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# Inter-comparison of optical absorption coefficients of atmospheric aerosols determined by a multi-wavelength photoacoustic spectrometer and an Aethalometer under sub-urban wintry conditions

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## ABSTRACT

The wavelength-dependent optical absorption coefficients (OACs) of atmospheric aerosols were measured at a sub-urban site in Budapest under wintry conditions simultaneously with a commercially available, seven-wavelength ( $7\lambda$ ) Aethalometer instrument, and with a recently developed four-wavelength (1064, 532, 355 and 266 nm) photoacoustic spectrometer ( $4\lambda$ -PAS). While there is a good agreement between the readings on the two systems in the near-infrared and in the visible, the OACs measured by the  $4\lambda$ -PAS in the UV region were systematically higher than the OACs extrapolated to the UV region from data measured in the visible by the Aethalometer, and the relative deviation between the readings of the instruments in the UV was found to increase exponentially as a function of the Ångström exponent fitted to the wavelength dependency measured by either system. This result indicates that the  $4\lambda$ -PAS has more diagnostic value than the Aethalometer, i.e. it has higher sensitivity to discriminate between different aerosol sources. Furthermore, our finding can significantly affect the results of radiative transfer calculations for atmospheric aerosols, in which wavelength-dependent optical absorption, usually measured by Aethalometers, is an important input parameter.

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## 1. Introduction

The wavelength-dependent optical absorption coefficient (OAC) of the light absorbing components (LAC) in atmospheric aerosols is one of those parameters that seriously influence their direct radiative forcing, yet are known only with large uncertainties (Penner et al., 2001; Schwartz, 2004; Lack et al., 2006; Solomon et al., 2007). It is generally assumed that this wavelength dependency can be accurately described by the following relation:

$$b_{\text{abs},\lambda} = a \times \lambda^{-\alpha}, \quad (1)$$

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where  $b_{\text{abs},\lambda}$  is the wavelength-dependent light absorption coefficient (in unit of  $\text{Mm}^{-1}$ , where  $1 \text{ Mm}^{-1} = 10^{-8} \text{ cm}^{-1}$ ),  $a$  is a wavelength independent constant,  $\lambda$  is the wavelength (in unit of nm), and  $\alpha$  – the Ångström exponent – is an aerosol specific optical parameter (Ångström, 1929, 1930).

Until recently it was a generally accepted practice to measure the OAC at a single wavelength and assume a near-linear dependence of the OAC on the inverse of the wavelength (i.e. using an Ångström exponent close to one in Eq. (1)). This approach can indeed be justified in case of refractory black carbon (rBC) aerosol emitted from flaming combustion. However, the recent discovery of various additional components in LAC, such as the so called brown carbon, as well as different types of non-carbonaceous light absorbing particles make this approach less reliable (Andreae & Gelencsér, 2006; Hoffer et al., 2006). Ångström exponents markedly different from unity were observed both under laboratory and field conditions, and some measurements indicated that a single Ångström exponent can be used to describe the wavelength dependency of the OAC of LAC only in narrow wavelength regions (Moosmüller et al., 2011).

Three different measurement techniques are usually applied to determine the OACs of aerosols.

