WCCAP Report

Absorption Photometer Workshop 2007

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1 Introduction

The first absorption photometer workshop was held in Leipzig at the Leibniz Institute for Tropospheric Research, November 2005. In March 2007 a second workshop was conducted to answer specific questions. This report summarizes results of the second workshop. It is planned to publish the results of both workshop in 2008.

1.1 Background

Spectral aerosol light absorption is an important parameter for the assessment of the radiation budget of the atmosphere. Many on-line measurement techniques for aerosol light absorption, such as the Aethalometer, the Particle Soot Absorption Photometer (PSAP), and the Multi Angle Absorption Photometer (MAAP) have been available for several years. The commercial single-wavelength PSAP was modified to measure at three wavelengths (Virkkula et al. , 2005a). A 7-wavelength Aethalometer became commercially available, which covers the visible (UV/VIS) to near-infrared (NIR) spectral range (λ=370–950 nm).

A limiting factor for all filter based absorption photometers is the cross sensitivity to particle scattering. Furthermore a loading dependent calibration factor complicates the evaluation of a calibration function. In the last years several correction schemes were developed for different types of instruments. Bond et al. (1999) introduced a first scattering correction for the PSAP. The correction is limited to low filter loadings and to low single scattering albedos. The Multi Angle Absorption Photometerie (MAAP) reduces the cross sensitivity to scattering but there is still a scattering artifact of about 1% of scattering which was seen in the workshop 2005. This value also was seen by Petzold et al. (2005).

Thus the focus of the 2nd workshop was to investigate scattering artifacts depending on particle size and loading. The data collected during both workshops will be used in future to find better correction schemes for absorption photometers. In this report we can give new correction functions we only will present a set of data which points out where we expect uncertainties using standard correction schemes (e.g. Bond, 1999, Weingartner, 2003).

1.2 Goals of the inter-comparison workshop

Specific goals are:

a) to determine the unit to unit variability of instruments,

b) to determine the relative sensitivities of the different instruments types to absorbing and scattering particles,

c) to investige particle size and loading effects.

Several experiments were done which were dedicated to achieve the specific goals. Different types of aerosols were used.

A brief overview of the workshop is given in the following chapters. A description of the experimental setup including the instruments and the aerosol generation is given in Chapter 2. In Chapter 4 a characterization of the instruments is given. The results of the inter-comparison are given in Chapter 5, and a discussion follows in Chapter **Fehler! Verweisquelle konnte nicht gefunden werden.**.

1.3 Organization

The workshop was held at the Leibniz Institute for Tropospheric Research (IfT) in Leipzig from 12 to 23 March, 2007. IfT provides several laboratories for the workshop. The laboratories were equipped with the aerosol generators and systems for a basic aerosol characterization. 36 participants attended the workshop with 34 systems for measuring the particle absorption.

Table 1: List of participants

surname	**first name**	**institution**
Angelov	Hristo	Basic Environmental Observatory Moussala at INRNE / BEO
Bizjak	Mirko	Environmental Agency of the Republic of Slovenia, Ljubljana, Slovenia
Covert	Dave	University of Washington Department of Atmospheric Sciences Seattle WA USA 98195-4235
dos Santos	Sebastiao	Joint Research Centre, Ispra, Italy
Engström	Erik	MISU, Department of Meteorology Stockholm University
Garland	Rebecca	mpi Mainz
Gruening	Carsten	Joint Research Centre, Ispra, Italy
Henzing	Bas	TNO - Netherlands Organization for Applied Scientitic Research, Apeldoorn, The Netherlands
Hoffer	András	University of Pannonia, Department of Earth and Environmental Sciences, Veszprém, Hungary
Imre	Kornelia	University of Pannonia, Department of Earth and Environmental Sciences, Veszprém, Hungary
Ivanow	Peter	Basic Environmental Observatory Moussala at INRNE / BEO
Karlsson	Hans	Hans Karlsson, Department of Applied Environmental Science(ITM), Stockholm university

Komppula	Mika	Leibniz Institute for Tropospheric Research / Finnish Meteorological Institute
Laborde	Marie	EcoTech
Lunder	Chris	NILU, Norwegian Institute for Air Research, Norway
Marinoni	Angela	ISAC, Institute for Atmospheric Science and Climate - CNR, Bologna, Italy
Moerman	Marcel	TNO D&V, Oude Waalsdorperweg 63, 2597AK The Hague, The Netherlands
Müller	Thomas	IFT, Leibniz Institute for Troposheric Research, Leipzig, Germany
Pichon	Jean-Marc	LaMP/OPGC, Laboratoire de Météorologie Physique, Clermont-Ferrand, France
Plass-Dülmer	Christian	DWD, Meteorologisches Observatorium Hohenpeissenberg, Germany
Roger	Jean-Claude	LaMP/OPGC, Laboratoire de Météorologie Physique, Clermont-Ferrand, France
Sachin	Gunthe	mpi Mainz
Sangeeta	Sharma	Environment Canada, 4905 Dufferin Street, Toronto, Ontario, Canada
Shao-Meng	Li	Environment Canada, 4905 Dufferin Street, Toronto, Ontario, Canada
Sheridan	Patrick	NOAA
Teinilä	Kimmo	Finnish Meteorological Institute
Verhagen	Henk	TNO - Netherlands Organization for Applied Scientitic Research, Apeldoorn, The Netherlands
Viana	Mar	CSIC, Spanish Research Council, Barcelona, Spain
Virkkula	Aki	Finnish Meteorological Institute
Wiedensohler	Alfred	IFT, Leibniz Institute for Troposheric Research, Leipzig, Germany
Wilhelm	Reinhard T.	DWD, Meteorologisches Observatorium Hohenpeissenberg, Germany
Ya-Qiang	Wang	Centre for Atmosphere Watch and Services (CAWAS), Chinese Academy of Meteorological Sciences (CAMS)
Nowak	Andreas	IFT, Leibniz Institute for Troposheric Research, Leipzig, Germany
Tuch	Thomas	IFT, Leibniz Institute for Troposheric Research, Leipzig, Germany

Table 2: Instruments and instrument IDs

Instrument class	Instrument	Number of instruments
Nephelometer Measured quantity: scattering coefficient	TSI, multi-wavelength	14
	TSI, single-wavelength	2
	Ecotech, multi-wavelength	1
	Ecotech, single-wavelength	3
	Radiance Research	2
Absorption photometer Measured quantity: Absorption coefficient	MAAP	8
	PSAP, multi-wavelength	4
	PSAP, single-wavelength	8
	PSAP, home made	4
	Aethalometer, multi-wavelength	4
	Aethalometer, single-wavelength	3
Measurement of reference absorption	Photoacoustic absorption photometer	2
	Extinction cell, home made (difference of extinction and scattering)	1

2 Experimental Setup

2.1 Aerosol Sources

The aerosols used during the workshop were:

a) Ambient aerosol: Ambient aerosol was used for an inter-comparison of al instruments in parallel. For that experiment the air conditioning of the laboratory was switch off and the windows were opened.

b) Ammonium sulfate: For investigation of the cross sensitivity to purely scattering particles. Ammonium sulfate particles were generated using an atomizer.

c) Soot: The sensitivity to strongly absorbing particles was measured with soot (Printex 75) generated by an atomizer.

