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Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops

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Abstract

Absorption photometers for real time application have been available since the 1980s, but the use of filter-based instruments to derive information on aerosol properties (absorption coefficient and black carbon, BC) is still a matter of debate. Several workshops have been conducted to investigate the performance of individual instruments over the intervening years. Two workshops with large sets of aerosol absorption photometers were conducted in 2005 and 2007. The data from these instruments were corrected using existing methods before further analysis. The inter-comparison shows a large variation between the responses to absorbing aerosol particles for different types of instruments. The unit to unit variability between instruments can be up to 30% for Particle Soot Absorption Photometers (PSAPs) and Aethalometers. Multi Angle Absorption Photometers (MAAPs) showed a variability of less than 5%. Reasons for the high variability were identified to be variations in sample flow and spot size. It was observed that different flow rates influence system performance with respect to response to absorption and instrumental noise. Measurements with non absorbing particles showed that the current corrections of a cross sensitivity to particle scattering are not sufficient. Remaining cross sensitivities were found to be a function of the total particle load on the filter. The large variation between the response to absorbing aerosol particles for different types of instruments indicates that current correction functions for absorption photometers are not adequate.

1 Introduction

Aerosols influence the radiation balance of the Earth through scattering and absorption of solar radiation. The importance of the direct effect of aerosols on climate has been pointed out by many authors (e.g. Charlson et al., 1991; Hansen et al., 1997; IPCC, 2001; Andreae, 2001). In order to study the role of aerosols on the radiation balance and reduce the uncertainties in the prediction of the direct effect of aerosols

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on climate change, field experiments have been conducted in the last decade, covering different aerosol characterization investigations at different locations (e.g. TARFOX, Russell et al., 1999; LACE 98, Ansmann et al., 2002; ACE-1, Bates et al., 1998; ACE-2, Raes et al., 2000; INDOEX, Ramanathan et al., 2001; SAMUM 1, Heintzenberg, 2009; EUCAARI, Kulmala et al., 2008). These studies reveal a large impact of aerosols on the transmission and reflection of solar radiation in the atmosphere, where scattering aerosols are responsible for the reflection of part of the solar irradiation back into space, and thus responsible for cooling because less radiation reaches the Earth surface. Absorbing aerosols may locally warm the atmosphere and influence meteorological processes and climate. The relative contributions of scattering and absorption are expressed through the single scattering albedo. There is a large uncertainty in the single scattering albedo and its global distribution. The latter can nowadays be estimated by using satellites (Veihelmann et al., 2007), but the technique is still at an early stage and relies on the determination of aerosol type from these satellite data. However, even when the aerosol type is known with some degree of confidence (Robles-Gonzalez et al., 2006), the absorption properties are poorly determined. Aerosol particles usually do not have a unique chemical composition: they may be either externally mixed as individual particles of a single composition or they may be internal mixtures of two or more major constituents with their own optical characteristics, which may not be representative of the mixture.

The use of dedicated instruments to determine the particle absorption coefficient from in situ measurements bears a large uncertainty. The aim of this paper is to determine the sources of these uncertainties through detailed analysis of systematic laboratory experiments using a representative sample of different types and makes of absorption photometers commonly deployed during field campaigns and at a range of long-term monitoring sites. This broad suite of instruments allows for multiple instrument inter-comparisons and instrument characterizations. The characterization of scattering instrumentation has been presented elsewhere (Anderson et al., 1996; Heintzenberg et al., 2006; Müller et al., 2009).

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Aerosol light absorption measurements typically show larger and more poorly understood uncertainties than extinction and scattering measurements. An important issue is the lack of a generally accepted reference or calibration standard. Network stations often rely on filter-based measurements where aerosols are collected on a fiber-filter matrix and the absorption is determined from the rate of change of light transmission through the particle loaded filter. However, it is well known that filter based techniques do not provide a true aerosol absorption coefficient and major corrections are needed. Several problems have been identified. Multiple scattering increases the optical path in the filter leading to enhanced absorption (Liousse et al., 1993; Bond et al., 1999; Weingartner et al., 2003). With increasing filter loading, the optical path in the particle loaded filter generally decreases, which effectively reduces the absorption below its true value (LaRosa et al., 2002; Reid et al., 1998; Weingartner et al., 2003). Another problem concerns particle-related scattering effects (Liousse et al., 1993; Petzold et al., 1997; Bond et al., 1999; Weingartner et al., 2003; Lindberg et al., 1999; Petzold et al., 2005). Scattering of the incident light by particles increases the filter reflectance and hence reduces the transmission through the filter, which results in apparent absorption. Other problems include ill-defined spectral sensitivities for certain types of instruments, drift of flow, spot sizes that deviate from those provided by manufacturers, etc. Since all these factors affect the results of the measurement, they need to be well-characterized and corrected for.

Dedicated instrument inter-comparisons and laboratory studies are needed to solve the problems described above and to better understand the measurements. Only a few studies have been performed and reported in the literature. In The First International Workshop on Light Absorption by Aerosol Particles, held at Colorado State University in 1980, many fundamentally different techniques measuring light absorption were compared (Gerber, 1982). After this rather comprehensive international workshop, instrument performance has been substantially improved and new instruments have been introduced. In 1999, a soot characterization experiment took place in the AIDA aerosol chamber in Karlsruhe, Germany (Saathoff et al., 2003). In that experiment,

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several different instruments for measuring carbon mass concentrations were compared using laboratory generated aerosols, e.g. diesel soot, spark generated “Palas” soot and internal mixtures of diesel soot and ammonium sulfate. The objective of the Reno Aerosol Optics Study (RAOS) conducted at the Desert Research Institute Reno in 2002, was to study the aerosol scattering, absorption and extinction under controlled conditions (Sheridan et al., 2005). The focus was to evaluate the accuracy of different measurement techniques. In 2007 the responses of four different instruments to fractal soot particles were inter-compared (Slowik et al., 2007). Another inter-comparison experiment with six different methods, including filter-based methods for measuring black carbon and elemental carbon is given in Park et al. (2006). All the above studies have the common feature that instruments from different manufacturers and/or using different techniques were compared. However, they do not provide statistics on the performance of multiple instruments of the same make and type.

In this article, results are presented from two absorption photometer workshops which were conducted in 2005 and 2007 at IfT (Leibniz Institute for Tropospheric Research) Leipzig, Germany. The first of these workshops was held in the framework of WMO Global Atmosphere Watch (GAW) and in collaboration with the EU FP6 Network of Excellence ACCENT (Atmospheric Composition Change: A European Network) in 2005. This workshop is denoted as GAW2005 throughout this manuscript. The second workshop was part of an EU FP6 Integrated Infrastructures Initiatives (I3) project EUSAAR (European Supersites for Atmospheric Aerosol Research), in collaboration with GAW and ACCENT. This workshop is denoted as EUSAAR2007. The goals of the workshops and the frameworks in which they were organized are presented in Sect. 2. In Sect. 3, the approach and experimental set up is described. The workshops focused on filter-based light absorption methods since these are widely used in the global aerosol monitoring networks, despite all the necessary known and poorly understood corrections. Most groups that run multiple stations can not afford to place expensive multiple-wavelength photoacoustic or cavity ring-down extinction instruments at all of their sites. A benefit of workshops such as these would be a better understanding of the

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filter-based instruments, with the goal of eventually being able to relate the filter-based absorption measurements to one of the more robust reference methods. The selected instruments MAAP, PSAP, and Aethalometer are introduced in Sect. 4. A rather complete instrument characterization is presented in Sect. 5. We present the unit-to-unit variability between instruments of the same make and type, after applying commonly used correction algorithms in Sect. 6. A summary of the results and the conclusions and recommendations are presented in Sect. 7.

2 Goals and objectives

The objective of EUSAAR is the integration of measurements of atmospheric aerosol properties performed in a distributed network of 20 high quality European ground-based stations. The measurements include physical and optical properties of carbonaceous aerosols. The overall objective of the EUSAAR activity on optical properties is to integrate and harmonize measurements of aerosol optical properties at the EUSAAR sites, with the outcome of having a sustainable and reliable observation network for aerosol optical data across Europe with known and high quality that are readily accessible in a common format. This requires the development of standard procedures for routine measurements of optical parameters (aerosol scattering coefficient, aerosol absorption coefficient and aerosol optical depth). Because different types of instruments are used for this purpose, it is difficult to assess the quality of the data and compare results from the various stations. Therefore, specific objectives and standard operating procedures were developed to ensure that data from the network stations are harmonized:

1. Develop a protocol providing aerosol optical measurements to ensure that they all adhere to a schedule of regular calibration and quality assurance for the relevant instruments and make data available on-line in a common format.

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2. Develop calibration procedures for the various instruments at the EUSAAR sites for measurement of aerosol optical properties of aerosol scattering and absorption coefficient and of aerosol optical depth.
3. Develop quality assurance (QA) procedures to determine uncertainties for the instrumentation and data on aerosol optical properties at the EUSAAR sites.
4. Harmonize aerosol optical property data bases which are accessible via a single webpage.

The objectives of the GAW and EUSAAR workshops discussed in this paper address specific objectives 2 and 3, for absorption measurements, in support of the EUSAAR, GAW, and ACCENT activities on a) Training and education, b) Development of QA procedures, and c) Establishing the scientific basis to provide data of high quality with known uncertainty.

The specific goals of the workshops were:

- To characterize instruments.
- To determine the variability amongst several photometers of the same type/manufacturer.
- To compare absorption photometers of different types.
- To determine the response of absorption photometers to well-characterized generated aerosol.
- To train users of filter-based absorption photometers on the use and maintenance of these instruments to obtain the optimum results.

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3 Approach and experimental set up

3.1 Tasks

The tasks to be achieved for these objectives were:

I *Instrument characterization:*

5 a) Determination of the effective wavelength for which the absorption coefficient is valid. Emitted spectral radiation of light sources in absorption photometers was measured. For broad emission spectra, the spectral sensitivity of the detector was included.

b) Determination of filter spot sizes.

10 Bond et al. (1999) observed variation in spot sizes for PSAPs and included measured spot sizes in their correction method. Actual spot sizes that differ from the spot size that is included in the instrument software, directly translates to erroneous absorption coefficients. Spot sizes (or rather spot areas) of PSAPs, MAAPs and Aethalometers were measured.

15 c) Aerosol flow characterization.

Incorrect flow directly translates to an erroneous absorption coefficient. As pointed out by Anderson et al. (1999), the effect of spot-area and flow correction is potentially larger than the corrected instrument unit-to-unit absorption coefficients. Another flow characterization concerns the face velocity, i.e. the ratio of volume flow and spot area. Face velocities were increased and lowered to estimate upper and lower bounds for optimal operation.

20 II *Determination of the variability of the results amongst several photometers of the same type/manufacturer, following correction by widely used absorption photometer correction functions.*

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III *Comparison of absorption photometers of different types* to determine differences between instruments as a consequence of their characteristics, measurement principles and corrections.

5 IV *Determination of the response of absorption photometers to well-characterized, laboratory generated aerosol and ambient aerosol.*

Inter-comparison experiments for strongly absorbing (carbon black) and moderately absorbing (ambient air) aerosols. Sensitivity to purely scattering aerosol was investigated with ammonium sulfate, which does not absorb light at wavelengths in the visible part of the electromagnetic spectrum.

10 Experimental runs with ambient air were done in order to compare the response to a real atmospheric aerosol. Ambient aerosol particles may consist of an unknown mass fraction of organics and may be coated with absorbing or non-absorbing liquids. Artifacts due to evaporation and/or condensation may be possible. Experiments which deal with these effects have been done in recent years (e.g. Subramanian et al., 2007; Lack et al., 2008; and Cappa et al., 2008). In Lack et al. (2008) and Cappa et al. (2008) it was found that PSAP overestimates absorption significantly in the presence of organic matter. Dedicated experiments concerning the response to organics were not performed during the workshops, because this issue was far beyond the scope of the workshops.

20 3.2 Experimental set-up

The instruments were compared using ambient aerosol. This comparison included:

- a) Inter-comparison of instruments of a particular make and type to investigate instrument to instrument variability;
- b) Comparison of different types of instruments to determine relative sensitivities to particle absorption. For the latter experiments, the air conditioning of the

laboratory was switched off and the windows were opened. It was ensured that all instruments were placed at a similar distance to the windows.

Solutions of ammonium sulfate and carbon black (Printex 75, Evonik Degussa GmbH) were atomized for aerosols with defined composition. The aerosol was dried by diffusion dryers and fed into a 0.5 m³ stainless steel mixing chamber as shown in Fig. 1. Absorption photometers were connected to six of the eight output ports of the mixing chamber. Two output ports were used for additional aerosol characterization using a Scanning Mobility Particle Sizer (SMPS) and an aerodynamic particle sizer (APS, TSI model 3321) to measure number size distributions, and an integrating nephelometer (TSI, model 3563) to measure scattering coefficients.