- The difference method (DM) is based on determining absorption from the difference between extinction and scattering measured by an extinction cell and a Nephelometer, respectively (Smith & Atkinson, 2001; Schnaiter et al., 2003, 2005; Pettersson et al., 2004). Since in this method absorption is determined from a small difference of two large quantities, its applicability is limited to high pollution events, or to laboratory studies on aerosols (primarily on rBC particles) in high concentrations (Bond et al., 1999).
- Filter based techniques, where the aerosol laden gas sample is drawn through a filter on which the particles are deposited. The increase of deposited aerosol mass with time results in a decrease of light transmission through the filter, which is measured continuously (Bond et al., 1999). The most frequently used instruments applying this technique are the Aethalometer (Hansen et al., 1984; Weingartner et al., 2003, Fialho et al., 2005), the particle soot absorption photometer (PSAP) (Bond et al., 1999) and the multi-angle absorption photometer (MAAP) (Petzold and Schönlinner, 2004). On-line aerosol absorption measurements are most frequently carried out with this technique. However, as the measurements are not performed on airborne aerosols, but rather on samples deposited on a filter, these measurements suffer from a variety of artefacts originating from the photon-filter and photon-aerosol interactions, including multiple scattering and shadowing effects. Efforts have been made to compensate these artefacts, and site-specific correction factors have been introduced, which have proved to improve the reliability of the method considerably. However, up to now there is no correction scheme, which is generally valid for atmospheric aerosols having largely varying chemical and physical properties (Bond et al., 1999; Arnott et al., 2005; Virkkula et al., 2005; Schmid et al., 2006; Collaud Coen et al., 2010).
- The photoacoustic (PA) method is based on determining light absorption from the amplitude of the sound wave generated by the absorption of modulated light (Rosencwaig, 1980). One of the most important advantages of this method is that the PA signal is linearly proportional to light absorption, and is completely insensitive to light scattering. Furthermore, contrary to the other techniques, a PA system is suitable for measuring the absorption of airborne aerosols directly, based on calibration with a light absorbing gaseous analyte, without the need for site-specific correction factors (Roessler & Faxvog, 1980; Sigrist, 1994). The stability and the reliability of recently developed PA systems make them useable as reference instruments in measurements for the optical absorption coefficient of artificial or atmospheric aerosols, although due to sensitivity limitations (minimum detectable concentration with the current PA systems is approx.  $1 \mu\text{g}/\text{m}^3$  of rBC) only at polluted urban sites (Arnott et al., 1999; Krämer et al., 2001).

Some advanced versions of the above listed instruments also offer the possibility to determine the OAC in an extended wavelength range such as the seven-wavelength Aethalometer operating from 370 to 950 nm, or the two-wavelength PAS operating at 405 nm and 870 nm (Lewis et al., 2008). To the best of the authors' knowledge, the widest wavelength coverage is achieved with our recently developed four-wavelength PAS ( $4\lambda$ -PAS) operating at 1064, 532, 355 and 266 nm (Ajtai et al., 2010).

From the wavelengths at which the LAC absorbs solar radiation, our knowledge on the OAC in the UV region is the most limited. Although solar radiation below  $\sim 300$  nm is totally absorbed by oxygen and ozone at high altitudes, and does not contribute to aerosol radiative effects, absorption measurements in the UV are expected to have high diagnostic value for aerosol source apportionment. Yet the OACs in the UV are typically determined by the extrapolation from measurements in the visible using various assumptions, since reliable measured data are not available mostly because of the lack of appropriate instrumentation.

This paper presents the results of OAC measurements on atmospheric aerosols conducted in a sub-urban area (Budapest, Hungary) using a conventional  $7\lambda$ -Aethalometer and the  $4\lambda$ -PAS under wintry conditions. The OACs determined by the two instruments were inter-compared in the overlapped wavelength region and out of the common wavelength range, especially focusing the UV region where the measured ( $4\lambda$ -PAS) and extrapolated ( $7\lambda$ -Aethalometer) results were compared.

## 2. Experimental

The results presented here were obtained during a field measurement campaign between 10 and 16 March 2010. The instruments for measuring the OAC were operated inside a temperature and humidity controlled mobile measurement

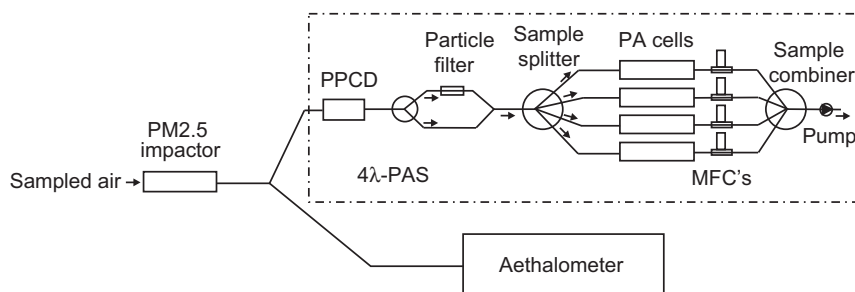


Fig. 1. Experimental scheme showing the gas handling of the  $4\lambda$ -PAS instrument (within the dotted-line box) in details. (See text for further details.)

station, installed in a three-ton van (courtesy of Hilase Ltd.) at the observation site of the Hungarian Meteorological Service (Gilice tér Budapest, GPS position:  $47^{\circ}25'54.18''\text{N}$ ,  $19^{\circ}11'1.22''\text{E}$ ). The daily average air temperature during the campaign gradually increased from  $-4$  to  $4$  °C and showed moderate daily variation with no precipitation throughout the period.