The aerosols b), c) were fed into a mixing chamber (0.5 m^3) which was used as a buffering volume. The instruments were connected to the eight output ports of the mixing chamber. Two ports were used for additional aerosol characterization using a DMPS/APS for measuring the number size distribution and integrating nephelometer (TSI, model 3563) the scattering coefficient was measured using an integrating nephelometer for measuring the particle scattering coefficient.

2.2 Aerosol characterization

2.2.1 Nephelometer

The total scattering coefficient and the backscattering of the aerosols were measured using an integrating nephelometer (TSI, model 3563) three wavelengths (450 nm, 550 nm, and 700). Because of a truncation error the recorded total scattering and backscattering coefficients are smaller than the “real” scattering coefficients. A correction of this so called “truncation error” (Anderson et al., 1996) was not applied, because the scattering correction for the PSAP data according to Bond et al. (1999) requires the uncorrected data from the nephelometer.

2.2.2 DMPS and APS

The particle number size distributions were recorded using a Differential Mobility Particle Sizer (DMPS) in the size range 10 nm to 600 nm. Larger particles were recorded using an Aerodynamic Particle Sizer (APS, TSI model 3321). A more detailed description can be found in the report of the first workshop.

3 Instrument descriptions

In total twelve commercial PSAPs (Radiance Research) and four home made PSAPs were compared during the workshop. Furthermore eight MAAPs, seven Aethalometers, one extinction cell and two photoacoustic photometers were used.

3.1 PSAP

The functional principle of the Particle Soot Absorption Photometer (model PSAP, Radiance Research, Seattle, WA) is described in Bond et al. (1999). The PSAP with the nominal wavelength 567 are referred as the *old* PSAP. In the *new* PSAP the light source was replace by a diode emitting light at a shorter wavelength of about 532 nm. In addition the opal glass plate between light source and particle filter was replaced by a diffusely scattering hemisphere. A further modification of PSAP led to the 3-wavelength PSAP (Virkkula et al., 2005a). The optical wavelengths are approx. 467, 531, and 650 nm. The wavelengths of all PSAPs were checked using an optical spectrometer. The method and results are given in Chapter 4.3.

In the last years correction schemes were developed by Bond et al. (1999) (in the following referred as *Bond correction*) and a et al. (2005). Although the Bond correction was for the *old* PSAP type, we apply it to the other types of PSAPs at all wavelengths. With this approach we want to ease the inter-comparison of all PSAPs, especially for an investigation of the unit to unit variability. Because of the fundamental approach for correcting the scattering artifact and the extrapolation of this correction to other wavelengths the correction scheme is described in more detail.

This correction method accounts for loading and scattering correction. A correction of the aerosol flow and correction of spot size has to be done. All these corrections were included in the correction method which is referred as *Bond correction*. These corrections were developed for the *old* PSAP having a nominal wavelength of 567 nm. However the applied scattering correction uses scattering coefficients at 550 nm, the center wavelength of the green channel of a nephelometer (TSI, model 3563). It is worth note that the scattering coefficients used for the Bond correction are not corrected for the so called truncation error (Anderson 1996, Heintzenberg 2006).

The change of the optical wavelength required to calculate scattering coefficients for the other wavelengths. The interpolation and extrapolation of scattering coefficients was done using the Ångstroem exponent α which is defined as: $\sigma_{sp}(\lambda_2) = \sigma_{sp}(\lambda_1) \cdot \left(\frac{\lambda_2}{\lambda_1} \right)^{\alpha}$.

With the scattering coefficients measured at 3 wavelengths an average Ångstroem exponent is calculated. The Ångstroem exponent was used for an interpolation of scattering coefficients for wavelengths between 450 nm and 700 nm.

The formula for the Bond correction is:

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

In this formula A is the sample spot area, V the volume flow, τ the optical transmission and I(t) the measured intensity.

3.2 MAAP

The MAAP) (multi angle absorption photometer) was recently introduced and 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). The advantage of the MAAP compared to PSAP and Aethalometer is the in-situ correction of the scattering artifact.

3.3 Aethalometer

Four types of Aethalometers were used during the workshop. These were two white light Aethalometers (AE-9 and AE-10), one 2λ-Aethalometer (AE 21), and three 7λ- Aethalometers (AE-31). A more profound description of the Aethalometers can be found in the user manual (Hansen, 2005). To get the absorption coefficient the Aethalometer data need to be corrected.

The Aethalometer in principle measures the absorbance A(λ) ($A(\lambda) = -\ln\left(I(\lambda) / I(\lambda)_0\right)$). The Absorbance is related to the total BC (Black Carbon) concentration [BC] by $A(\lambda) = Sigma_{BC}(\lambda) \cdot [BC]$. The specific attenuation $Sigma_{BC}$ depends on the optical wavelength (see Table 3.1). It is

worth to note, that the specific attenuation is not a physical constant. The specific attenuation coefficient is the product of the mass absorption coefficient and a correction function for the Aethalometer, which is needed to relate the temporal evolution of the absorbance to the physical absorption coefficient.

Table 3: Specific attenuation coefficient used for Aethalometer (Hansen (2005))

λ[nm]	$Sigma_{BC}$ [m^2/g]
370	39.5
470	31.1
520	28.1
590	24.8
660	22.2
880	16.6
950	15.4

The first correction of the Aethalometer data to absorption coefficients including a loading and scattering correction was reported by Weingartner (2003) for laboratory aerosols. Further correction methods were given by Arnott (2005) and Schmid (2006). The latter one is based on the method of Weingartner, but the resulting absorption coefficients are lower by about 30% for ambient aerosol. This discrepancy of calibrations has to be taken into account, when comparing absorption coefficients measured with Aethalometer and PSAP or MAAP.

3.4 Custom made photometers

Four custom made instruments were used during the workshop. The optics is similar to the commercial PSAP. Three of them are one wavelength PSAP like instruments (NILU, ITM, Lund). The main difference is the implementation of a rotating disc filter holder.

3.5 Photoacoustic absorption spectrometer

Two photoacoustic absorption spectrometers (PAS) were intended to be the reference instrument for measuring the absorption coefficient. Differences between these instruments were discussed in a separate chapter.

3.6 Extinction minus scattering

A three wavelength extinction cell was used to determine the absorption coefficient by subtracting the also measured scattering from the extinction coefficient. A detailed description can be found in Virkkula et al. (2005b). The data provided by this approach couldn’t be used because of a low signal to noise ratio. After the workshop a leak in the cell was found what causes the high noise.