Before the GAW2005 workshop the mixing chamber was tested for possible differences of aerosol concentration at the outlet ports. Particle number concentrations at the outlet ports were measured with two Condensation Particle Counters (CPC), which were checked for measuring the same concentration before testing the chamber. Eight tests with different combinations of ports were done. The aerosol source was dried ambient air with a total aerosol flow of about 15 lpm through the chamber. It was found that differences in aerosol concentration were smaller than 1.5% for all eight ports. Afterwards filtered air with the same flow rate was fed into the chamber. The aerosol concentration was reduced by half after about 10 min. After three hours no particles were measured. This test showed that it is possible to use one chamber for different types of aerosols after flushing the chamber with particle free air. Measurements of ambient air were done in a different way. With the mixing chamber it would be impossible to compare all instruments at the same time because of the limited number of aerosol outlets of the chamber. Thus for ambient air measurements the instruments were placed with their aerosol inlets at the same distance to the windows of the laboratory and the windows were opened. The laboratory air conditioning was switched off during these experiments. The relative humidity was measured in the inlet of a nephelometer and was always lower than 35%, even during overnight runs.

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3.3 Aerosol characterization

3.3.1 Particle scattering coefficient

Scattering and backscattering coefficients were measured using an integrating nephelometer at wavelengths of 450, 550, and 700 nm. This nephelometer measures the integrated intensity of light scattered at angles between 7° and 170°. This limitation and an imperfect light source result in underestimation of the scattering coefficient. A correction for this so called “truncation error” (Anderson and Ogren, 1998) can be done if the refractive index and the particle size distribution are known. Sometimes, the uncorrected scattering coefficients are used, e.g. when applying the scattering correction for commercial PSAPs proposed by Bond et al. (1999).

3.3.2 Particle number size distribution

The particle number size distributions were measured using an SMPS in the size range from 10 nm to 600 nm of electrical mobility diameter. Larger particles, in the aerodynamic size range from 0.5 to 20 μm, were measured with the APS. The aerodynamic diameter d_{aer} is related to the equivalent geometrical particle diameter d_p by

$$d_p = d_{aer} \cdot \sqrt{\chi_p / \rho_p}, \quad (1)$$

where ρ_p is the particle density and χ_p is the dynamic shape factor of the particle. For ambient aerosol and ammonium sulfate we used a particle density of 1.7 g/cm³ and a dynamic shape factor of unity. Using a dynamic shape factor of unity, the electrical mobility diameter equals the geometrical diameter. In case of carbon black, the same values were used, since the particle number concentration measured in the size range of the APS were too low and do not contribute significantly to the volume concentration or optical properties.

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3.4 Aerosol characterization results

The particle number size distributions of ambient aerosol, ammonium sulfate, and carbon black used in the GAW2005 workshop are shown in Fig. 2 and those for the EU-SAAR workshop are shown in Fig. 3. Physical and optical characteristics are given in Tables 1 and 2. The effective radius, defined by

$$R_{\text{eff}} = \frac{\int r^3 n(r) dr}{\int r^2 n(r) dr}, \quad (2)$$

is the area weighted mean radius of the particle number size distribution. The single scattering albedo

$$\omega_0 = \frac{\sigma_{\text{sp}}}{\sigma_{\text{sp}} + \sigma_{\text{ap}}} \quad (3)$$

is calculated for the actual wavelengths of the Radiance Research 3 λ -PSAP as measured during the RAOS experiment in 2002 (Sheridan et al., 2005). PSAP data were corrected using the Bond correction scheme (Bond et al., 1999) and scattering coefficients were measured with a nephelometer as described above.

4 Absorption photometers description

Several types of instruments for measuring aerosol light absorption coefficients are commercially available. Filter-based instruments measure the rate of change of transmittance through a fiber filter as particles are deposited. The complex relationship between change in light transmission and aerosol absorption and scattering on the filter requires a calibration of these filter-based methods.

The Aethalometer (Magee Scientific, Berkeley, USA; Hansen et al., 1984) is offered in different configurations. The model AE31 measures light transmittance through the filter at seven wavelengths, from 370 to 950 nm. The ability to measure multispectral

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absorption coefficients provides insight in the chemical composition of the absorbing material. Corrections for this instrument type were developed by several investigators (Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006; Collaud Coen et al., 2009). Another correction of the loading effect was shown by Virkkula et al. (2007), in which the reported BC concentration is not converted to absorption coefficients. The Particle Soot Absorption Photometer (PSAP, Radiance Research, Seattle, USA), originally measured light absorption at one wavelength in the green, and correction schemes for this instrument were developed by Bond et al. (1999) and Virkkula et al. (2005). A three wavelength model was developed later, with wavelengths of 467 nm, 530 nm and 660 nm (Virkkula et al., 2005). Corrections applied to data of the PSAP and the Aethalometer require the knowledge of the particle scattering coefficient, often measured with nephelometers. An inherent correction method for minimizing the cross sensitivity to particle scattering was realized for another instrument type, the Multi Angle Absorption Photometer (MAAP, Thermo ESM Andersen Instruments, Germany). In addition to the filter light transmittance, the MAAP measures the reflectivity of the filter at two angles. A radiative transfer model implemented in the MAAP relates the measured signals to the particle absorption coefficient (Petzold et al., 2004). The MAAP and the Aethalometer utilize a filter tape drive mechanism providing automatic filter advance, which facilitates long term monitoring of aerosol absorption.

The instruments tested during the workshops GAW2005 and EUSAAR2007 are presented in Tables 3 and 5, respectively. Instruments for aerosol characterization were similar for both workshops and are given in Table 4. In order to compare reported values, measured absorption and scattering coefficients were corrected to standard temperature and pressure conditions (STP, 0°C and 1013.25 hPa). A more detailed description of the instruments is given in the following sections.

4.1 PSAP

The operating principle of the PSAP is described in Bond et al. (1999). The PSAP with a nominal wavelength of 565 nm is referred to as the *old* PSAP. In the *new* PSAP the

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light source was replaced by a diode emitting light at a shorter wavelength of about 530 nm. In addition, the opal glass plate between light source and particle filter was replaced by a diffusely scattering hemisphere. A prototype, 3-wavelength PSAP was developed as described by Virkkula et al. (2005) with optical wavelengths of 467, 530, and 660 nm. This instrument differed slightly from the commercial version developed later, particularly with regard to the optical diffuser. The wavelengths of all PSAPs in the workshops were checked using an optical spectrometer. Methods and results are presented in Sect. 5.

PSAP correction schemes were developed by Bond et al. (1999) (in the following referred to as *Bond correction*) and Virkkula et al. (2005). For the PSAP inter-comparison, most notably the unit-to-unit variability, the Bond correction was applied to all types of PSAP, although it was developed for the *old* PSAP type instruments. The Bond correction accounts for flows and spot sizes that differ from the values used for internal calculations and for loading and scattering artefacts. The scattering corrections were developed for the *old* PSAP having a nominal wavelength of 565 nm, whereas the applied scattering correction uses scattering coefficients at 550 nm, the center wavelength of the green channel of a TSI-nephelometer (TSI, model 3563). The scattering coefficients used for the Bond correction are not corrected for the so called truncation error (Anderson, 1996; Heintzenberg, 2006; Müller et al., 2009). Correction of the 3λ PSAP requires the corresponding scattering coefficients. The interpolation and extrapolation of scattering coefficients was done using the scattering Ångström exponent α_{sp} which is defined by:

$$\alpha_{\text{sp}}(\lambda_1, \lambda_2) = \frac{-\ln(\sigma_{\text{sp}}(\lambda_2)/\sigma_{\text{sp}}(\lambda_1))}{\ln(\lambda_2/\lambda_1)} \quad (4)$$

With scattering coefficients measured at three wavelengths, an average scattering Ångström exponent has been calculated. The overall correction, which is based on

the procedure described in Bond et al. (1999), is given by

$$\sigma_{\text{ap}}(t) = \frac{A}{Q \cdot \Delta t} \ln \left(\frac{I(t - \Delta t)}{I(t)} \right) \cdot \frac{1}{1.317 \cdot \tau + 0.866} - 0.016 \cdot \sigma_{\text{sp}} \quad (5)$$

with sample spot area A , volumetric flow rate Q , time interval between readings Δt , optical transmission relative to a blank filter τ , and the measured intensity $I(t)$.

The approach of Virkkula et al. (2005) was not used because during the analysis of the data of the present work it was found, that in the Virkkula et al. (2005) paper there is an error in flow correction which affects the factors of the transmission correction function. The error leads to an underestimation of absorption coefficients. An erratum article is under preparation.

4.2 MAAP

The MAAP measures the radiation transmitted through and scattered back from a particle-loaded filter. A two-stream radiative transfer model is used to minimize the cross sensitivity to particle scattering. A detailed description of this method can be found in Petzold et al. (2004). Although MAAP measures absorption coefficients, the values reported by the instrument are given as mass concentration m_{BC} of black carbon (BC) using a mass specific absorption coefficient of $Q_{\text{BC}} = 6.6 \text{ m}^2/\text{g}$.

The MAAP operation manual gives the operating wavelength as 670 nm. During the Reno Aerosol Optics Study (RAOS), MAAP absorption was compared (Petzold et al., 2005) to a reference absorption measurement (Sheridan et al., 2005). The reference absorption coefficient was calculated both as the difference between measured extinction and scattering coefficients and as determined using photoacoustic photometry. Absorption coefficients measured by the reference technique were adjusted to 670 nm using the Ångström law with an Ångström exponent of 1.02. From regression analysis of the MAAP and the reference absorption, a regression line with a slope of 0.99 ± 0.01 was calculated for pure black carbon particles.

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However, during the GAW2005 workshop, it was found that the optical wavelength of MAAP is 637 ± 1 nm instead of 670 nm. The full width at half maximum (FWHM) of the emitted light is about 18 nm. The consequences of this wavelength mismatch are:

- a) The MAAP can be used for aerosol particles with an absorption Ångström exponent¹ close to unity to give directly the absorption at 670 nm.
- b) As described above, the MAAP compares excellently with the photoacoustic reference adjusted to 670 nm, whereas the real MAAP wavelength is 637 nm. For an Ångström exponent of 1.02, the absorption coefficient at 637 nm should be 5% higher than at 670 nm. Hence, for this Ångström exponent, the MAAP *measured* absorption coefficient at 637 nm is 5% low and should therefore be corrected by multiplication by a factor of 1.05. If the actual Ångström exponent is known, the appropriate correction factor should be used.

Throughout the entire manuscript, the following correction was applied to MAAP data:

$$\sigma_{\text{ap}}^{637\text{nm}} = m_{\text{BC}} \cdot Q_{\text{BC}} \cdot 1.05, \quad (6)$$

where m_{BC} is the equivalent mass concentration of black carbon reported by the instrument and $Q_{\text{BC}} = 6.6 \text{ m}^2/\text{g}$.

4.3 Aethalometer

Several versions of Aethalometers were used in both workshops (listed in Tables 3 and 5). A more complete description of the Aethalometers can be found in the user manual (Hansen, 2005).

The Aethalometer measures the attenuation $\text{ATN}(\lambda, t)$ defined by:

$$\text{ATN}(\lambda, t) = -\ln(I(\lambda, t)_0 / I(\lambda, t)), \quad (7)$$

¹ The absorption Ångström coefficient α_{ap} is defined similar to the scattering Ångström exponent in Eq. (4).

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where I_0 is the intensity of light that passes through a pristine portion of the filter and I is the intensity of light that passes through the particle-laden filter. The change in light attenuation by filter loading during a time interval Δt defines the attenuation coefficient σ_{ATN} as:

$$\sigma_{\text{ATN}} = \frac{\text{ATN}(\lambda, t + \Delta t) - \text{ATN}(\lambda, t)}{\Delta t} \cdot \frac{A}{F}, \quad (8)$$

where A is the area of the filter spot and F is the volumetric flow rate. The aethalometer internal software converts the measured attenuation coefficient into equivalent black carbon mass concentration (m_{BC}) using :

$$m_{\text{BC}} = \frac{\sigma_{\text{ATN}}}{\text{SG}_{\text{BC}}}, \quad (9)$$

where $\text{SG}_{\text{BC}} = 14625/\lambda$ [m^2/g] (wavelength given in nm) is the spectral mass specific attenuation cross-section. The Aethalometer reports m_{BC} rather than the attenuation coefficient σ_{ATN} , which is needed to calculate the absorption coefficient, but σ_{ATN} can be calculated from the reported lamp intensity. However, because the recorded m_{BC} is internally calculated from higher precision data than those provided in the aethalometer data output file (Magee Scientific, Berkley, USA, personal communication, 2008), m_{BC} should be used to calculate σ_{ATN} . In this paper σ_{ATN} is used as derived from m_{BC} by multiplication with SG_{BC} (Eq. 9).