The OACs were measured simultaneously with a  $7\lambda$ -Aethalometer (Magee Scientific) at 370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm and 950 nm, and with the  $4\lambda$ -PAS (courtesy of Hilase Ltd.) at 266 nm, 355 nm, 532 nm and 1064 nm. The experimental set-up is shown schematically in Fig. 1. Both instruments were connected to the same  $\text{PM}_{2.5}$  cut-off inlet. The Aethalometer was operated at a flow rate of 5 l/min in an automated mode (i.e. the filter tape advanced whenever light transmission at 370 nm decreased to 75% of the value measured on an aerosol free filter), while the  $4\lambda$ -PAS was operated at a flow rate of 1 l/min. A measurement cycle with the  $4\lambda$ -PAS lasted for 20 min, and in order to make the readings of the two instruments better comparable, the 2 min long measurement of the Aethalometer was averaged to this value. Further details of the two instruments are given in the following section.

### 2.1. The $4\lambda$ -PAS instrument

When modulated light is absorbed by a gas sample, the energy is released as heat generating pressure waves at the modulation frequency of the exciting light. These waves are amplified in an acoustic resonator (placed within the so called photoacoustic cell) when the laser modulation frequency is set to coincide with one of the resonance frequencies of the acoustic resonator. The amplified pressure wave can be detected by a microphone attached to the resonator. The PA response is proportional to the concentration as well as to the absorption coefficient of the absorbing species. More detailed description of the photoacoustic method can be found elsewhere (Miklós et al., 2001; Moosmüller et al., 2009; Bozóki et al., 2011).

The  $4\lambda$ -PAS applied here is an upgraded version of the instrument used in a previous aerosol chamber experiment (Ajtai et al., 2010). Briefly, it has four detection cells, each of which is illuminated with one of the light beams; thereby it measures the OAC at four different wavelengths simultaneously. The instrument is controlled by its built-in programmable electronic unit, which makes its operation fully automatic. From the signals measured in the detection cells, the electronic unit calculates the optical absorption coefficients of the aerosol within the gas sample with the help of calibration factors determined during an independent gas phase calibration of the instrument (Ajtai et al., 2010).

Three major upgrades have been made to the previously described instrument, which was originally built for laboratory applications. First of all, it has been made portable by building it into a rugged metal frame with dimensions of  $65\text{ cm} \times 65\text{ cm} \times 130\text{ cm}$ . The PAS now weighs approximately 100 kg. Second, the optical set-up was rearranged in such a way that the optical power in each detection cell was increased and consequently the minimum detectable optical absorption coefficient was improved to 0.17, 0.7, 3.5,  $6.7\text{ Mm}^{-1}$  @ 1064, 532, 355 and 266 nm, respectively. Finally, the gas handling system of the instrument has also been upgraded to suppress possible interference effects from gaseous molecules, which is more likely to happen in field operation. The aerosol filter that was already built into the previous version of the system can be used to eliminate the effect of  $\text{NO}_2$  absorption at 532 nm. However, for highly reactive gases, such as ozone, the filter cannot be used for signal suppression; therefore, the gas handling of the instrument is supplemented with a Parallel Plate Carbon Denuder (PPCD), right after its sampling inlet. The PPCD is designed to remove ozone, which has strong absorption at 266 nm and could thus interfere with the OAC measurements on LAC in the UV. The transmission efficiency of the PPCD is practically 100% for aerosols, but it completely eliminates ozone, as it was verified in a laboratory study in which  $\text{O}_3$  was generated by an ozone generator (MLU400) in a concentration range covering the atmospheric ozone concentration by a large margin, and was fed to the PPCD. The  $\text{O}_3$  concentration was measured with a high sensitivity ozone detector (MLU700) after the PPCD, and it was found to be below the detection limit of the MLU700 in all cases.

### 2.2. The $7\lambda$ -Aethalometer instrument

In an Aethalometer, in which sampled air flows with a volumetric flow rate of  $F$ , and particles are deposited on a filter having an area of  $A$ , the sample volume ( $\Delta t \times F$ ) can be visualized as a cylinder with a basal area  $A$  and a length  $l = \Delta t \times F/A$ .