4 Instrument characterization and calibrations

4.1 Flow-rate

The flow rates of all instruments were calibrated using a primary flow standard. The Operating conditions of the instruments were room temperature and atmospheric air pressure. The absorption and scattering coefficients were corrected to STP (standard temperature and pressure, T=0°C and P=1013.25 hPa) conditions.

PSAP: The aerosol flow is corrected to STP conditions.

MAAP: Depending of the instrument settings the values are given for volume or mass flow at STD. The temperature used for STP correction can be change be the user. A flow calibration following the instructions in the user manual was done for all MAAPs.

Aethalometer: Depending of the instrument settings the values are given for volume or mass flow at STD. The temperature and pressure used for STP correction can be change be the user.

4.2 Spot sizes

The calculation of the absorption coefficient requires the “true” area of the sample spot. Variations from the nominal area given by the manufacturer have to be corrected. Therefore it is necessary to measure the area of each sample spot.

PSAP

The spot sizes for each PSAP were measured several times. The diameter ranges from 5.3 to 4.8 mm for the individual instruments. On average the spot diameter was 5.02 for all PSAPs. The variability and the average of all PSAP are similar to the results of the first workshop. It is recommended that for all PSAP an individual spot correction factor is introduced as described in Bond (1999)

MAAP

No deviation from the nominal spot size can be observed.

Aethalometer

Spot sizes were not measured during the workshop. In the report of the first absorption photometer workshop in 2005 several sample spot were measured.

4.3 Optical wavelength of PSAP light sources

The spectral emission the light sources of absorption photometers were measured using a fiber optic spectrometer (HR2000, Ocean Optics Inc.). In the 2nd absorption photometer workshop only the spectra of PSAPs were measured, because of different types of light sources and large unit to unit variations in the emitted spectra. In the first workshop also the spectral emission of MAAP and Aethalometer was measured.

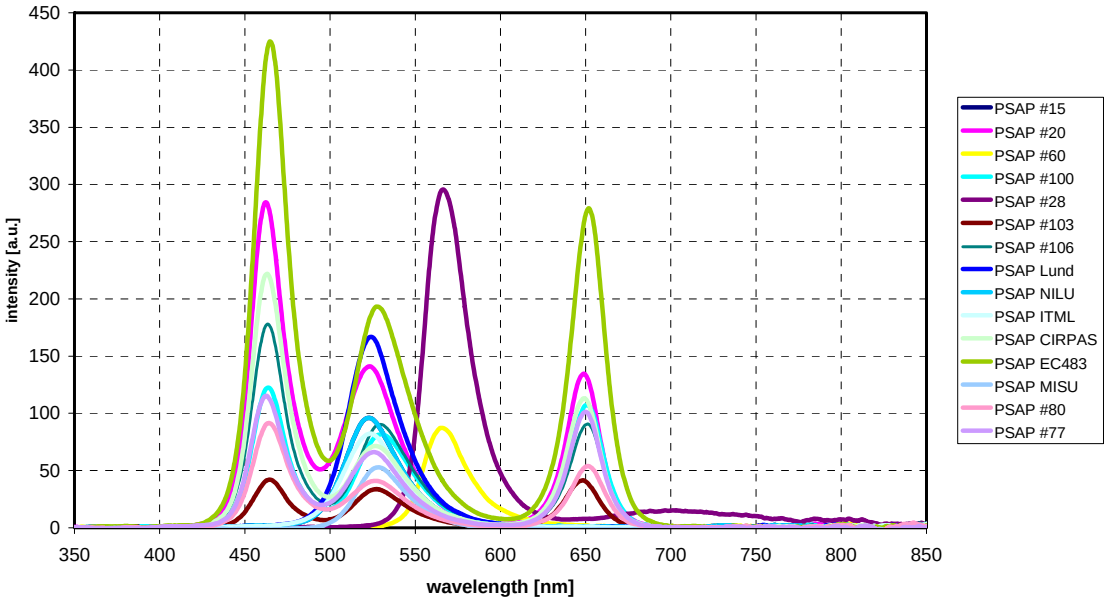

Figure 1: Spectral emitted radiation of PSAP light sources.

5 Results

For inter-comparison of instruments working at different wavelengths all instruments were corrected to 532 nm using the measured Angstroem absorption exponent of 1.08 for ambient aerosol.

5.1 Reference absorption

It was intended to derive reference absorption from the two photoacoustic absorption photometers (DMT-PAS and MPI-PAS) and the extinction cell (extinction minus scattering).

The MPI-PAS suffers from a not working automatic zero measurement. Thus data were corrected afterwards using time periods with filtered particle free air. A possible drift of the zero during overnight runs is possible and can not be accounted for. An inter-comparison (Figure 2) of DMT-PAS and MPI-PAS show that the relative sensitivity differs by a factor between 2.1 and 1.4 depending on the type of aerosol. This issue is still under discussion. The second possible reference measurement was the measurement of the difference of extinction and scattering. The extinction cell showed up a relatively high noise level, which doesn't allow the use of the extinction cell for reference measurements. After the workshop the reason of the high noise was found in a leak in the cell. Figure 3 shows a comparison of extinction minus scattering with MPI-PAS.

Because of these problems it was decided to use the absorption reported by MAAP as secondary reference. The reasons for the choosing MAAP are:

(1) During the RAOS experiment good agreement between MAAP and extinction minus scattering was found,

(2) The MAAP has the lowest cross sensitivity to particle scattering (result of the first absorption photometer workshop)

(3) The unit to unit variability is almost negligible (see Fig. 8).

An inter-comparison of MAAP and MPI-PAS (Fig. 4) shows a good correlation between both methods. The high intercept of the correlation line might be caused by an incorrect zero measurement of the PAS as described above.

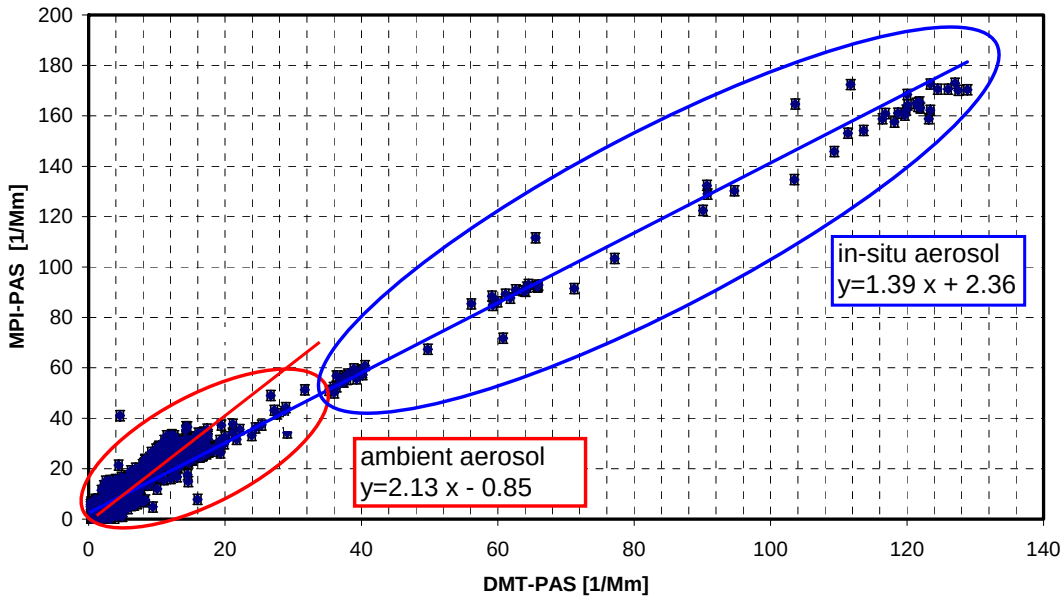

Figure 2: Inter-comparison of photoacoustic absorption photometer with ambient aerosol and soot. Both systems were corrected to a wavelength of 532 nm.