The attenuation coefficients σ_{ATN} are converted to absorption coefficients σ_{ap} using the Weingartner et al. (2003) correction function:

$$\sigma_{\text{ap}} = \sigma_{\text{ATN}} / (C \cdot R(\text{ATN})), \quad (10)$$

where the factor $C=2.14$ is introduced for the correction of multiple light-scattering effects of the filter fibers. More recently, Collaud Coen et al. (2009) evaluated a newly developed and four already existing, aethalometer correction schemes and concluded that this value for C is too low and should be at least 2.9 (average C values for four

datasets varied between 2.9 and 4.3. However, Collaud Coen et al. (2009) recommend further analysis to extend their results to obtain a more universal multiple scattering correction factor. Awaiting the results from such research, we used the original value $C=2.14$ as suggested by Weingartner et al. (2003). The factor R accounts for the reduction of the optical path length in the filter with increasing filter load:

$$R(\text{ATN}) = (1/f - 1) \cdot [\ln(\text{ATN}) - \ln(10\%)] / [\ln(50\%) - \ln(10\%)] + 1, \quad (11)$$

where $f = a(1-\omega_0)+1$ with $a=0.87$. All Aethalometers are corrected by the same experiment/measurement-period average f value.

This correction is referred to as the *Weingartner correction* in the rest of the paper.

5 Absorption photometer characterization

5.1 Emission wavelengths of different absorption photometers

The spectral emitted radiation of absorption photometers was measured with a grating spectrophotometer (HR2000, Ocean Optics Inc.) equipped with a fiber optic connector. One end of an optical fiber was connected to the spectrophotometer and the other end was held into the measurement head of the photometer and measured the directly emitted light of the diodes. A wavelength dependency of the filter transmittance is negligible, since the spectral width of the emitted light is small compared to the spectral transmittance of the filter (see Arnott, et al., 2005). The light emitted by the PSAP and the Aethalometer was measured directly; for the MAAP it was only possible to collect some stray light. Examples of measured spectra are shown in Fig. 4. The measured intensity spectra $I(\lambda)$ were corrected for the spectral sensitivity of the spectrometer detector $S_S(\lambda)$ and the grating efficiency $\chi_S(\lambda)$. The spectral sensitivity of the photometer detector $S_P(\lambda)$ was also taken into account. Values for $S_P(\lambda)$ were taken from datasheets of typically used silicon detectors. The *effective* wavelength is defined

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as the first moment of the sensitivity corrected spectra

$$\lambda_{\text{eff}} = \frac{\int \lambda \cdot I_{\text{corr}}(\lambda) d\lambda}{\int I_{\text{corr}}(\lambda) d\lambda}, \quad (12)$$

with the sensitivity-corrected intensity

$$I_{\text{corr}}(\lambda) = \frac{S_P(\lambda)}{S_S(\lambda) \cdot \chi_S(\lambda)} \cdot I(\lambda) \quad (13)$$

5 Results from these measurements are summarized in Table 6 and are discussed below.

Several types of PSAPs (Radiance Research) were tested in these workshops. Some instruments (serial number SN<48²) were delivered with a peak wavelength at 565 nm. A tail of the emitted radiation at longer wavelengths causes an effective wavelength of about 585 nm. For a typical wavelength dependence of λ^{-1} the ratio of absorption coefficients at 565 and 585 nm is 1.035. One newer Instrument (SN 71) was measured to have a symmetrical intensity distribution with a peak wavelength at 522 nm and a FWHM of 20 nm. The three-wavelength PSAPs show peak wavelengths at 467 (FWHM 20 nm), 531 (FWHM 40 nm), and 650 nm (FWHM 22 nm), slightly different than the detector weighted averaged wavelengths of 467, 530 and 660 nm given for the prototype instrument in Virkkula et al. (2005) and given in the manual by the manufacturer (cf. Table 6). The measured spectral radiances (without sensitivity correction) are shown in Fig. 4a. The three intensity spectra of 3 λ -PSAPs could not be measured separately because the instrument switches between the different light sources at a frequency which is faster than the integration time of the spectrophotometer.

15
20 Spectra of the seven-wavelength and two-wavelength Aethalometer models are shown in Fig. 4b and c. Only spectra of one specific instrument of each type are shown, because no significant differences between instruments of the same type were observed. Spectra of white light Aethalometers are not shown. Reasons are the unknown spectral sensitivity of the detector of the Aethalometer and the upper limit of

²The serial number at which the light source was changed is not precisely known.

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the wavelength range of the Ocean Optics spectrophotometer. Because of the limited wavelength range we were not able to measure the whole emission spectrum. Estimates of the emitted spectral radiation and the effective wavelength of this Aethalometer are reported by Weingartner et al. (2003), who specify the effective wavelength to be 855 nm for unloaded filters. Weingartner et al. (2003) also noted that the effective wavelength depends on the particle loading, which causes a wavelength dependent attenuation of the transmitted light ($I(\lambda)$ in Eq. 13). This fact makes it much more difficult to accurately estimate the effective wavelength for white light Aethalometers.

For the comparison of different instruments, the measured absorption coefficients have been adjusted to a *reference* wavelength. This requires knowledge of the spectral variation of the absorption or scattering coefficients, which is given by the Ångström exponent. The absorption Ångström exponent was calculated using a 3 λ -PSAP after application of the Bond correction. The absorption Ångström exponent was determined separately for PSAP and Aethalometer, respectively. The scattering Ångström exponent was calculated in a similar way from the scattering coefficients measured with a nephelometer at wavelengths of 450, 550 and 700 nm.

5.2 Spot size

Spot size areas of absorption photometers are needed for calculation of absorption coefficients. For instance, the spot area is used in the Bond correction of PSAP to account for the difference from a *reference* spot area. During both workshops spot areas were measured with optical reticles for PSAP and the MAAP and with vernier calipers for the Aethalometer.

For each PSAP, spot areas of six (GAW2005) and eleven (EUSAAR2007) filters were measured by different workshop participants. Average and standard deviation of spot areas were calculated for each PSAP. The standard deviation is determined from the uncertainty in the measured spot area for different instruments. In Table 7, average spot areas of all PSAPs and the average of the standard deviations are given. On average, spot areas are about 6% smaller (EUSAAR2007) and 1% larger (GAW2005)

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than the reference spot area of 20.43 mm^2 used in the Bond correction. Here it should be noted, that the manufacturer uses a reference spot area of 17.85 mm^2 for calculating the absorption coefficient. The uncertainty (standard deviation) in the measured spot size for one individual filter by different people was on average 4%. Figure 5 shows a plot of the standard deviation versus the spot area for different PSAPs. Clearly, the standard deviation increases with increasing spot area. This behavior could be explained by bad sealing rings. It is noted that different types of filter holders are available for the PSAP and that some types of holders appear to seal better than others.

Aethalometers can be purchased with two different spot sizes, i.e. the “High Sensitivity” (HS) spot size measuring 0.5 cm^2 and the “Extended Range” (ER) spot area of 1.67 cm^2 . The choice of spot area may depend on the level of pollution at a monitoring site and the sensitivity that is needed. The greatest sensitivity is achieved with a small spot area and the highest air flow. The disadvantage of greater sensitivity is that transmission goes down in a shorter period of time, which leads to more interruptions of data due to filter transport. During the GAW2005 workshop the spots of three HS and one ER and during the EUSAAR2007 workshop the spots of three HS and two ER types of instruments were measured.

To test the objectivity of spot area measurements, the spot areas for all instruments available during EUSAAR2007 were measured three times, by two persons. The standard deviation of the distribution of the measurements for a single spot (0.02 cm^2) is similar to the standard deviation of the average spot sizes collected from various instruments (0.03 cm^2) provided in Table 8 which shows the average spot areas for all Aethalometers for both workshops. The average spot areas for the high sensitivity spots are 4% larger (EUSAAR2007) and 8% lower (GAW2005) than those reported by the manufacturer. Likewise, for the extended range spots, the areas were 2% larger (EUSAAR2007) and 4% smaller (GAW2005) than those specified by the manufacturer. In view of these results and their standard deviations (Table 8), we conclude that for Aethalometers the spot sizes do not significantly deviate from the spot sizes reported by the manufacturer. Therefore, we did not apply any correction.

Spot sizes of all available MAAP instruments did not show a significant variation within the accuracy of the measurement (0.1 mm in diameter). Consequently, it is not expected that variability in spot sizes has an effect on the determination of absorption coefficients with a MAAP.

5.3 Flow rates

To estimate the sensitivity of the absorption photometer response to different face velocities, the instrument pump flows were varied during EUSAAR2007, after the pressure and temperature sensors and flow rates were calibrated. For the MAAP, the standard flow rate was 16.7 liters per minute (lpm). In total, seven MAAPs were compared. Prior to the sensitivity test, the unit-to-unit variability was determined. The unit-to-unit variability is defined by the coefficient of variation (CV), which is the ratio of the standard deviation and the average absorption coefficients measured simultaneously with a number of instruments. For this set of instruments $CV=3\%$. Before the sensor calibrations, CV was 11%; hence a proper flow calibration is a key to proper functioning of the instruments.

The set was split into two groups, one group consisting of three instruments (“standard set”) which was continuously operated at a standard flow of 16.7 lpm. The second group, consisting of four instruments (“test set”), was operated at different flows of 16.7 lpm, 10 lpm and 6 lpm. At 16.7 lpm, the unit-to-unit variabilities of the standard and test sets were 2% and 3%, respectively. These values are comparable to the overall unit-to-unit variability ($CV=3\%$). The absorption coefficient obtained with the test set was on average 1.9% higher than for the standard set. For a subsequent experiment of 13 h, the flow rate of the test set was lowered to 10 lpm. In that experiment, the absorption coefficient of the test set was 1.7% lower than that for the standard set and the unit-to-unit variability increased to 4%. The unit-to-unit variability of the standard set decreased to 1% in this period. For the following nighttime period (12.5 h), the flow of the test set was lowered to 6 lpm. The unit-to-unit variability of the test set increased to 13% and the average absorption coefficient decreased by 5% with respect to the

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standard set. Average deviations between standard and test sets were smaller than 2% for flow rates larger than 10 lpm and 5% for a flow rate of 6 lpm. The latter deviation of the average absorption coefficient (5%) is larger than the unit-to-unit variability of the instrument at standard flow. We can explain neither the larger unit to unit variability nor the deviation to the standard set. Possible causes are that the flow regulation does not work properly at 6 lpm, and/or particles are embedded differently in the filter matrix at low flow. We conclude that running a MAAP with a flow rate of 6 lpm does not provide reliable results. The results from these experiments confirm that the minimum flow rate should be 8.3 lpm as recommended in the instrument manual.

The Aethalometer flow rate recommended by the manufacturer is 2–6 lpm. For the “High Sensitivity” (HS) and “Extended Range” (ER) spot sizes, the recommended flow rates correspond to different face velocities for the two available spot sizes. For HS and ER, 2–6 lpm corresponds to face velocities of 67–200 and 20–60 cm/s, respectively. Due to the limited number of instruments and the larger unit-to-unit variability, as compared to the MAAP, the suite of instruments could not be divided into two sets as was done for the MAAP. Instead, the face velocity of one of the Aethalometers was changed for a period of time that is long enough to average out instrument noise precision. During the Aethalometer flow rate sensitivity experiments, MAAP data were not available. Data shown for this experiment were corrected with the Weingartner correction.

To investigate sensitivity of the absorption coefficient to a change in face velocity, two Aethalometers with an ER spot were consecutively operated at flows of 1.9, 4.0, 5.9, and 6.8 lpm. The response of the instruments with changed flow was compared to the response of the same instrument during a reference period of three days with all instruments running at the standard flow of 4 lpm. To facilitate the comparison we introduce the face-factor ff:

$$ff = \frac{(\sigma_{\text{abs}}(\text{instrument})/\sigma_{\text{abs}}^*)|_{\text{flow experiment}}}{(\sigma_{\text{abs}}(\text{instrument})/\sigma_{\text{abs}}^*)|_{\text{reference period}}} \quad (14)$$

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where σ_{ap}^* is the average absorption coefficient of the 5 Aethalometers that run at the standard flow. The Aethalometer running at 1.9 lpm was not sensitive to the new face velocity, $ff=1.01$. For 5.9 and 6.8 lpm the face factors were 1.06 and 0.90, respectively.

For Aethalometers with HS spots, we operated one Aethalometer with a flow of 2.9 lpm and another one with 6.5 lpm. For the low-flow experiment, stability problems with the external pump of this Aethalometer were experienced and results cannot be trusted. The experiment with high flow resulted in $ff=1.16$. This is a rather large impact that exceeds the unit-to-unit variability.

These flow experiments indicate that Aethalometers should not be operated with flows at the high end of the recommended range. We observed significant changes, overestimations as well as underestimations in absorption coefficients that are larger than the instrumental noise and unit-to-unit variability. However, the number of sensitivity experiments was rather limited and a good reference was missing, so that an ensemble of instruments that themselves suffer from large unit-to-unit variability had to be used. Therefore, the experiments performed were not conclusive as regards the upper or lower limits for flow operation.

A flow test for PSAP was conducted during the GAW2005 workshop. One PSAP was operated with a constant flow rate of 1.13 lpm. A first test set of four PSAPs were operated with 2.25 lpm and another test set with only one PSAP with 1.01 lpm. After two hours the filters were replaced and the flow rates for test set one were set to about 0.5 lpm. The flow rate of test set two was set to 2.27 lpm. Ratios of the test sets with the reference PSAP were calculated for the two flow experiments. After changing the flows the ratios changed up to 15%, but no correlation between the ratios and flow rates were found. From this experiment no dependence of the absorption coefficient to the flow rate can be deduced.

