Calibration of Aethalometers with Different Soot Aerosols

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Introduction

The characterization of carbonaceous particles is a very active field, especially with respect to its effects on human health and atmospheric radiative properties. The aethalometer [1] is the most frequently used technique for measuring so-called black carbon (BC) mass concentrations in real-time. In the last years, new aethalometers have been developed operating with several light sources with narrow bandwidths ranging from the near ultraviolet to the near infrared.

Aethalometers continuously sample aerosol particles on a quartz fiber filter and measure the attenuation (*ATN*) of light transmitted through this filter. The change of *ATN* is then converted into the aerosol absorption coefficient (b_{abs}). Due to the interactions of the incident light within the fibre matrix, the precise determination of b_{abs} is a difficult task. The aim of this work is to present correction factors for the use of the aethalometer [2].

Definitions

The **absorption coefficient (b_{abs})** is defined with Beer-Lambert's law

$$I = I_o \cdot e^{-b_{abs} \cdot x}$$

I_o is the intensity of the incoming light
I is the remaining light intensity after passing through a medium with the thickness *x*.

The **attenuation (*ATN*)** is typically given as percentage values and is defined by

$$ATN \equiv \ln\left(\frac{I_o}{I}\right)$$

According to the Beer-Lambert's law, the aerosol absorption coefficient of the filtered aerosol particles b_{ATN} **(=attenuation coefficient)** is defined as

$$b_{ATN} \equiv \frac{A}{Q}\frac{\Delta ATN}{\Delta t}$$

A is the filter spot area,
Q is the volumetric flow rate
ΔATN is the change in *ATN* during the time interval Δt

Empirical Model

It is well known that b_{ATN} differs significantly from the „true“ aerosol absorption coefficient b_{abs} of airborne particles. Therefore, the calibration factors *C* and *R(ATN)* are introduced:

$$b_{abs} = b_{ATN}\frac{1}{C \cdot R(ATN)}$$

C is > 1 and describes the **multiple scattering** of the light beam at the filter fibers in the unloaded filter.
R accounts for the "**shadowing effect**", which is caused by a reduction of light intensity in the filter by the embedded light absorbing particles. *R* varies with the amount of particles embedded in the filter.

Calculation of BC Concentration

The exact knowledge of *C* and *R* is also of importance if the attenuation data is converted to BC mass loadings. The aerosol black carbon mass concentration M_{BC} (in units of g m^{-3}) is related to the absorption and attenuation coefficients by

$$M_{BC} = \frac{b_{abs}}{\sigma_{abs}} = \frac{b_{ATN}}{\sigma_{ATN}\, R(ATN)}$$

σ_{abs} and $\sigma_{ATN} \equiv \sigma_{abs} \cdot C$ are the mass specific absorption and attenuation cross-sections (in units of m^{2}g^{-1}), respectively.

Experimental

The calibration experiments were performed during an extensive soot aerosol campaign in the AIDA aerosol chamber of the Research Center Karlsruhe in 1999. During this campaign the optical properties of artificially generated soot, Diesel engine soot, ammonium sulfate aerosol particles, and mixtures of the above components were characterized. b_{abs} of the filtered particles was measured with two different aethalometers (an AE10 operating with white light (broadband) and an AE30 with multiwavelength capability). Parallel to these absorption measurements, a reference value of b_{abs} was determined by measuring the extinction (b_e) and scattering coefficient (b_s) of the airborne particles with an extinction spectrometer and a three-color integrating nephelometer.

Conclusions

- With increasing filter load the optical path in the aethalometer filter decreases. As a result, an increased underestimation of the measured aethalometer raw signals occurs with increasing filter loads. This artefact is very distinct for "pure" soot particles where a filter change causes an apparent increase of the measured signals by a factor of about 1.6.
- Provided that the proper numerical corrections are performed, the AE30 instrument allows for the measurement of the aerosol light absorption coefficient (b_{abs}) or of the black carbon (BC) mass concentration over a wide spectral range (λ = 450 – 950 nm). The empirical corrections require information on the light scattering behaviour (i.e. light scattering coefficient) of the sampled particles.
- The determination of b_{abs} with the AE10 aethalometer is a difficult task because of an ill-defined spectral sensitivity of this instrument.
- Another instrumental artefact was observed when high concentrations of semi-volatile gaseous species adsorbed on the aethalometer filter fibers and thus enhanced multiple light scattering. Therefore, denuders are required in front of the aethalometer in cases where an aerosol with relatively high concentrations of condensable gaseous species is sampled.