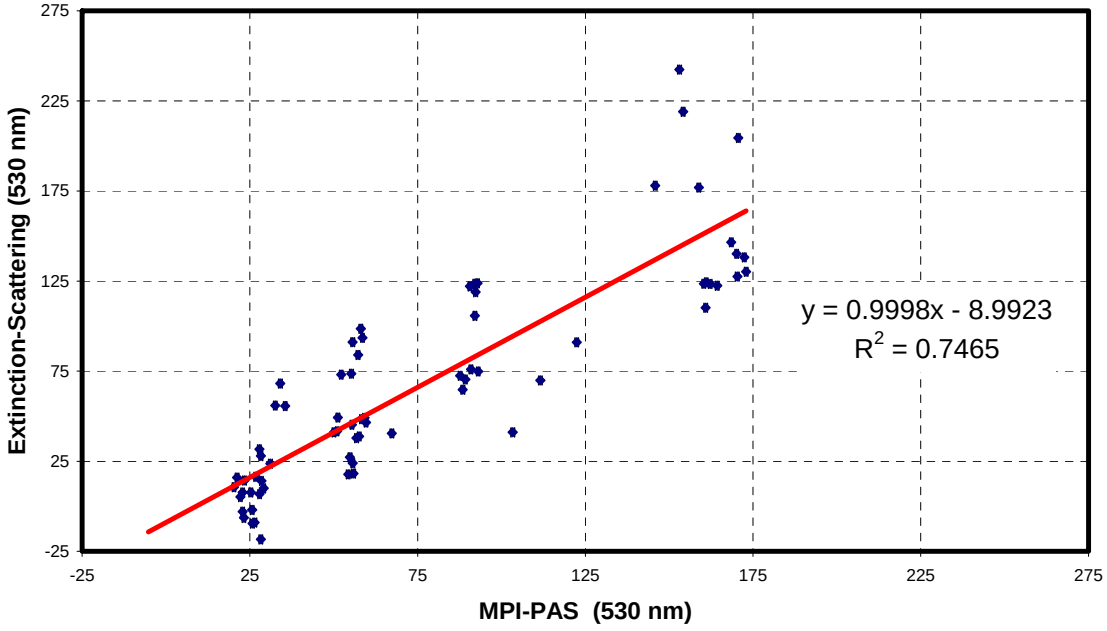

Figure 3: Comparison of extinction minus scattering vs. MPI-PAS using soot. Wavelengths are corrected to 532 nm.

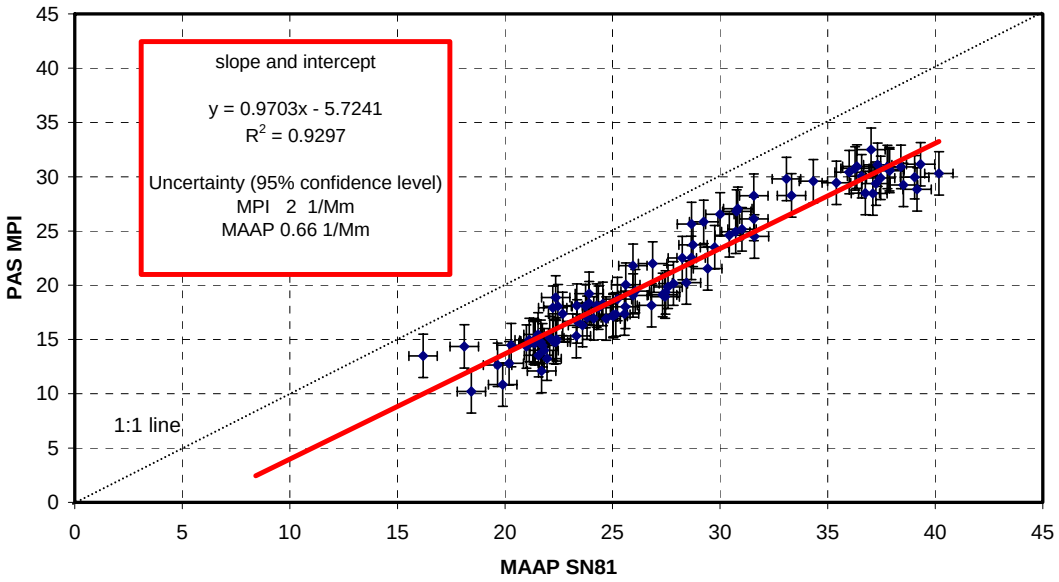

Figure 4: MPI-PAS versus MAAP. Instruments were corrected to a wavelength of 532 nm.

5.2 Inter-comparison of PSAP

One overnight run was selected for inter-comparison of ten Radiance Research PSAPs. Flow and spot size corrections were done for each PSAP and the absorption coefficients were corrected to 532 nm. The average and standard deviation are plotted in Figure 5. Average standard deviations and relative standard deviations are given for 3 ranges filter loading, a moderately loading with transmission Tr between 1 and 0.7, a high loading (0.5<Tr<0.7), and an overloading (Tr<0.5). The standard deviation is much larger then the noise level of PSAP, which is about 0.08Mm^{-1} (c.f. Figure 6).

For correction of the home made PSAP the bond correction was applied. Figure 7 shows an inter-comparison Radiance Research PSAPs. For home made PSAP are well correlated to the Radiance Research PSAPs. One of the home made PSAP has a sensitivity which is about 20% higher. Possible reasons for this deviation are a wrong flow or spot measurement. Anyway, the high linear correlation coefficient shows that the Bond correction can be applied to the home made PSAPs.