5.4 Noise test

The instrumental noise was determined by the analysis of filtered, particle-free air (relative humidity < 30%) measurements. Absorption coefficients of different types of

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photometers were corrected using the standard correction schemes (Bond and Weingartner). Averaging times were one minute for the PSAP and MAAP, and three minutes for the Aethalometer. The instrumental noise is given by the standard deviation of the distribution of absorption coefficients for a single instrument. The noise was calculated for several instruments of the same type. Table 9 summarizes results of both workshops. For each instrument type and selected wavelength, the averaging time and the number of instruments are listed. The noise is averaged for all instruments of the same type.

Instrumental noise for the PSAP determined during GAW2005 was 0.06 Mm^{-1} or smaller for all three wavelengths of a 3λ -PSAP, and on average was 0.36 Mm^{-1} for 1λ -PSAPs. The difference in noise for both instrument types cannot be explained, since it is not known which changes have been made to the light source, the detector, and the electronics in the 3λ -PSAPs. During EUSAAR2007, the average noise of six 3λ -PSAPs was similar to the GAW2005 results, 0.07 Mm^{-1} , and within experimental error. The average noise of two 1λ -PSAPs was lower (0.15 Mm^{-1}) compared to GAW2005.

The noise characteristics of the PSAP and the dependence on the integration time (Δt) were investigated by Springston and Sedlacek (2007). It is assumed that the time between two consecutive measurements is equal to the integration time. From analysis of the error propagation they showed that the PSAP signal noise should be proportional to $\Delta t^{-1.5}$. Theoretically this dependence should be valid for all types of filter based absorption photometers. In contrast, our experiments showed that the noise of the PSAP varies with $\Delta t^{-1.3}$ and the noise was determined to be 1.6 Mm^{-1} for an averaging time of 2 s. Using a power law with an exponent of 1.3, the noise should be 0.02 Mm^{-1} for an averaging time of 60 s. However, the lowest values (3λ -PSAP) measured during the GAW2005 and EUSAAR2007 workshops are higher by a factor of 2.5 than this theoretical value.

The noise of the MAAP, using one minute averaging time, was determined as 0.08 Mm^{-1} during GAW2005 and 0.22 Mm^{-1} for EUSAAR2007. We have no explanation for this rather large difference between the two workshops. In the user manual,

the detection limit for two minutes averaging time (95% confidence level) is given as 0.66 Mm^{-1} , which corresponds to a noise level (single standard deviation) of about 0.33 Mm^{-1} . Even with a shorter integration time the noise level observed during both workshops was lower than that reported in the user manual.

The noise level for Aethalometers was investigated with instrument-minimum averaging time. During GAW2005 the minimum averaging time for 7λ -Aethalometers (AE31) was three minutes, whereas the minimum averaging time of a white light Aethalometer (AE10) was two minutes. For convenience of comparison, we apply the theoretical $\Delta t^{-1.5}$ noise dependence (Springston and Sedlacek, 2007) to relate the obtained 2-minute noise values to noise values that would have been obtained if the instrument “averaging period” was 3 min. As pointed out by Springston and Sedlacek (2007) this result is not quite the same as recording the data during 2-min and then averaging the data during post processing. This latter technique yields a noise reduction proportional to the square root of the averaging time, but does not allow comparison of noise values obtained by instruments running with different instrument averaging times. The effect of (post processing) averaging time on the Aethalometer noise level is illustrated in Fig. 6. The noise values for the four 7λ -Aethalometers that were operated on filtered air during EUSAAR2007 were averaged over 2, 4, 6, 10, and 30 min. Noise values for all wavelengths were combined as no wavelength dependence was observed. The data points (measurements) closely follow the fitted line (inverse square root of time dependence). During GAW2005 filtered air measurements were averaged over 3 min, the noise level of AE31 photometers was 0.42 Mm^{-1} at a wavelength of 370 nm and 0.17 Mm^{-1} at a wavelength of 880 nm. The noise of a white light Aethalometer (AE10) was much higher with a value of 1.84 Mm^{-1} .

During EUSAAR2007, noise levels were determined for measurements at a 2 min averaging time for the 7λ -Aethalometers and a 1 min instrument-averaging time for the single wavelength aethalometers. As above, and for convenience of comparison to the GAW2005 Aethalometer-noise estimates, all presented values were converted to 3 min instrument averaging times (Springston and Sedlacek, 2007). The noise levels

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for two single wavelength Aethalometers (880 nm) and the two 7λ-Aethalometers with extended range spots (1.67 cm²) and the two 7λ-Aethalometers with high sensitivity spots (0.5 cm²) were estimated independently. The standard deviations of filtered air measurements of Aethalometers with ER spots (1.5 Mm⁻¹) were markedly higher than the standard deviations for instruments with HS spots (0.7 Mm⁻¹). For both types of Aethalometers, the pump flow was 4 lpm, which corresponds to different face velocities for both types of spots. Springston and Sedlacek (2007) showed in their Eq. 4, that the absorption coefficient noise and face velocity are inversely proportional. Adopting their relation, we reduced the estimated noise levels of the Aethalometers with ER spots by a factor of 1.67/0.5. After the mathematical conversion to the same face velocity as the HS spots, the 3-min average noise value of the two ER-7λ-Aethalometers in this study was 0.24 Mm⁻¹. The 3-min average noise value of the HS-λ-Aethalometers in this study was 0.38 Mm⁻¹. For the single wavelength Aethalometers, the 3 min averaging noise value was 0.19 Mm⁻¹, where we reduced the noise of one of the aethalometers by a factor of two because of its deviating spot size of 1.0 cm². After transformation of all Aethalometers to a face velocity of 133 cm/s, corresponding to 4 lpm and HS spot sizes, the average noise was 0.27 Mm⁻¹ for a 3 min instrument time averaging, with maximum and minimum noise of 0.41 Mm⁻¹ and 0.14 Mm⁻¹, respectively.

6 Instrument intercomparison

6.1 Reference instrument

The MAAP was used as “reference instrument” for absorption measurements (cf. Sect. 4.2). The MAAP certainly suffers as all filter based methods to a cross sensitivity to scattering. Therefore MAAP can not be a “true” absorption reference instrument, as e.g. extinction minus scattering measurements, but there were several reasons to use MAAP like a reference method for this study.

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This choice is based on the results from the RAOS study (Sheridan et al., 2005), with good agreement between photoacoustic spectrometer (PAS) measurements and absorption obtained from the difference between extinction (from an extinction cell) and scattering (from a nephelometer). Also the low unit to unit variability, given by the standard deviation of several MAAPs running in parallel, makes this instrument a good reference. During the GAW2005 and EUSAAR2007 workshops, other instruments had been expected to provide extinction-scattering or photoacoustic data to obtain the reference absorption, but all instruments failed for one reason or another. Therefore the MAAP was the only alternative. Time series of average MAAP values and standard deviations are shown in Fig. 7 for both workshops. Both time series were measured with dry ambient air. Unit to unit variability was on average 5% during GAW2005 and 3% during EUSAAR2007.

6.2 Ambient air

To account for the wavelength dependence of the aerosol absorption, PSAP and Aethalometer absorption coefficients were adjusted to the MAAP wavelength of 637 nm. The wavelength adjustment uses the respective absorption Ångström exponent measured with a 3λ-PSAP and 7λ-Aethalometer. For PSAPs, the Ångström exponent for absorption was 1.14 during GAW2005 and 1.08 and 0.99 for two different experiments during EUSAAR2007. For Aethalometers, the applied Ångström exponent for absorption obtained from channels 520, 590, 660, and 880 nm, was 0.97 for GAW2005 and 1.06 for EUSAAR2007, respectively. Prior to wavelength adjustment, absorption coefficients measured with PSAPs and Aethalometers were corrected using the Bond and Weingartner corrections, respectively. The relative sensitivity ε^n of an individual instrument “ n ” is defined by the ratio of the wavelength adjusted absorption coefficients divided by the average of the absorption coefficients measured with the MAAP.

$$\varepsilon^n = \sigma_{\text{ap}}^n(637\text{nm}) / \text{avg}[\sigma_{\text{ap}}^{\text{MAAP}}(637\text{nm})] \quad (15)$$

Relative sensitivities for PSAPs and Aethalometers were averaged for instruments of the same type and wavelength. The results are presented in Table 10. The variability of the sensitivity, given by the unit to unit variability (standard deviation/mean), is also presented in Table 10.

To avoid effects of “overloading” of the PSAP filter, data were only used when the transmittance was between 1.0 and 0.7. Data evaluation of GAW2005 showed a lower sensitivity to light absorption in ambient aerosol for the PSAPs compared to the MAAP. The relative sensitivities were between 0.79 and 0.86 after adjusting the data to the MAAP wavelength of 637 nm. The variability in the sensitivity for the various wavelengths was small as compared to the unit-to-unit variability which ranged from 0.07 and 0.27. Figure 8 shows the absorption coefficients measured with the 3 λ - PSAP vs. those measured with the MAAP, without wavelength adjustment.

During EUSAAR2007, experiments with ambient air showed a higher sensitivity of PSAP compared to GAW2005. The relative sensitivity for two experiment runs are 1.05 and 0.99 and the corresponding uncertainties are 8% and 10%, respectively. The correlation of absorption coefficients adjusted to 637 nm is shown in Fig. 9 for one run. The sensitivity to for low loadings (transmittance between 1 and 0.7) is 1.03 and for higher loading (transmittance smaller 0.7) the sensitivity is 0.96. Reduced sensitivity at higher loading implies that the applied Bond loading correction, that accounts for the reduction of the optical path length in the filter with increasing filter load, is not sufficient for transmittance smaller than 0.7. This finding is in agreement with that of Bond et al. (1999).

During EUSAAR2007, two periods were selected to study Aethalometer sensitivity. These periods were selected because both MAAP and nephelometer data were available. The Weingartner method (Sect. 4.3) to convert the attenuation coefficients σ_{ATN} to absorption coefficients σ_{ap} influences the unit-to-unit variability through the loading correction factor R (cf. Sect. 4.3). The average of all MAAPs and the truncation corrected nephelometer data were used to calculate the single scattering albedo. During period 1 the average single scattering albedo (637 nm) was 0.75 (0.67–0.81) and $f=1.22$.

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For period 2 the single scattering albedo (637 nm) was 0.87 (0.76–0.91) and $f=1.12$. For the obtained absorption coefficients, we obtained $CV=9\%$ (period 1) and $CV=8\%$ (period 2) for all instruments at 880 nm and $CV=8\%$ (period 1) and 9% (period 2) for the four 7λ -Aethalometers at all wavelengths. Comparing absorption coefficients measured with Aethalometer and MAAP, we conclude that the multiple scattering part of the Weingartner correction is not sufficient to overcome the overestimation of the absorption coefficient measured by Aethalometers for the aerosol measured during the workshop. The ratio $\sigma_{\text{abs,aeth}}/\text{avg}(\sigma_{\text{MAAP}})$ is about 1.6 (Table 10). The value $C=2.14$ was determined for experiments where no SOA was produced (“pure” soot). Higher C values were found in the AIDA experiments (Saathoff et al., 2003) when SOA was also present in the chamber. This is probably caused by the condensation of semi-volatile material on the fibres, which leads to higher C values. Collaud et al. (2009) compared MAAP and Aethalometers at several field sites in Europe and also found higher C values in the range of 2.9 to 4.3. Figure 10 shows the absorption coefficient at 660 nm versus the absorption coefficient measured by MAAP at 637 nm.

The absorption Ångström exponent deduced from Aethalometer attenuation coefficients is 1.06 ± 0.08 . After the loading correction (Weingartner et al., 2003), the Ångström exponent increases to 1.18 ± 0.08 . This increased wavelength dependency does not mean that the loading correction is wavelength dependent; it is merely the result of increasing ATN values (and thus a stronger loading correction) with decreasing wavelength. The effect of such large deviations in Ångström exponents that can be used to make the conversion is obvious, e.g. using an Ångström exponent of 1.18 instead of 1.06 results in about 15% higher absorption coefficients at 370 nm (starting at 637 nm). This fact and the uncertainty in the wavelength dependence of the absorption coefficient make comparison at various wavelengths rather difficult.

In light of these difficulties, the MAAPs and the four 7λ -Aethalometers were compared only at a single wavelength (660 nm channel adjusted to 637 nm). Using the relative sensitivity introduced at the beginning of this section, we find that absorption coefficients deduced using the Weingartner correction are higher than the reference

absorption coefficients by a factor of 1.6 ± 0.2 during EUSAAR2007 and 1.37 ± 0.1 during GAW2005, respectively. As mentioned before in this paper the C value found by Weingartner et al. (2003) is rather low. Collaud et al. (2009) found values of at least 2.9; application of these values would lead to sensitivities closer to unity.

The determination of the light absorption coefficient with AE10 Aethalometers is difficult because of the ill-defined spectral sensitivity. However, for the sake of intercomparability, we applied the Weingartner correction and adjusted the absorption coefficient to 637 nm by adopting the effective wavelength for white light Aethalometers as specified by Weingartner et al. (2003) to be 840 nm (855 for unloaded filters). The so-obtained relative sensitivity of AE10 was 1.21 during GAW2005.

