigma_abs=sigma_abs(ATN) for all sites. The relationship between the C and the scattering coefficient should be reported – do scattering particles in the filter

increase the attenuation coefficient?

In addition to this, the Aethalometers are compared to different instruments: two different versions of PSAP, CLAP and MAAP. The authors need to substantiate that the comparison of the Aethalometers to these different instruments is relevant. No comparison between the PSAPs, CLAP and MAAP is reported. The authors should at least sum up the results of laboratory inter-comparisons if no comparisons for ambient Arctic measurements is available.

Specific comments

Page 2, line 22: “below the detection limit”. As the authors later point out, the detection limit is a function of the time between two consecutive measurements and the averaging time. One can lower the detection limit by integrating the sample for a longer time. The sentence needs to be modified or the time resolution (5 min?) needs to be specifically mentioned.

P. 3-5: it would be a good idea to report the inlet cuts (PM2.5. . .), flows (or face velocities), operational wavelengths of the filter photometers for all sites. The conditions for triggering the change of tape should also be reported (8 hours in Pallas, for example, elsewhere an ATN limit).

P. 5, l. 14: “Initially, when no aerosol particles have been deposited onto the filter, light is transmitted through the filter with an intensity I_0.” This is not true, the Aethalometers measure I_0 (intensity of light transmitted through the reference part of the filter without any sample) at the same time as the intensity of light I transmitted through the sample. Please change.

P. 5, l. 20 and repeated later: “uncorrected light absorption coefficient (σ0)”. This is the “attenuation coefficient”, please see above. Please change accordingly.

P. 5, l. 31: “14625/λ m2g–1”. Please use unitless expressions, for example “16.6 m2g–1 at 880 nm, scaling inversely with the wavelength” or something similar.

P. 7. Section 2.4: the comparison of the so-called “reference” instruments needs to be elaborated (please see above). Has there been an intercomparison of the instruments in question? In ambient conditions or in the laboratory? How are published results of other intercomparisons relevant for the results reported in the manuscript?

P. 8., l. 9: is the 2% relative uncertainty for spot area specific to Aethalometers or to all filter photometers. Can the diameter be measured to 0.1 mm? How do relative and absolute uncertainties of the “reference” filter photometers influence the results?

P. 8, l. 19: “The one wavelength Aaethalometer at Summit was interpolated from 880 nm to 637 nm using a α of –1 in Eq. (5).” Please correct the typo “Aaethalometer” -> “Aethalometer”. What is the real value of the absorption Angstrom exponent α? Could the lower C determined at Summit be (partly) due to the systematic bias due to the extrapolation from 880 nm to 635 nm using an α value that is too small? Are there any multi-wavelength measurements of absorption at Summit (sat least a short time series)?

P. 9, l. 22: “Moreover, lateral flow can influence both the signal and reference detectors, and thus ATN, through deposition of aerosol particles that do not originate from the sample air stream.” This is highly unlikely, as the particles will get filtered out at the edges of the filter material, not above the light detectors in the filter photometers. Please substantiate the claim or remove the sentence.

P. 10-11, Fig. 1: The sources of the drift in the laboratory experiment with the absolute filter are intriguing. The authors should at least offer some hypothesis on them and comment whether they are relevant for ambient measurements. How does the additional pressure drop due to the filter influence the measurements? Does it in fact introduce additional drift? Are there jumps or transients when tape is moved and measurements restarted? Do semi-volatile organic compounds adsorb on the filter and cause a signal, appearing as drift? Is the air conditioning in the laboratory important? Does drift have a wavelength dependence (indicating sample deposition rather than a

fixed electronic drift)?

P. 11-12, Fig. 3: Was the absolute filter attached to the Aethalometer or to the sampling line leading to the Aethalometer? Were other instruments attached to the same sample line? Were there any pressure drops, jumps, transients in the sample line, or events in the measurement room, resulting in movement of the filter in the Aethalometers? Any movement or drift in the filter position influences the measurement of ATN. The authors need to comment on these possible sources of drift and the observed transients.

P. 13, l. 32-33: "Thus, the drift uncertainty seen in Fig (3) becomes 0.003–0.03 Mm–1 after multiple scattering correction is applied." The effect of the reduction of noise on the uncertainty of C and the effect of thus derived C on the uncertainty of the absorption coefficient should be presented more clearly, starting perhaps here.

P. 14, Table 4: The determination of the C within ACTRIS "grey literature" reports would be a valuable reference here.

P. 14, l. 22-25: "Third, it has to be acknowledged that there can be a bias in the absolute Cref values because of imperfect corrections of filter artefacts in the reference instruments (Backman et al., 2014; Müller et al., 2011). However, this bias should not substantially alter the ATN dependency because filter changes were not performed in sync." This is an oversimplification. At constant concentration and equally spaced movements of tape (but not synchronized, and dependent on the flow of the individual instruments) an back-side-of-an-envelope calculation shows that the effect can be up to 20%. This is unlikely for global background sites in the Arctic, but this needs to be shown at least in the Supplement of the manuscript.

P. 15, Figure 9: The regression equation on the figure is missing sigma_ap as the independent variable, making it confusing for the reader.

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