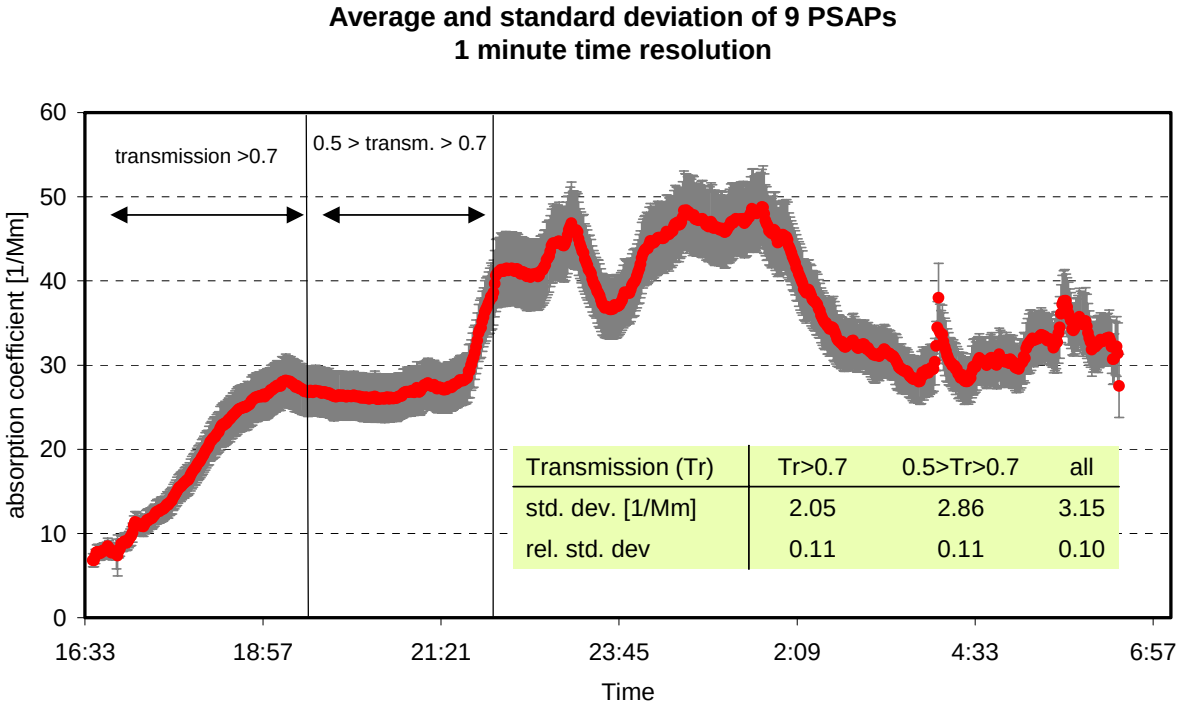

Figure 5: Average and standard deviation of nine Radiance Research PSAPs.

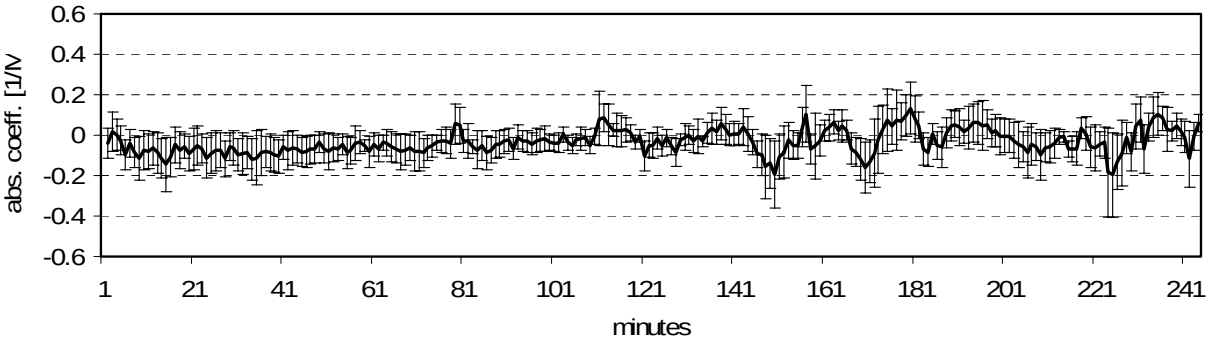

Figure 6: Noise test of ten PSAPs with particle free air. Shown are the average and standard deviation with a time resolution of one minute.

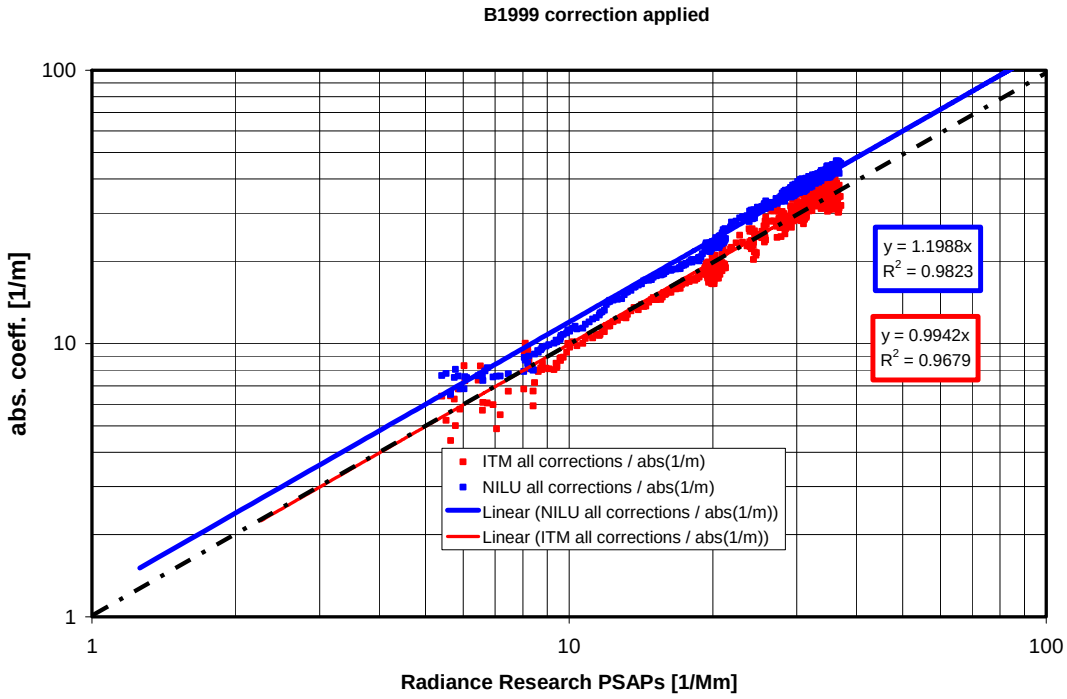

Figure 7: Inter-comparison of two home made PSAPs with Radiance Research PSAP.

5.3 Inter-comparison of MAAP

An inter-comparison of MAAPs before and after re-calibration of the aerosol flow, and the dependence on the flow rate is shown in Figures 8 and 9. It can be seen that the relative standard deviations after re-calibration is significantly smaller. After re-calibration the relative standard deviation amounts about 3% and the absolute standard deviation is about 0.05Mm^{-1}. A possible effect due to different flow rates (1000 and 600 l/h) was not seen.