The optical attenuation is determined by a simple optical transmission measurement through the particle laden filter on each of the seven wavelengths generated by diode lasers. The optical power transmitted through the blank filter ( $I_0$ ) and the optical power transmitted through the particle laden filter ( $I$ ) are connected through Beer's law

$$I = I_0 e^{-(b_{\text{aeth},\lambda,t})l} = I_0 e^{-(b_{\text{aeth},\lambda,t})(\Delta t F/A)}. \quad (2)$$

The optical absorption coefficient  $b_{\text{aeth},\lambda,t}$  measured by the Aethalometer is given by

$$b_{\text{aeth},\lambda,t} = \frac{A}{\Delta t F} \ln\left(\frac{I_0}{I}\right). \quad (3)$$

As the instability of the light sources or the electronics could affect the reliability of the Aethalometer instrument, two light detectors are used in the instrument. One measures the intensity of light passing through the aerosol-laden filter spot, and the other measures the same quantity on the blank part of the filter. Eq. (3) assumes that: (i) the absorption of the aerosol does not change when being collected in a thin layer, (ii) interaction between the filter and aerosol optics can be neglected, and (iii) particle morphology does not change upon deposition, or the change does not affect the particle optics.

Due to the applied methodology (filter sampling), the OACs obtained by this instrument are affected by various sampling and analytical artefacts mostly referred to as multiple scattering and shadowing effects. To provide a better estimate on the OAC, the measured data need correction. There are several different correction schemes described in the literature. The correction procedure introduced by Weingartner et al. (2003) includes the correction factors  $C$  and  $R(ATN)$  as

$$b_{\text{abs},\lambda} = \frac{b_{\text{aeth},\lambda,t}}{C \times R(ATN)_{\lambda,t}}. \quad (4)$$

In Eq. (4),  $b_{\text{abs},\lambda,t}$  corresponds to the OAC of the measured aerosol sample after performing correction on the raw data of the Aethalometer. The correction factor  $C$  stands for multiple scattering, while  $R(ATN)_{\lambda,t}$  takes into account the shadowing effect based on the gradual accumulation of the aerosols in the filter.

For the sub-urban site of our measurement the value of  $C=2.14$  and the following expression for  $R(ATN)_{\lambda,t}$  are used (Weingartner et al., 2003; Sandradewi et al., 2008a, b)

$$R(ATN)_{\lambda,t} = \left(\frac{1}{f_\lambda} - 1\right) \times \frac{\ln(ATN_{\lambda,t}) - \ln(10\%)}{\ln(50\%) - \ln(10\%)} + 1, \quad (5)$$