Results

Parameterisation of the "Shadowing Effect"

During all experiments it was found that the uncorrected aethalometer signals decrease with increasing particle load on the filter (see Fig. 1). This results in an increasing underestimation of the measured aethalometer raw signal with increasing filter load. This artefact is due to a "shadowing effect", which is caused by a reduction of the light intensity in the filter by the embedded light absorbing particles. The magnitude of this effect is linearly related to the single scattering albedo (defined as $\omega_o = b_s/b_e$) of the sampled aerosol, i.e. it is very distinct for "pure" soot particles (ω_o ~ 0.2) and almost negligible for the aerosol present at the high alpine site Jungfraujoch (ω_o > 0.7). The difference at the remote site is explained by the incorporation of a high amount of scattering material in the fiber matrix, which partially compensates for the "shadowing effect".

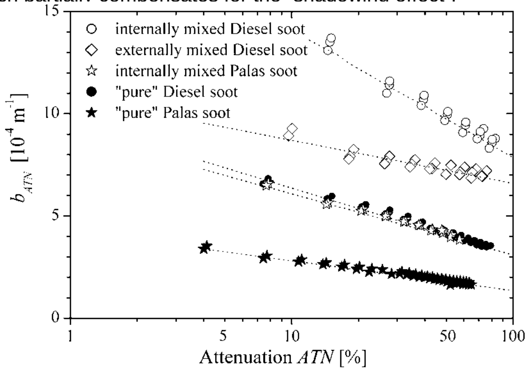

Figure 1: Measured aethalometer signals (symbols) as a function of attenuation for different aerosols. An empirical curve (dotted lines) was fitted to the data which allows a parameterisation of the observed shadowing effect.

Parameterisation of Multiple Scattering in the Unloaded Fiber (*C*-Factor)

Another important instrumental artefact is caused by multiple light scattering in the fiber matrix resulting in increased light intensity and thus in enhanced light absorption of the filtered particles. Comparisons with the reference method showed that most investigated aerosols experience an average enhanced absorption of *C* = 2.14 ± 0.21 in the filter. Significantly higher values (*C* ~ 3 - 4) were determined during experiments where soot particles were coated with organic material. Here, semi-volatile oxidation products (such as pinonaldehyde) were formed by the ozonolysis of α-pinene. The higher *C* values are attributed to a condensation artefact from gaseous species that condense on the "fresh" aethalometer filter in the first few minutes after every filter change.

Wavelength Dependent Comparison with the Reference Method

As an example, Fig. 2 shows the wavelength dependent comparison of the "true" absorption coefficient (reference) with the absorption coefficient deduced from the filter based attenuation measurement. Here, the aethalometer data were corrected for the above-mentioned "shadowing effect" and multiple scattering. A good agreement is found between the AE30 and the reference method. The ill-defined spectral sensitivity of the AE10 is reflected in an overestimation of b_{abs}.

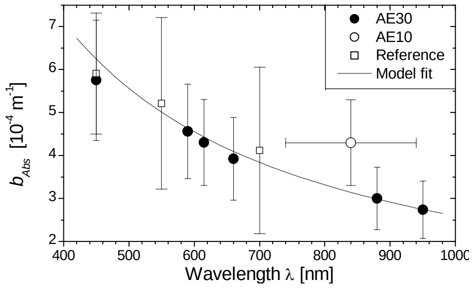

Figure 2: Comparison of the corrected absorption coefficients determined by two aethalometers with the "true" reference values for a mixture of Diesel soot and ammonium sulfate particles.

References

[1] Hansen, A.D.A., H. Rosen, and T. Novakov, The aethalometer - an instrument for the real-time measurement of optical absorption by aerosol particles, *Sci. Tot. Environ.*, *36*, 191-196, 1984.

[2] Weingartner, E., H. Saathoff, M. Schnaiter, N. Streit, B. Bitnar, and U. Baltensperger, Absorption of light by soot particles: Determination of the absorption coefficient by means of aethalometers, *J. Aerosol Sci.*, in press, 2003.