Relative sensitivities differ strongly between GAW2005 and EUSAAR2007. For GAW2005 the relative sensitivities are significantly smaller for both PSAP and Aethalometer. Absorption coefficients were in a moderate range from 8 to 15 Mm^{-1} (GAW2005) and from 12 to 23 Mm^{-1} (EUSAAR2007). For EUSAAR2007 and GAW2005 differences in the particle number size distribution (cf. Figs. 2 and 3) were observed, which could be an indication of different particle composition and thus optical properties. The sensitivity to OC is not well understood (e.g. Lack et al., 2008) and can differ between PSAP, MAAP, and Aethalometer. Thus an artifact due to organics is possible but not proven. Besides, differences in the particle composition and number size distribution, we do not have an explanation for the different sensitivities between both workshops.

Figure 11 shows the unit to unit variability of corrected (Bond and Weingartner, respectively) absorption coefficients for all instruments of the same kind versus reference absorption (average of absorption coefficients measured by MAAPs). When comparing values for PSAP, MAAP, and Aethalometer, it should be considered, that the scaling in Fig. 11a–c differs. Values are shown for 1 and 10 min averaging times for MAAP and PSAP. The slope of linear regressions indicate that the MAAP has lower unit to unit variability (CV=3.2% and 3.8%) compared to the PSAP (CV is about 8%). For both types of instruments the standard deviation does not depend on averaging time. Thus

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the averaging time should only be considered when absorption coefficients are not much larger than the instrument noise. For absorption coefficients much larger than the noise, the precision of the instruments is dominated by unit to unit variability (systematic error) and not noise (statistical error). The regression line for the Aethalometer has a similar slope (with $CV=8.9\%$) as the PSAP but the points spread much more compared to the PSAP. This spread can be (partly) explained by the difference in instrument noise of both types of instruments. The noise of the Aethalometer is about 0.3 Mm^{-1} (3 min averaging time) compared to 0.08 Mm^{-1} for PSAP (1 min averaging time). The higher noise of Aethalometers causes the larger spreading of points around the regression line.

6.3 Ammonium sulfate

The sensitivity of the PSAP, Aethalometer and MAAP absorption photometers to particle scattering was investigated using the same aerosol (ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$) and same experimental setup during GAW2005. The properties of this aerosol are discussed in Sect. 3.4. White light Aethalometers were not used for this experiment because of their larger noise level compared to multi-wavelengths Aethalometers. Bond et al. (1999) showed that sensitivity to particle scattering is about 1.6% of the scattering coefficient, after application of a loading correction to the attenuation coefficient. However the uncertainty in the sensitivity to particle scattering is as large as the derived uncertainty itself. Virkkula et al. (2005) determined the cross sensitivity for a modified 3λ -PSAP to be between 1.3% and 2.1%, and for a commercial 1λ -PSAP to be 2.3%. In view of the large uncertainties these values are considered to be in agreement with Bond et al. (1999). The first investigation of the sensitivity to particle scattering for the Aethalometer was published by Weingartner et al. (2003) who obtained a ratio of $\sigma_{\text{ATN}}/\sigma_{\text{sp}} = 0.7\%$ for $(\text{NH}_4)_2\text{SO}_4$ for 450 nm and 660 nm. Applying the Aethalometer correction reduces absorption by a factor of about 2. Thus, based on reported sensitivities to particle scattering, the ratio of apparent absorption and scattering coefficient is significantly smaller for the Aethalometer than for the PSAP. Petzold et al. (2005) showed

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that for the MAAP the sensitivity to non absorbing aerosol is smaller than 3%.

The cross sensitivity was calculated using the corrected absorption coefficients divided by the scattering coefficient. The scattering coefficient was interpolated from adjacent wavelengths and, to be consistent with Bond et al. (1999), no truncation correction was applied to nephelometer data. The loading correction for PSAP was done according to Bond et al. (1999) and MAAP values were corrected internally by a radiative transfer model described in Petzold et al. (2004). Cross sensitivity for Aethalometer is given for absorption coefficients after Weingartner correction. Results for the available instruments and wavelengths are presented in Table 11.

The sensitivity to non absorbing aerosol for six MAAPs is on average 0.62% with a standard deviation of 0.06%. This value is significantly smaller than values reported by Petzold et al. (2005). After application of the Bond correction, three 1 λ -PSAPs still show a sensitivity between 0.89% and 2.18%, whereas two 3 λ - PSAPs show a sensitivity between 0.16% and 0.89%. These large uncertainties among different instruments and experiments reflect the large uncertainty already given by Bond et al. (1999). For the Aethalometer, the sensitivity ranges between 1.57% and 2.67% (for wavelengths 450–660 nm) which is significantly higher than the values derived by Weingartner et al. (2003) and more comparable to the sensitivities measured for PSAP. One possible reason for differences compared to other studies might be found in the particle size distribution and different ranges of particle loading, but cannot be explained satisfactorily.

All multi-wavelength photometers show that the sensitivity to scattering increases with increasing wavelength. For the Aethalometer, the sensitivity to non-absorbing aerosols is about four times higher for the IR-wavelength (950 nm) than for the UV-wavelength (350 nm). Wavelength dependencies of scattering corrections also were found in Arnott et al. (2005) for Aethalometer and Virkkula et al. (2005) for PSAP.

The measurements done during GAW2005 and results reported in literature indicate that there is still a lack of understanding the problem of the cross sensitivity to particle scattering. Until now, there is no physics-based model which explains the influence of scattering particles on filter based absorption measurements.

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An obvious problem in the correction given by Bond et al. (1999) is the lack of an explicit loading correction for non absorbing aerosols. That means that the loading correction, which was derived for strongly absorbing aerosols, is applied to attenuation coefficients for both absorbing and scattering particles. In a second step, a constant fraction of particle scattering is subtracted. For Aethalometers, the loading correction given by Weingartner et al. (2003) depends on the particle single scattering albedo. For ammonium sulfate the single scattering albedo is 1.0 and loading correction does not impact on the determined cross sensitivity. The multiple scattering correction reduces the cross sensitivity by a factor of $C=2.14$.

Experiments during EUSAAR2007 were designed to provide insight into the loading dependence of the sensitivity to particle scattering. Thus filters were loaded with ammonium sulfate as long as needed to see a loading effect. In most experiments the transmittance was smaller than 0.7, for at least one wavelength of the photometer. Figure 12 shows the sensitivity vs. transmittance for two experiments with a 3- λ PSAP. The Bond correction underestimates the sensitivity to scattering for low loadings, and for loadings resulting in transmittance smaller than ~ 0.9 , an over-correction occurs. The remaining span of sensitivities from +2.5% to -0.5% almost explains the large uncertainty in the scattering correction given by Bond et al. (1999) and seen during the GAW2005 experiment. It can also be seen that the cross sensitivity to scattering increases with wavelength.

Similar results can be found for the Aethalometer (cf. Fig. 13). The apparent absorption diminishes with increasing loading. Also the dependence on wavelength can be seen. The cross sensitivities span a range from 3.5% to 0.5%. This range is larger than that observed for PSAP. However, if a similar wavelength range is considered for PSAP and Aethalometers, thus excluding the Aethalometer UV and near infrared channels, the span of sensitivities reduces to 2.5% to +0.5%, which is slightly less than the range observed for PSAP.

Figure 14 shows the sensitivity to scattering versus transmittance for the MAAP. The cross sensitivity covers the range from 3% to 0.5%. Within this range, the sensitivities

given by Petzold et al. (2005) and measured in the GAW2005 workshop are in agreement.

These experiments clearly show that a loading correction for purely scattering particles is necessary. In this manuscript we cannot explain the physical reasons for the wavelength dependence, but we showed the necessity to account for that effect.

A further point worth mentioning is the problem of preloading of the filter. Preloading is defined here as the situation where particles have been collected on the filter before the start of the measurements when the transmittance is set to unity. It can easily be seen (Fig. 13) that the cross sensitivity to particle scattering is very sensitive to the loading state, and to the transmittance of the filter. To our knowledge, this is the first time that this effect has been observed for Aethalometer. A loading dependent sensitivity for PSAP was observed during the RAOS for PSAP (Fig. 8 in Virkkula et al., 2005). Thus it is hard to compare results from different experiments without having proof that filters were clean before starting the experiment run. It is hard to give an average value for sensitivities to particle scattering because it depends on loading. Thus a loading dependent correction for particle scattering would be desirable.

6.4 Soot

The relative response to soot (carbon black) was investigated during GAW2005. A MAAP was chosen to be the *reference* instrument. Results are shown in Table 12. The response was calculated for each wavelength channel after adjusting the wavelength to the wavelength of the MAAP. Single wavelength PSAPs showed sensitivities from 0.95 to 1.1, and the 3 λ PSAP has a sensitivity between 0.8 and 0.85. Two 7 λ Aethalometers (after the Weingartner correction) showed very different responses. For one instrument, the response was 2.1 and 2.6, and the other instrument had a significantly lower response ranging from 1.49 to 1.53. Differences between the absorption measured by the MAAP, PSAP and Aethalometer cannot be explained. Possible reasons could be the different approaches used for correction. Another reason could be the large unit to unit variabilities of the PSAP and Aethalometer.

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During EUSAAR2007 only few experiments were performed. Because of technical and experimental problems these data are not reliable. Thus we can not present the relative responses to soot for PSAP and Aethalometer for the EUSAAR2007 workshop.

7 Summary and conclusions

5 We have presented a characterization of the PSAP, MAAP, and Aethalometer with respect to effective wavelength, instrumental noise, unit to unit variabilities, and relative sensitivity to absorbing and non absorbing aerosol particles. Results from two absorption photometer workshops are shown. While for MAAP absorption coefficients were not corrected, absorption coefficients for PSAP and Aethalometer were corrected using
10 the Bond- and Weingartner corrections, respectively.

The emission wavelengths of photometers showed that those of the Aethalometer agree with values given by the manufacturer. The actual wavelength of the MAAP is 637 nm instead of 670 nm as specified by the manufacturer. Consequently, absorption coefficients reported by the MAAP should be multiplied by a factor of 1.05 to obtain the
15 absorption coefficient at the true wavelength of 637 nm. The emission wavelength of the commercial single wavelength PSAP was determined to be larger than the specified 565 nm. A broad tail to longer wavelengths shifts the effective wavelength to about 585 nm. The three wavelength PSAP showed wavelengths of 467, 531 and 650 nm. Custom made single wavelength PSAPs have a wavelength of 532 nm.

20 Measurement of spot sizes revealed, that for MAAP there is no significant variation between different instruments. For PSAP and Aethalometer spot sizes can differ up to 6% and 8% from the nominal value. Since spot size and flow rate are going directly into the equation for calculating the absorption coefficient, we recommend to measure regularly spot size and flow rate.

25 The MAAP showed instrumental noise levels of 0.08 Mm^{-1} and 0.22 Mm^{-1} for a one minute averaging time during the GAW2005 and EUSAAR2007 workshops,

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respectively. Noise levels for the 3 λ PSAP determined with a one minute averaging time were similar for both workshops and were in the range from 0.05 Mm⁻¹ to 0.07 Mm⁻¹. In contrast, single wavelength PSAPs showed larger noise with average values of 0.36 and 0.15 derived for GAW2005 and EUSAAR2007, respectively.

Aethalometer noise strongly depends on the wavelength. With an averaging time of three minutes, the noise of the AE31 Aethalometer at wavelengths of 370 and 880 nm was 0.42 Mm⁻¹ and 0.17 Mm⁻¹ (GAW2005). During EUSAAR2007, the Aethalometer noise was estimated to be 0.27 Mm⁻¹ for a 3 min instrument averaging time.

Absorption coefficients measured with PSAP and Aethalometer were compared to absorption coefficients of MAAP, since unit to unit variability of seven MAAPs decreased from 11 to 3% after a flow calibration during EUSAAR2007. The relative sensitivity for ambient aerosol of the PSAP compared to the MAAP was 0.8 for 3 λ -PSAP and 0.86 for 1 λ -PSAP during GAW2005. 1 λ - and 3 λ -PSAPs showed no significant difference. During EUSAAR2007, average sensitivities of PSAPs were between 0.99 and 1.05 for two independent experiments. Aethalometer relative sensitivities for the 660 nm channel were on average 1.37 and 1.6 for GAW2005 and EUSAAR2007, respectively.

Relative sensitivity to non absorbing aerosol has been determined from measurements with ammonium sulfate. An average scattering cross sensitivity of 1.6% is included in the Bond-correction. The remaining cross sensitivity to scattering was on average 0.62% during GAW2005. For PSAP the cross sensitivity was on average 0.45% for 3 λ -PSAPs at all wavelengths and 1.08% for 1 λ -PSAPs. During EUSAAR2007, a loading and wavelength dependence has been observed. Cross sensitivities for low loadings (transmittance close to unity) were between 2% and 2.7%. With decreasing transmittance, the cross sensitivity became negative (-0.5%). Thus the Bond correction underestimates the cross sensitivity at low loading (transmittance>0.9) and overestimates it at higher loadings.