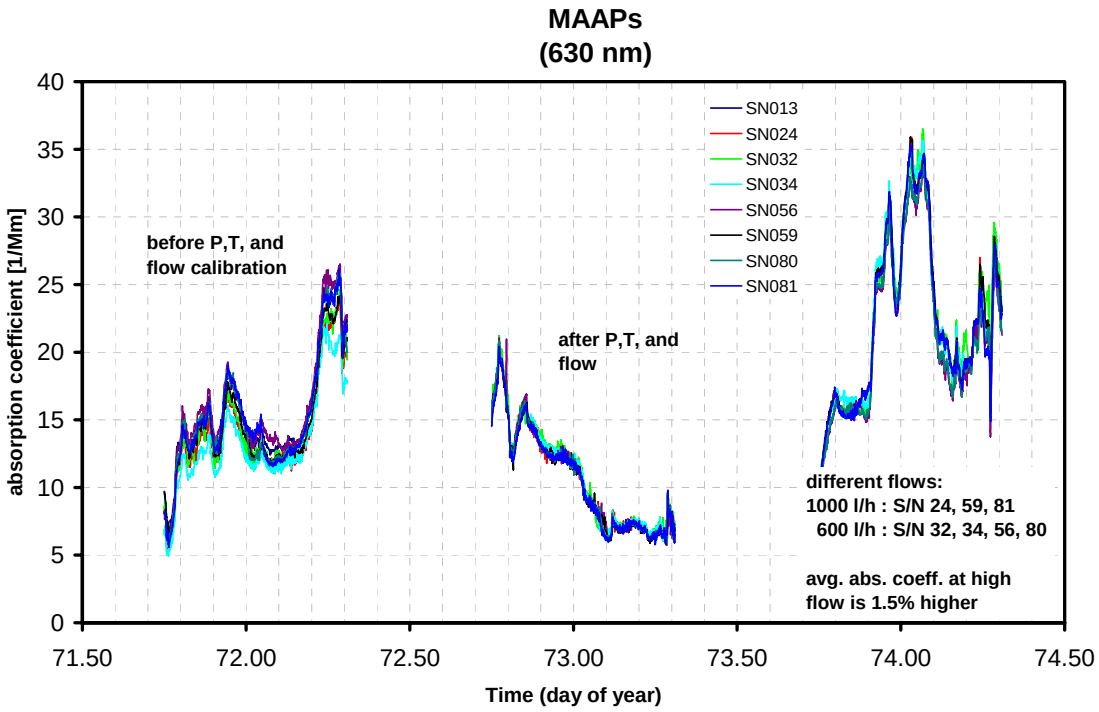

Figure 8: Inter-comparison of MAAPs before and after re-calibration. During the third period MAAPs were operated with a flow 1000 and and 600 l/h, respectively.

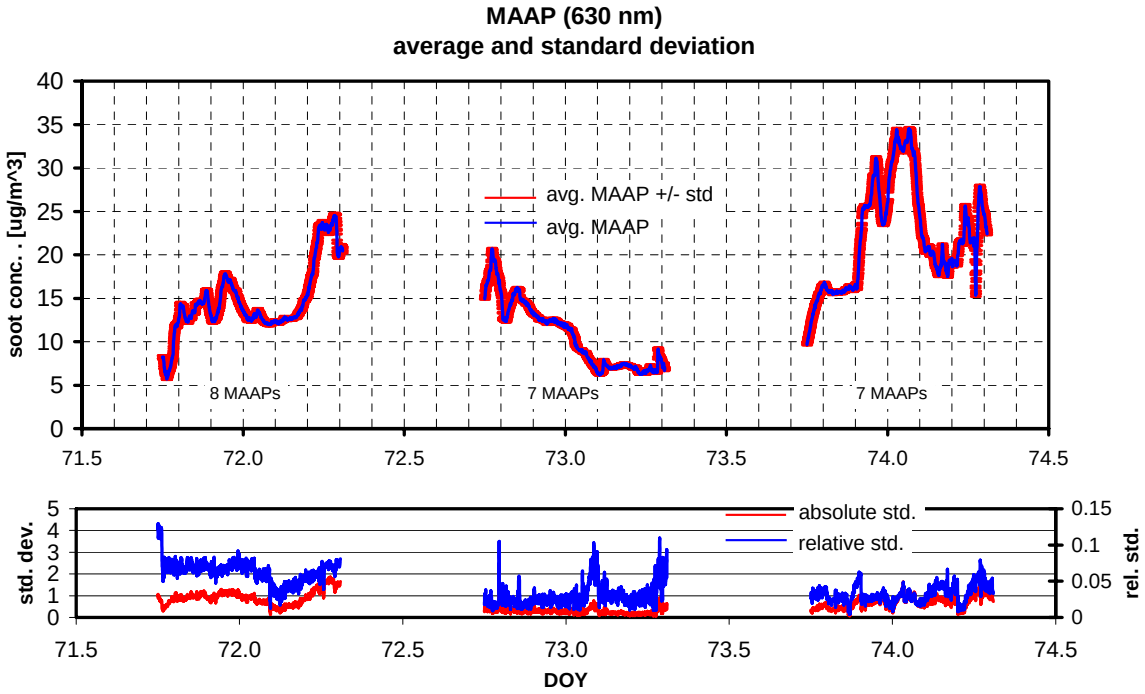

Figure 9: Average and standard deviations of MAAP before and after re-calibration and with different flow rates.

An inter-comparison of MAAP with PSAP and photoacoustic absorption photometers is shown in Figure 10. The PSAP and MAAP agree within their uncertainties which are dominated by the unit to unit variability.

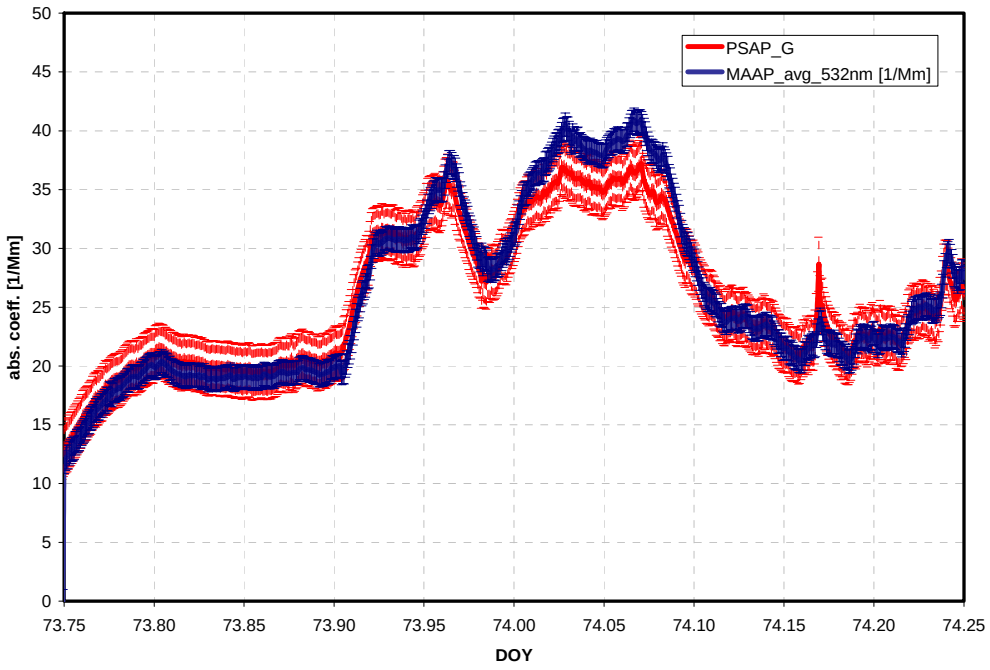

Figure 10: Inter-comparison of PSAP and MAAP.