The cross sensitivity of MAAP was between 0.5% and 0.69% during GAW2005. A loading dependent investigation during EUSAAR2007 showed average values for the

cross sensitivity of 2.6% at a transmittance of unity and about 1% at a transmittance of 0.7.

For purely scattering aerosol the applied Aethalometer correction reduces to a multiple scattering correction. A strong wavelength dependence was observed during GAW2005. At 370 nm and 880 nm the cross sensitivities were on average 1.2% and 4.3%, respectively. In addition to a wavelength-dependence, a loading dependence was also observed during EUSAAR2007. At high transmittances (>0.96) the cross sensitivity reached 4.5% at a wavelength of 880 nm, which decreased to 1% at a transmittance of 0.8. For a wavelength range similar to that of PSAP, thus excluding the Aethalometer UV and near infrared channels, the span of sensitivities for Aethalometers reduces to 2.5% to +0.5%, which is similar to the range observed for PSAP.

For all three types of absorption photometers we recommend to operate the instruments within the specifications and to follow the maintenance procedures given by the manufacturers.

The authors wish to state that reference to a particular manufacturer or company in this paper is not an endorsement of the particular manufacturer or company.

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Table 1. Average values of properties of aerosol types used during GAW2005; maximum and minimum values are given between parentheses. Absorption coefficients were measured by PSAP and MAAP and scattering coefficients were determined by a nephelometer.

aerosol type	ambient air	ammonium sulfate ¹	carbon black
Effective radius, R_{eff} [μm]	0.141	0.056	0.087
Single scattering albedo ω_0 at 637 nm	0.92 (0.90, 0.94)	1.0 (by definition)	0.46 (0.45, 0.47)
ω_0 at 530 nm	0.90 (0.89, 0.91)	1.0 (by definition)	0.35 (0.33, 0.50)
Scattering coefficients, σ_{sp} at 550 nm [1/Mm]	97.37 (67.4, 126.6)	95.6 (89.7, 100.6)	56.7 (18.7, 90.3)
Absorption coefficients, σ_{ap} at 637 nm [1/Mm]	11.8 (8.3, 15.4)	0.0 (by definition)	119.8 (76.6, 137.4)
Ångström scattering exponents α_{sp} (450 and 700 nm)	1.39 (1.25, 1.50)	2.57 (2.51, 2.64)	1.02 (0.96, 1.30)
Ångström absorption exponents α_{ap} (460 and 700 nm)	1.14 (1.09, 1.24)	not defined	0.80 (0.73, 0.86)

¹ The single scattering albedo of ammonium sulfate is set to unity. It is assumed that the absorption coefficient is zero.

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Table 2. Average values of properties of aerosol types used during EUSAAR2007; maximum and minimum values are given between parentheses. For Ammonium sulfate and carbon black the minimum and maximum values are omitted, since single scattering albedo and Ångström exponent were very stable during experiments. The optical properties of ammonium sulfate are the same as those given in Table 1 for GAW2005, only the effective radius is different.

aerosol type	ambient air, two experiments	ammonium sulfate, two experiments
R_{eff} [μm]	0.15	0.045 0.097
ω_0 at 637 nm	0.75 (0.67,0.81) 0.86 (0.76,0.91)	1.0
ω_0 at 530 nm	0.78 (0.72,0.83) 0.87 (0.80,0.91)	
Scattering coefficients, σ_{sp} at 550 nm [1/Mm]	60.8 (38.6, 75.7) 107.2 (64.4, 216.4)	1045 (980, 1116) 570 (309, 1272)
Absorption coefficients, σ_{ap} at 637 nm [1/Mm]	17.66 (9.88, 29.2) 12.0 (6.39, 21.26)	not measured
α_{sp} (450 and 700 nm)	1.91 1.6	3.2 0.61
α_{ap} (460 and 650 nm)	1.08 (1.01, 1.20) 0.99 (0.76, 1.20)	not defined

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Table 3. Absorption photometers at the GAW2005 workshop.

Type	Nominal wavelength(s) [nm]	Actual wavelength(s) [nm]	Manufacturer	Serial Numbers/ Identification
PSAP	565 ¹	585	Rad. Res. ²	48, 20A, 20B, 13
PSAP	565 ¹	522	Rad. Res. ²	71,
3λ-PSAP	470, 530, 660 ⁵	467, 531, 650	Rad. Res. ²	90A, 90B
PSAP	532		custom made	MISU, ITM
MAAP	670 ¹	637	Thermo Elec. Inc. ³	1A, 13, 30, 32, 49, 50
Aethalometer model AE31	370, 470, 520, 590, 660, 880, 950		Magee Scientific ⁴	483, 563, 337
Aethalometer model AE21	370, 880		Magee Scientific ⁴	426
Aethalometer model AE9	white light		Magee Scientific ⁴	910101
Aethalometer model AE10	white light		Magee Scientific ⁴	70010

¹ Nominal wavelength given by manufacturer differs significantly from wavelength measured during workshop.

² Radiance Research, Seattle, WA

³ Thermo Electron Corporation

⁴ Aerosol d.o.o., Ljubljana Slovenia (under license of Magee Scientific Company Inc.)

⁵ Sheridan et al. 2005

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Table 4. Instrumentation used for aerosol characterization during the GAW2005 and EUSAAR2007 workshops.

Type	measured property	Manufacturer
Nephelometer Model, 3565	particle scattering- and back- scattering coefficient at wavelengths 450, 550, and 700 nm	TSI
TDMPs ¹	particle number size distribution from 10 to 650 nm electrical mobility	custom made
SMPS ²	particle number size distribution from 20 to 650 nm electrical mobility	custom made
APS, model 3321	Particle number size distribution from 580 nm to 10 μm aerodynamic diameter	TSI

¹ GAW2005

² EUSAAR2007

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Table 5. Aerosol absorption photometers at the EUSAAR2007 workshop.

Type	Nominal wavelength(s) [nm]	Manufacturer	Serial Numbers
PSAP	565	Rad. Res. ²	15(leak), 20, 28, 60, 80
PSAP	467, 530, 660	Rad. Res. ²	103,106,100, 483
PSAP	531	Custom made	MISU
PSAP	523±1	Custom made	Lund, ITML,NILU
MAAP	670 ¹	Thermo Elec. Inc. ³	13, 24, 34, 56, 59, 80, 81
Aethalometer model AE31	370, 470, 520, 590, 660, 880, 950	Magee Scientific ⁴	217,427, 351, 408
Aethalometer model AE16	880	Magee Scientific ⁴	199, 531

¹ Nominal wavelength given by manufacturer differs significantly from wavelength measured during workshop.

² Radiance Research, Seattle, WA

³ Thermo Electron Corporation

⁴ Aerosol d.o.o., Ljubljana, Slovenia (under license of Magee Scientific Company Inc.)

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Table 6. Nominal and measured wavelengths of optical absorption photometers.

Photometer type	Nominal wavelengths [nm]	Measured wavelengths [nm]	Full width at half maximum [nm]	Number of instrument
PSAP	565	585±6	35	4
3λ-PSAP	467, 530, 660	467, 531, 650	20, 40, 22	1
Custom made PSAP		532±2	40	1
MAAP	670	637±1	18±1	7
Aethalometer model AE31	370 470 520 590 660 880 950	376±2 473±2 525±7 593±4 654±4 858±5 940±1	11±1, 30±2, 34±2, 17±3, 23±2, 85±6, 50±10	3
Aethalometer model AE21	370, 880	378, 867	15, 80	1

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Table 7. Measured PSAP spot areas. The ratio of measured spot areas to the reference spot area of 20.43 mm² is given in parentheses.

	EUSAAR2007	GAW2005
number of PSAPs	11	6
measurements for each PSAP	8	6
average area [mm ²]	19.23 (0.94)	20.72 (1.01)
standard deviation [mm ²]	1.25	1.96

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Table 8. Measured Aethalometer spot areas. HS and ER refer to models with “high sensitivity” and “extended range” spots with nominal spot area 0.5 cm^2 and 1.67 cm^2 , respectively. The ratio of measured and reference spot area is given between parentheses.

	EUSAAR2007	GAW2005	
HS	number of instruments	3	3
	measurements for each instrument	3	3
	average area [cm^2]	0.52 (1.04)	0.46 (0.92)
	standard deviation [cm^2]	0.03	0.06
ER	number of instruments	2	1
	measurements for each instrument	3	1
	average area [cm^2]	1.71 (1.02)	1.60 (0.96)
	standard deviation [cm^2]	0.03	–

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Table 9. Instrumental noise measured during the workshops for different photometer types.

Photometer	Workshop	Averaging time [min]	number of instruments	Noise [1/Mm]		
				average	Min	Max.
MAAP	GAW2005	1	6	0.08	0.06	0.13
3λ-PSAP blue	GAW2005	1	1	0.06		
3λ-PSAP green	GAW2005	1	1	0.05		
3λ-PSAP red	GAW2005	1	1	0.05		
1λ-PSAP green	GAW2005	1	3	0.36	0.30	0.46
7λ Aethalometer (370 nm)	GAW2005	3	1	0.42		
7λ Aethalometer (880 nm)	GAW2005	3	1	0.17		
AE10 white light Aethalometer	GAW2005	3	1	1.84		
AE9 whitelight Aethalometer ¹	GAW2005	–	–	–	–	–
MAAP	EUSAAR2007	1	7	0.22	0.22	0.23
3λ-PSAP blue	EUSAAR2007	1	6	0.07	0.04	0.16
3λ-PSAP green	EUSAAR2007	1	6	0.07	0.04	0.15
3λ-PSAP red	EUSAAR2007	1	6	0.06	0.04	0.14
1λ-PSAP green	EUSAAR2007	1	2	0.15	0.14	0.16
7λ Aethalometer (all λs) ER spot ²	EUSAAR2007	3	2	0.24	0.18	0.30
7λ Aethalometer (all λs) ER spot ²	EUSAAR2007	3	2	0.38	0.36	0.40
1λ-Aethalometer (880 nm)	EUSAAR2007	3	2	0.19	0.14	0.23

¹ Data of the AE9 Aethalometer were not analyzed because of a diffuse spot due to a bad sealing.

² For comparability Aethalometer noise is converted to 3-min averages and equal face velocity (see Sect. 5.4 for explanatory text).

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Table 10. Relative response of Aethalometer and PSAP compared to MAAP for ambient aerosol. Absorption coefficients were adjusted to 637 nm using average Ångström exponents. For PSAP the average Ångström exponent was calculated using all three wavelengths of the multi wavelength PSAP and amounts 1.14 for GAW2005 and 1.08 and 0.99 for two experiments during EUSAAR2007. For Aethalometer only four wavelengths 590, 660, and 880 nm were used to determine the average Ångström which amounts 0.97 for GAW2005 and 1.06 for EUSAAR. The uncertainty is calculated from the unit to unit variability of the instruments. Correction methods are Bond (Bond et al., 1999) for PSAPs and Weingartner (Weingartner et al., 2003) for Aethalometers.

Instrument	$\sigma_X/\sigma_{\text{MAAP}}$	Workshop
3 λ -PSAP, 650 nm	0.81±0.07	GAW2005
1 λ -PSAP, 585 nm	0.86±0.27	GAW2005
Aethalometer, 660 nm	1.37±0.11	GAW2005
Aethalometer, white light	1.21 ¹	GAW2005
3 λ and 1 λ -PSAPs		
Exp.1	1.05±0.08	EUSAAR2007
Exp.2	0.99±0.10	EUSAAR2007
Aethalometer 660 nm	1.6 ± 0.20	EUSAAR2007

¹ For the wavelength adjustment to 637 nm it is assumed that effective wavelength is 840 nm

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Table 11. Response to ammonium sulfate at GAW2005. PSAP and Aethalometer data were corrected following Bond et al. (1999) and Weingartner (2003), respectively, to show the remaining cross sensitivity to particle scattering.

Instrument and S/N	wavelength [nm]	relative apparent absorption, (σ_{ap}/σ_{sp})*100%
MAAP 049	637	0.69
MAAP 01A	637	0.65
MAAP 050	637	0.63
MAAP 013	637	0.62
MAAP 030	637	0.63
MAAP 032	637	0.51
PSAP 20B	585	1.72
PSAP 071	530	0.89
PSAP 20A	585	2.18
PSAP 90A	467 (B)	0.34
PSAP 90A	531 (G)	0.42
PSAP 90A	650 (R)	0.52
PSAP 90B	467 (B)	0.16
PSAP 90B	531 (G)	0.42
PSAP 90B	650 (R)	0.89
PSAP 013 (not included in avg.)	585	3.12
PSAP 048	585	1.64
Aeth. 483	370	1.14
	470	1.63
	520	1.79
	590	2.12
	660	2.53
	880	4.86
	950	4.74

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Table 11. Continued.

Instrument and <i>S/N</i>	wavelength [nm]	relative apparent absorption, (σ_{ap}/σ_{sp})*100%
Aeth. 563	370	1.30
	470	1.61
	520	2.67
	590	2.57
	660	2.09
	880	5.06
	950	5.47
Aeth. 426	370	0.95
	880	3.97
Aeth. 337	370	1.14
	470	1.58
	520	1.57
	590	1.96
	660	1.99
	880	2.42
	950	2.81

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Table 12. Relative responses to carbon black. PSAP and Aethalometer data were corrected according to Bond et al. (1999) and Weingartner et al. (2003). Wavelengths were adjusted using the Ångström absorption exponent.

Workshop GAW2005		
Instrument and S/N	Wavelength/nm	Wavelength adjusted relative sensitivity $\sigma_{ap} / \sigma_{ap}^{MAAP}$
PSAP 20B	565 (G)	1.05
PSAP 48	565 (G)	0.94
PSAP 90B	460 (B)	0.84
	530 (G)	0.83
	650 (R)	0.8
PSAP 71	530	1.1
Aeth. 337	370	2.6
	470	2.13
	520	2.1
	590	2.11
	660	2.11
	880	2.15
	950	2.22
Aeth. 483	370	
	470	1.52
	520	1.49
	590	1.51
	660	1.53
	880	1.53
	950	

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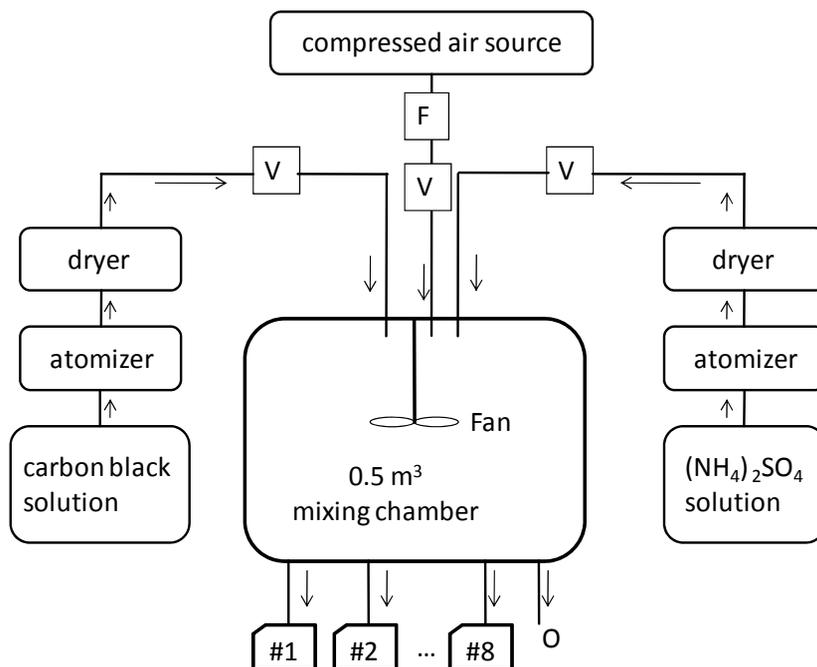


Fig. 1. Experimental setup for runs with ammonium sulfate and soot. Concentrations of ammonium sulfate and carbon black aerosols were adjustable. The aerosol was dried before entering the chamber. Direction of aerosol flow is indicated by arrows. In this diagram, F=filter, V=valve, O= overflow. Instruments were connected to output ports 1 to 8.

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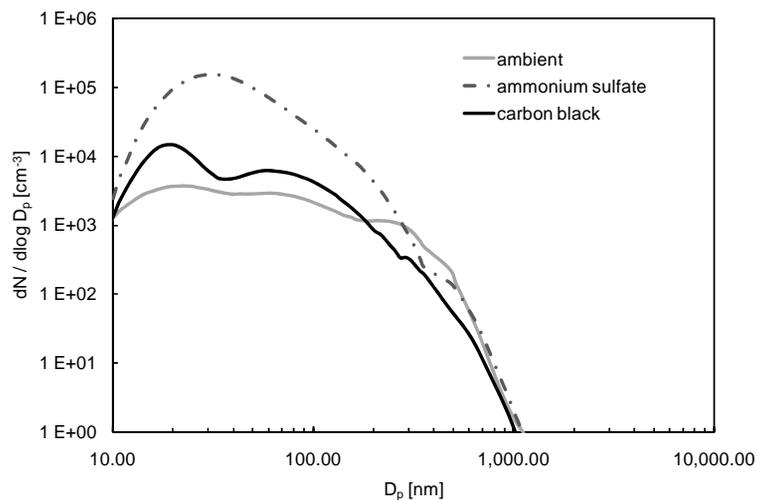


Fig. 2. Particle number size distributions for ambient air, ammonium sulfate, and carbon black during GAW2005. The particle number size distribution is a composite of number size distributions measured with a SMPS and an APS.

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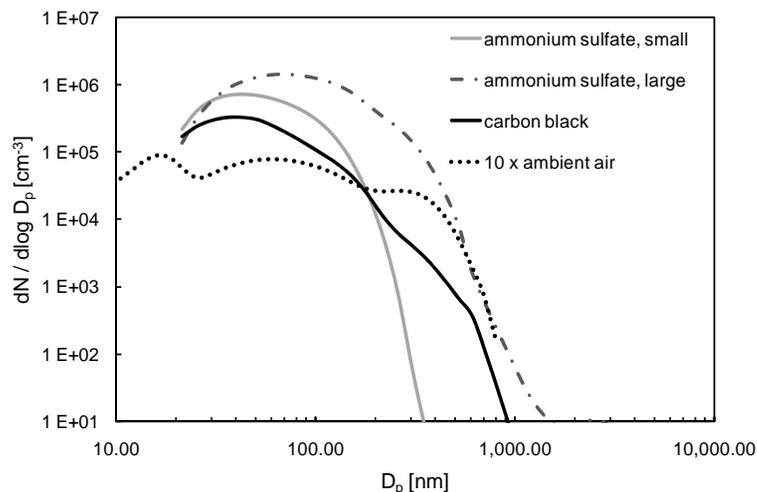


Fig. 3. Particle number size distributions for two sizes for ammonium sulfate, for carbon black, and for ambient air during EUSAAR2007. The number distributions for ammonium sulfate and carbon black are composites of number distributions measured with the SMPS and the APS. For ambient air APS measurements were not available. For visualization, the ambient air number concentration was multiplied by a factor of ten.

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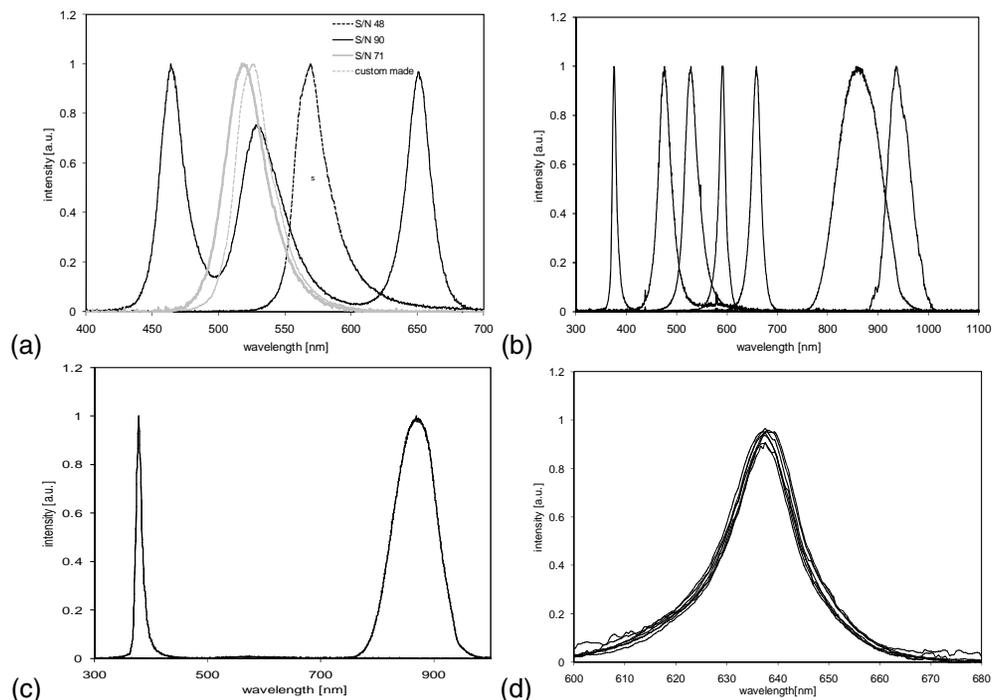


Fig. 4. Normalized spectral emission of light sources of absorption photometers. **(a)** different types of PSAPs; single wavelength Radiance Research PSAPs (S/N 48, S/N 71); three wavelength Radiance Research PSAP (S/N 90); custom made (ITM). **(b)** seven wavelength Aethalometer model AE31 (S/N 563). **(c)** two wavelength Aethalometer model AE21 (S/N 426) and **(d)** seven MAAPs.

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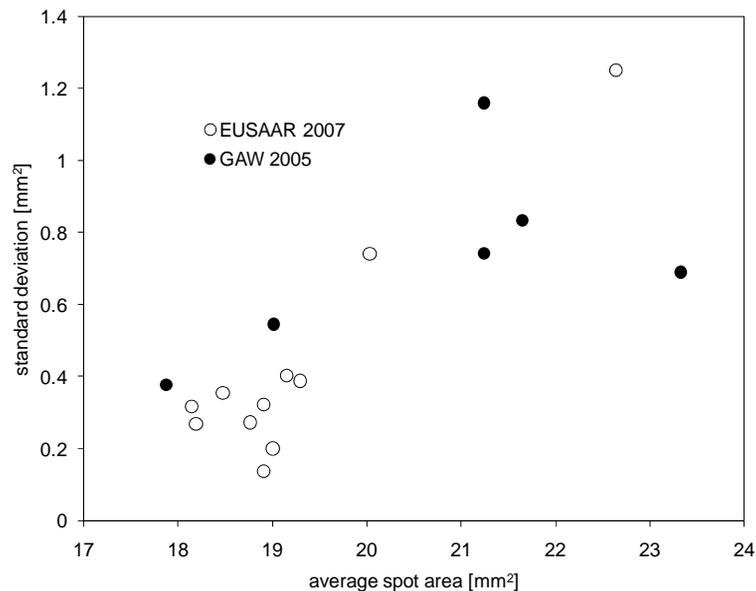


Fig. 5. Standard deviation of PSAP spot area versus average spot area. Each point is for one PSAP. The nominal spot area used by the manufacturers is 17.85 mm^2 .

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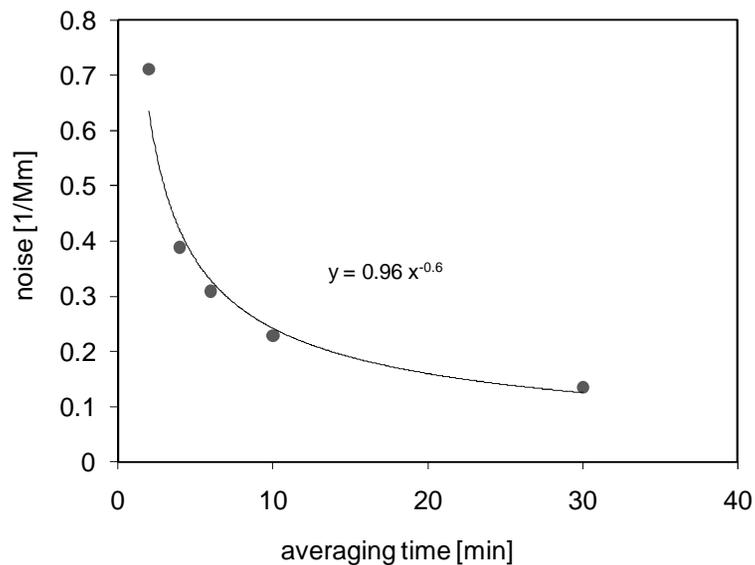
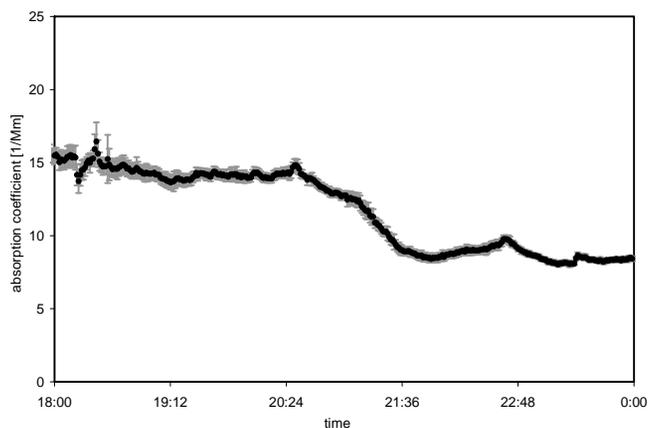


Fig. 6. Noise of Aethalometer vs. post-averaging time in minutes. The noise by post averaging almost follows the $\Delta t^{-0.5}$ law.