5.4 Aethalometer

For inter-comparison of Aethalometers a loading correction for ambient aerosol was developed. It was found that the ratio of measured attenuation with Aethalometer and the absorption coefficient measured with MAAP was a linear function of the total filter loading. This correction only is valid for the aerosols which are similar to the ambient aerosol during the workshop.

The unit to unit variability for Aethalometers is smaller that observed during the first absorption photometer workshop. One reason could be that the sample spots were well defined in contrast to the first photometer workshop, where we observed many diffuse spots.

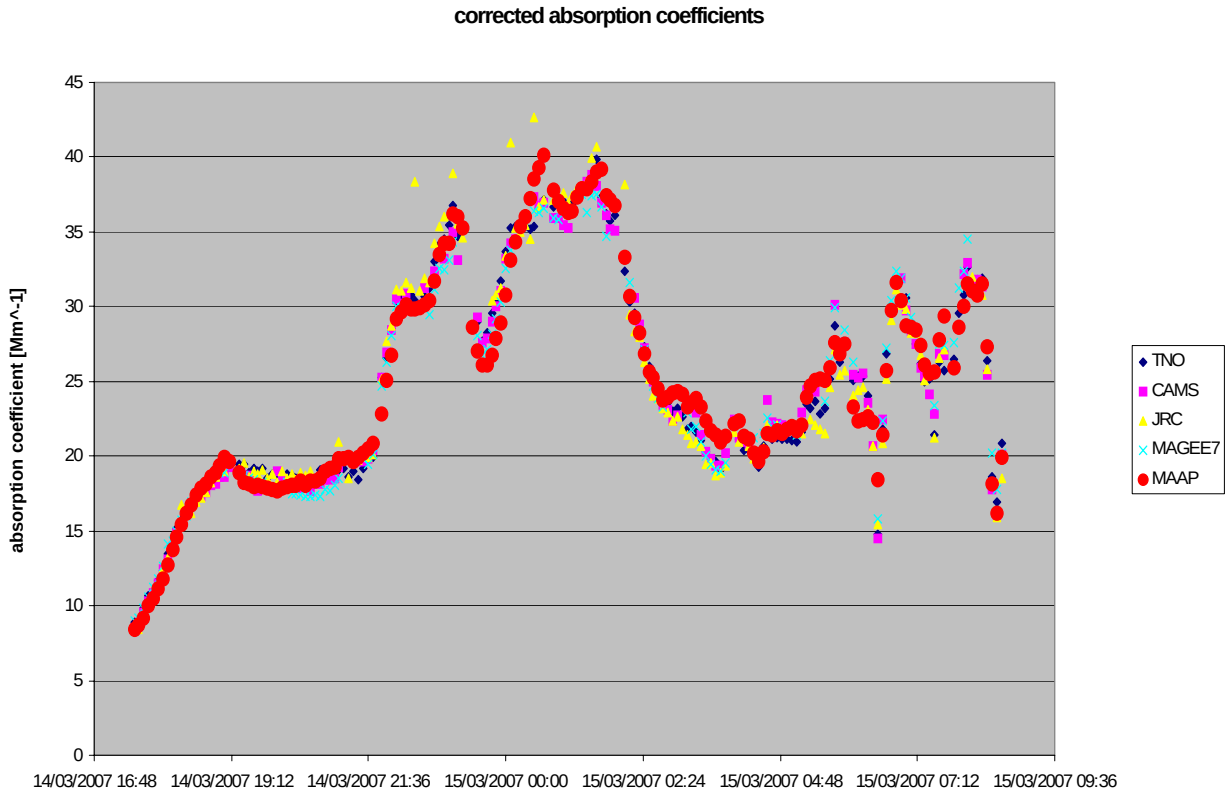

Figure 11: Corrected absorption coefficients of five Aethalometers.

5.5 Summary of instrument inter-comparisons

Table 4 and 5 summarizes the instrument results of the instrument characterization and the inter-comparisons.

Table 4: Summary of instrument characterizations

Instrument	**Noise** [1/Mm] temporal resolution		**Unit to unit variability**
PSAP	0.08	1 min	11%
Home made PSAP	0.3 (NILU) 1.7 (ITM)	10 min	— —
MAAP	0.05	1 min	3.1%
Aethalometer		2 min	~ 10 %
DMT-PAS	0.5	1 min	—
MPI-PAS	2	1 min	—
Absorption from extinction cell	up to 100%	—	—

Table 5: Summary of instrument inter-comparison. The inter-comparison of PSAP and MAAP in two experiments with ambient air we got different results.

Instrument	**slope**	**Intercept [1/Mm]**	**R^2**	**aerosol**	**corrections**
PSAP vs. MAAP	0.99 0.82	1.17 4.3	0.99 0.98	ambient	B1999 correction
home made PSAP vs. PSAP	1.10		0.97	ambient	B1999 correction
MPI-PAS vs. DMT-PAS	2.31 1.39	-0.85 2.36	0.90 0.96	ambient in-situ	λ=530 nm
MPI-PAS vs. MAAP	0.97	-5.72	0.93	ambient	λ=530 nm
Ext.-Sca. vs. MPI	0.99	-8.99	0.74	in-situ	λ=530 nm

5.6 Cross sensitivity to particle scattering

For investigation of the cross sensitivity to particle scattering ammonium sulfate aerosols were generated by an atomizer. The particles number size distribution was measured using particle size spectrometers (DMPS/APS) and the scattering coefficient was measured at three wavelengths. The Angstroem scattering exponent calculated from the spectral scattering coefficient is an indicator of the particle size, the smaller the particles the higher the value of the Angstroem exponent.

The cross sensitivity to particle scattering is defined as the ratio of the apparent interpreted absorption and the scattering coefficients. Figure 12 shows the cross sensitivity to particle scattering for MAAP versus the loading. The loading shown in Fig. 12 is the Optical depth of scattering particles accumulated on the filter.

Is can be seen that there is a cross sensitivity ranging from about 5% for low filter loadings down to 0.2% for high filter loadings. Form this measurement it is not clearly visible if there is an additional size effect. A possible size effect is covered by the loading effect.