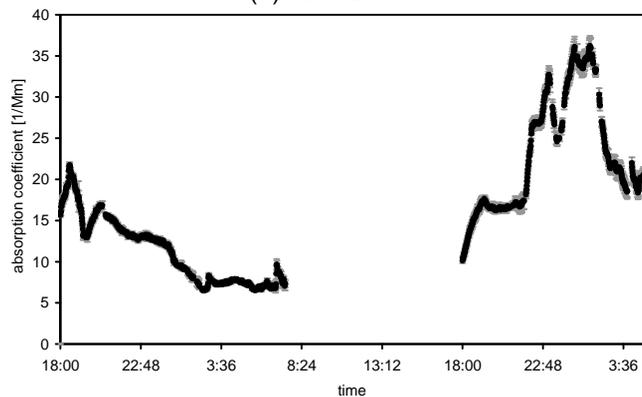
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(a) GAW2005



(b) EUSAAR2007

Fig. 7. Time series of MAAPs during the GAW2005 **(a)** and EUSAAR2007 **(b)** workshops. Error bars are the standard deviation of six (GAW2005) and seven (EUSAAR2007) MAAPs.

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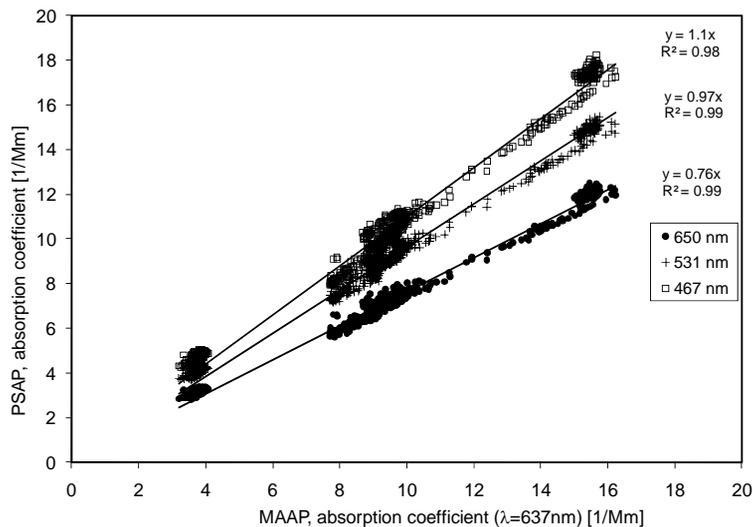


Fig. 8. Ambient air runs during GAW2005. PSAP absorption coefficients vs. MAAP absorption coefficients (637 nm) for the three PSAP wavelengths. The linear regression is forced through the origin. PSAP data were corrected using the Bond correction.

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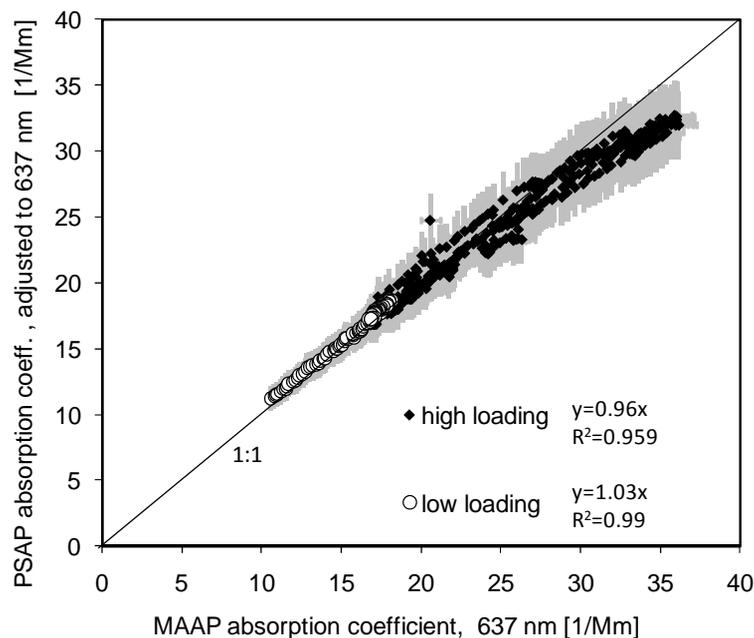


Fig. 9. PSAP absorption coefficients adjusted to 637 nm vs. MAAP absorption coefficients at 637 nm. PSAP data were corrected using Bond et al. (1999). The error bars represent the unit to unit variability of 3% and 8% for the MAAP and the PSAP, respectively. Data points for low loading are for data with PSAP transmittance between 1.0 and 0.7; high loading is for data with PSAP transmittance between 0.7 and to 0.2. (EUSAAR2007; ambient air).

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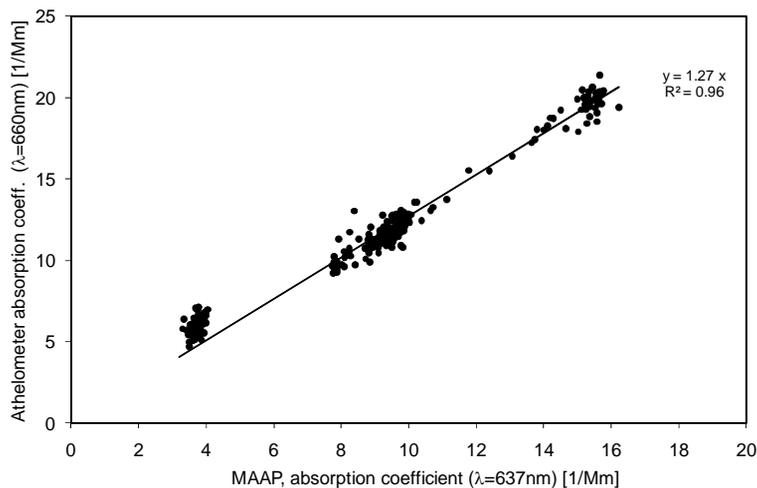


Fig. 10. Ambient air runs during GAW2005. Aethalometer (model AE31, 660 nm) absorption coefficient vs. MAAP absorption coefficient ($\lambda=637\text{ nm}$). Aethalometer data were corrected using Weingartner et al. (2003).

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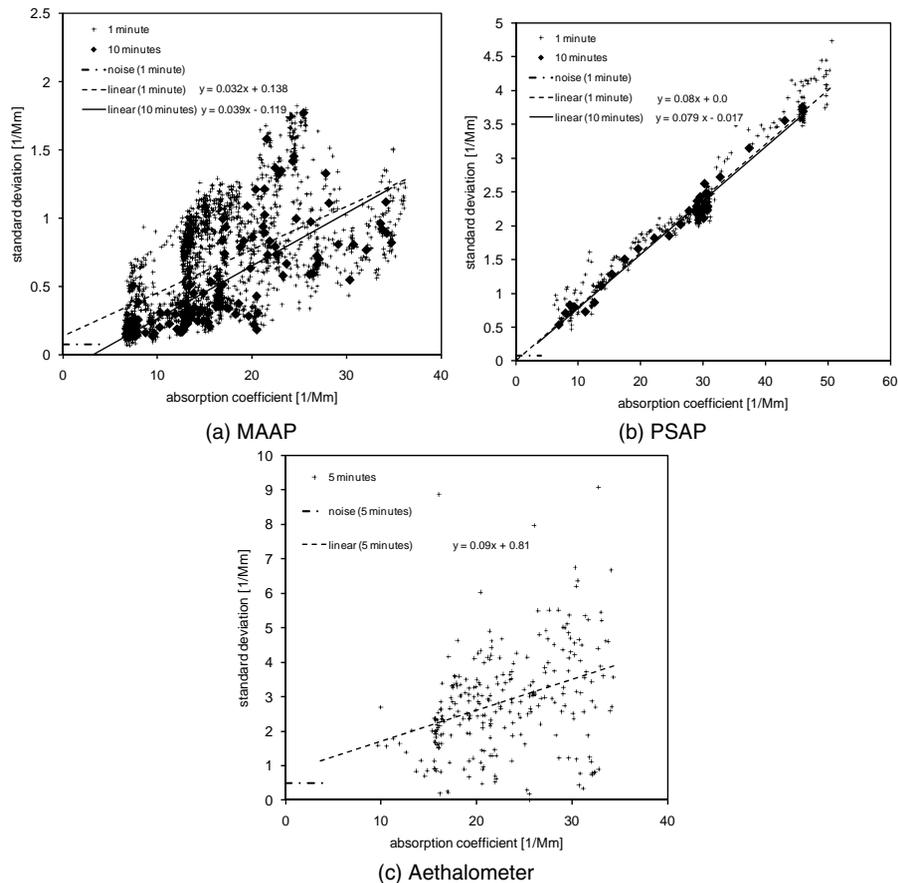


Fig. 11. Unit to unit variability (standard deviation of several instruments) vs. absorption coefficient for **(a)** MAAP (seven instruments), **(b)** PSAP (six 3 λ -PSAP, green) and **(c)** Aethalometer (four 7 λ -Aethalometers, 660 nm). PSAP and MAAP data are shown for the highest time resolution of one minute and for an averaging time of ten minutes. Aethalometer data are shown for an averaging time of 5 min. The noise level (one standard deviation) is indicated. (EUSAAR2007).

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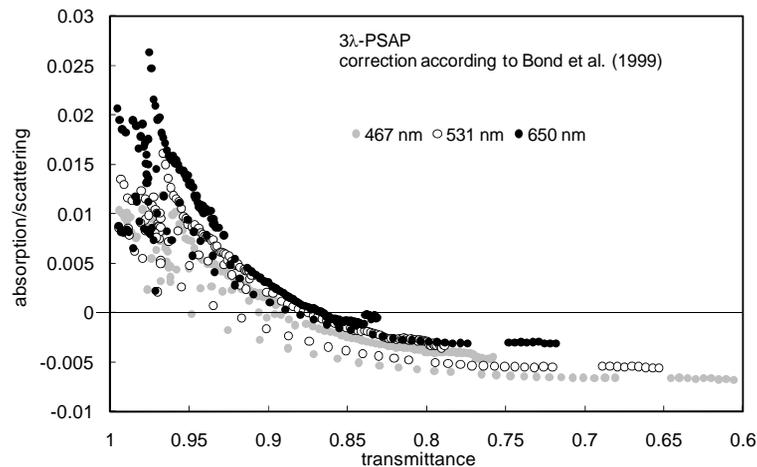


Fig. 12. Remaining cross sensitivity of absorption to particle scattering as a function of filter transmittance for the 3λ-PSAP. Absorption coefficients were corrected using Bond et al. (1999).

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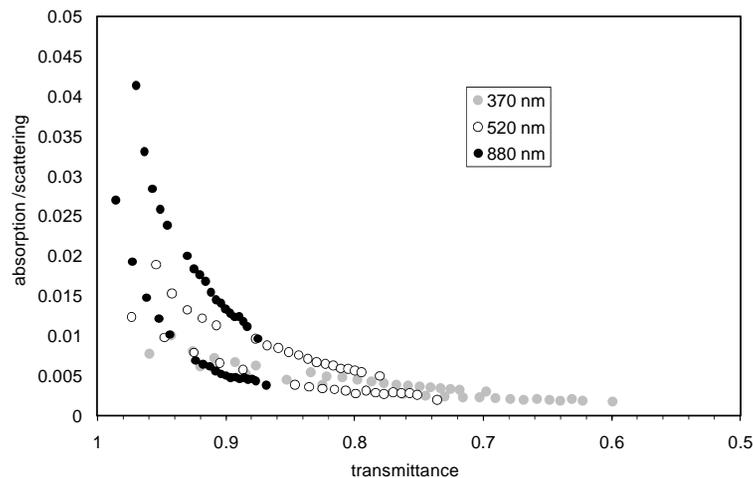


Fig. 13. Sensitivity to particle scattering as a function of filter transmittance for the 7 λ -Aethalometer. Shown are the ratios of absorption coefficients divided by scattering coefficients for three wavelengths. Absorption coefficients were corrected using Weingartner et al. (2003).

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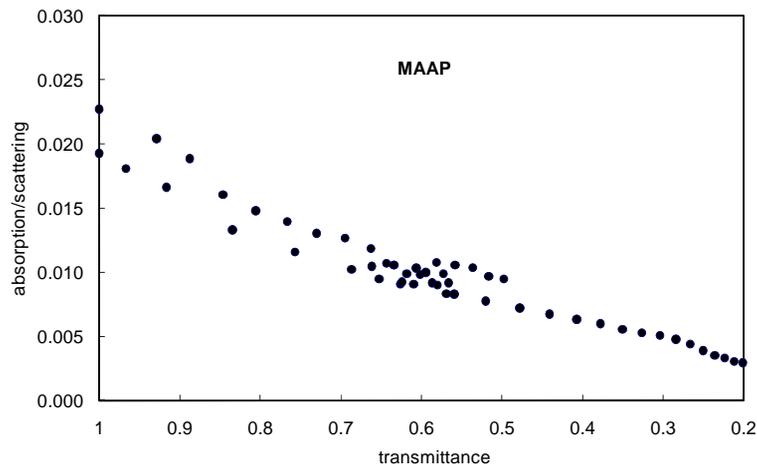


Fig. 14. Cross sensitivity of absorption to particle scattering as a function of filter transmittance for MAAP.

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