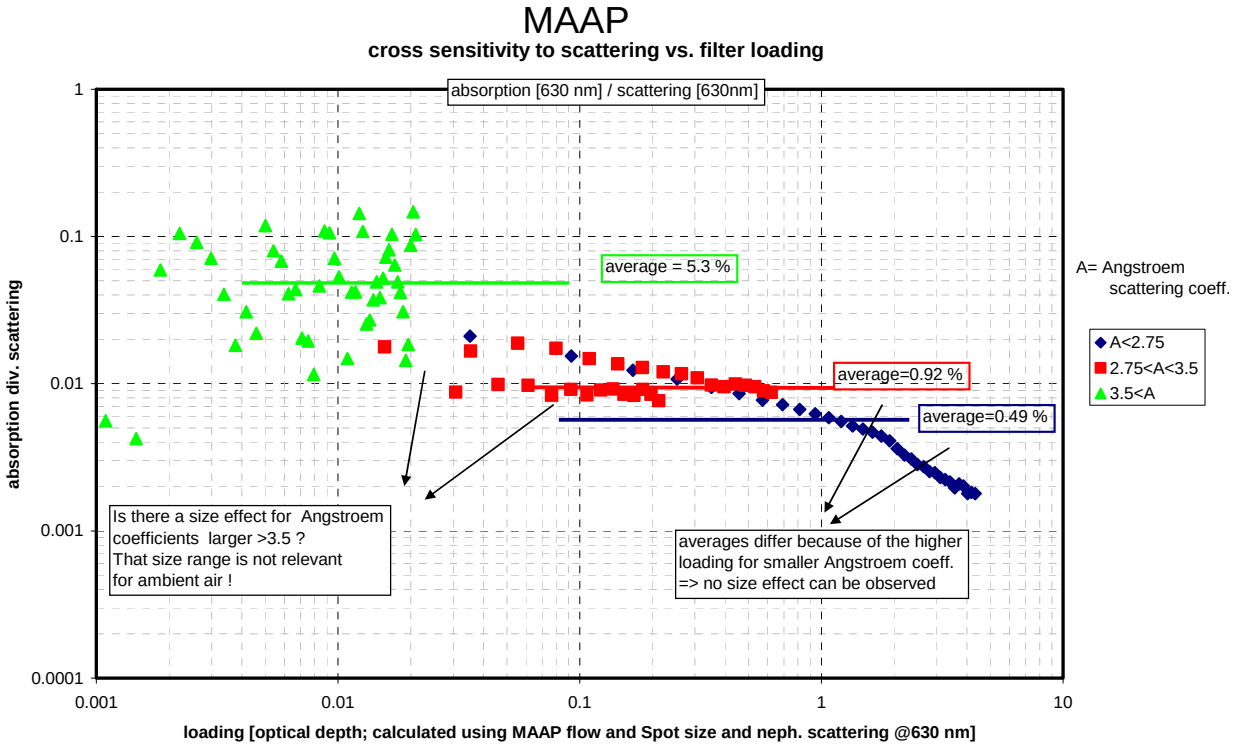

Figure 12: Cross sensitivity of MAAP to particle scattering.

A similar plot is shown for PSAP (see Figure 13). The loading is not plotted as an optical depth but as the optical transmission. For an unloaded filter the transmission is unity. When calculating the cross sensitivity, the PSAP prior was corrected using the Bond correction but without scattering term. The constant scattering correction factor introduced by Bond is plotted as green dotted line. Obviously the value derived by Bond is an average value for a certain range of loadings. Thus for low and high loadings the Bond correction is an underestimation and overestimation, respectively. A size effect is not clearly visible. In order to see a size effect the experiments have to be done with a better accuracy.

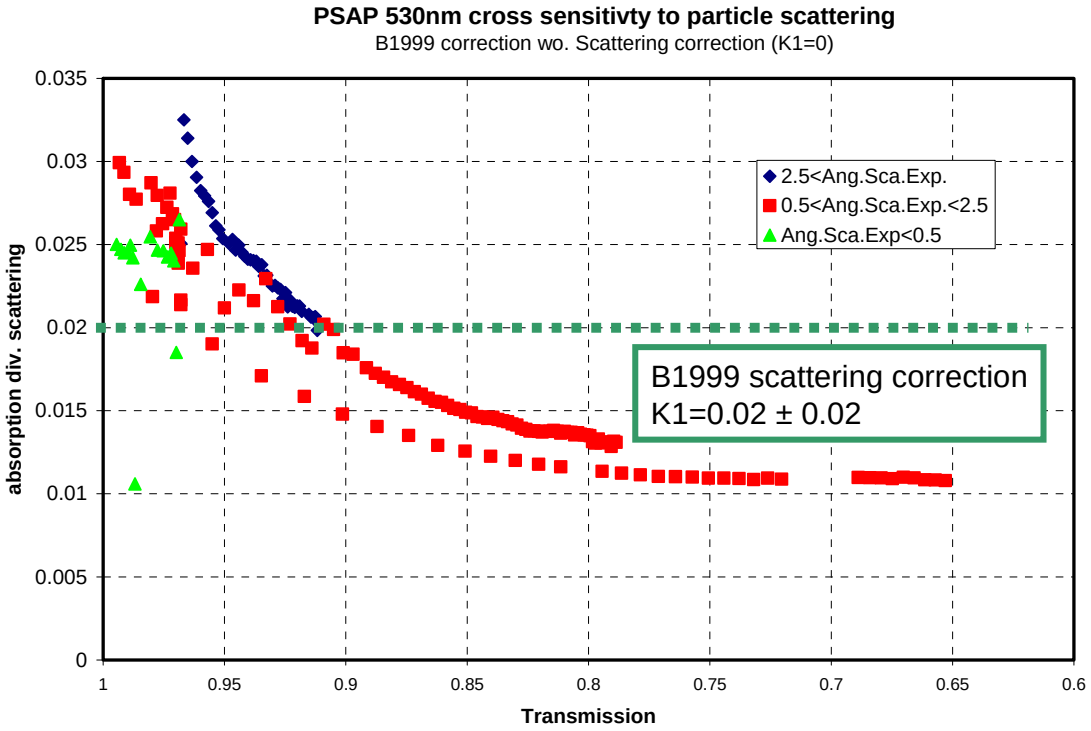

Figure 13: Cross sensitivity of PSAP to particle scattering.

6 Summary

The sensitivity to scattering particles is still a not well understood problem. The method introduced by Bond et al. (1999) is a first order correction. The dependence on the particle scattering was attributed to the total particle scattering (neglecting the truncation error of nephelometer). Till now a theoretical based model is missing which includes the particle backscattering in addition to the total particle scattering.

The results of the absorption photometer workshop show that there the need for better corrections function. The correction of scattering artifacts is one of the major goals for future work. Corrections function for absorbing and scattering aerosols can not be developed independently, because these functions are coupled. The data of the absorption photometer workshop of 2007 will be used for developing correction functions.

7 References

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