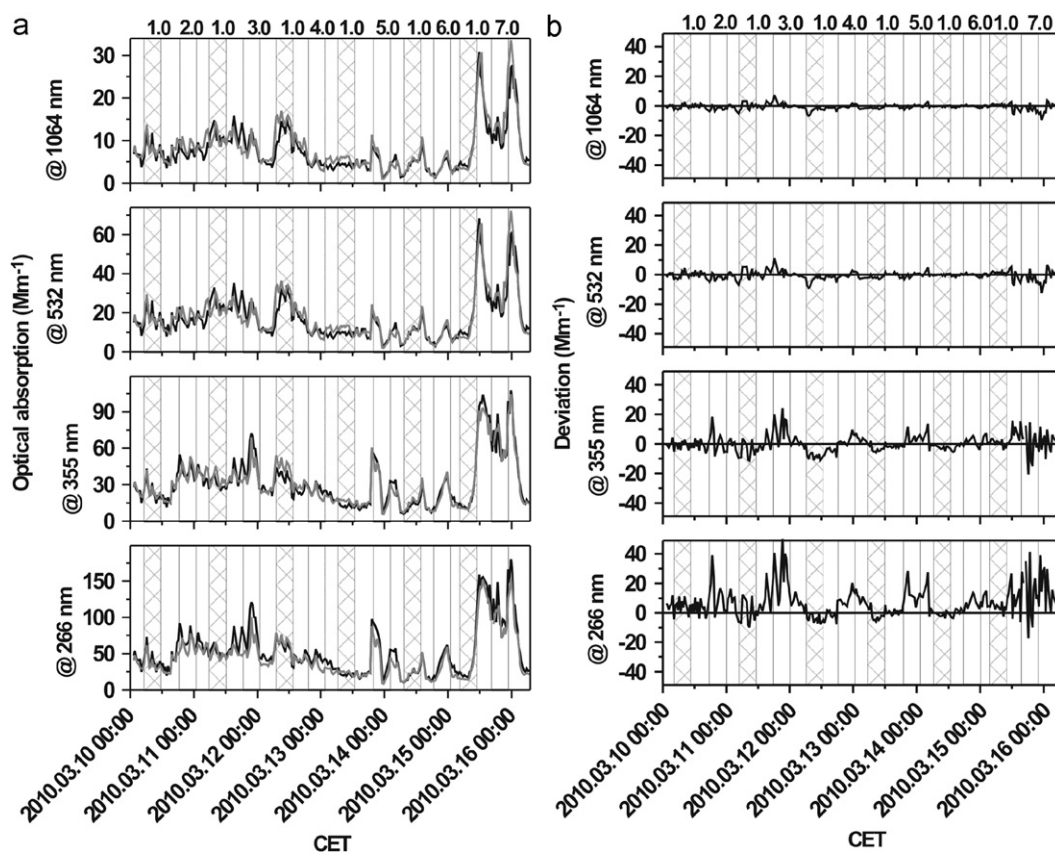
where  $ATN_{\lambda,t}$  represents the time and wavelength-dependent light attenuation measured by the Aethalometer, and  $f_\lambda$  is a free parameter that relates to the slope of this linear function. Due to the lack of simultaneous measurements of the aerosol scattering coefficients in this study,  $f_\lambda$  could not be determined as a function of the single scattering albedo. Alternatively,  $f_\lambda$  values were determined by the differences between the ratio of the absorption coefficient before and after each filter tape advancement step. The cumulative uncertainty of the absorption coefficient determined in this way is below 20% for any of the wavelengths.

### 3. Results and discussion

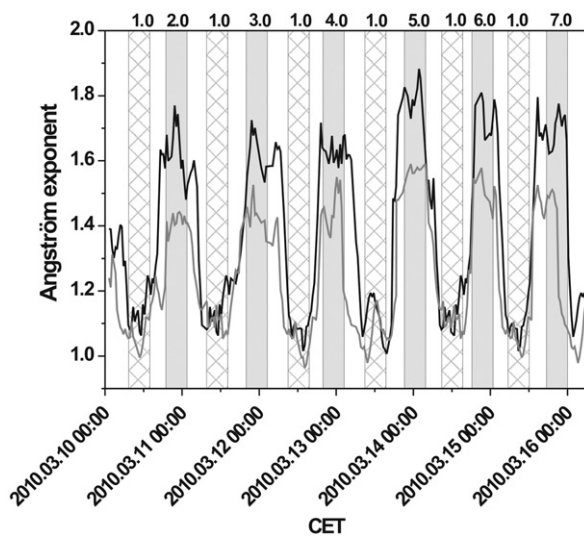
In Fig. 2 the optical absorption coefficients measured by the  $4\lambda$ -PAS and the  $7\lambda$ -Aethalometer instruments (Fig. 2a), as well as the difference between the readings of the two instruments (Fig. 2b) are shown. Since the two instruments operate at different wavelengths, their measurement results were made directly comparable by inter- and extrapolating the data of the  $7\lambda$ -Aethalometer to the measurement wavelengths of the  $4\lambda$ -PAS using Ångström exponents numerically fitted to the Aethalometer readings. For the 1064 nm only the longer wavelengths (i.e. 520–950 nm), while for the 355 and 266 nm only the shorter wavelengths (i.e. 370–520 nm) of the Aethalometer were used in the numerical fitting.

It can be seen in Fig. 2 that in general the measurement results of the two instruments are strongly correlated. The relative deviation between the readings of the two instruments averaged over the measurement period is less than  $\pm 10\%$  for all wavelengths, which can be considered to be a good agreement taking into account that in a study in which the performances of Aethalometers were compared a disagreement up to 40% between their readings was reported (Reid et al., 1998; Slowik et al., 2007). On the other hand, for the 266 nm in Fig. 2b it can be clearly seen that the difference between the readings becomes repeatedly positive over intervals lasting for several hours, while for other intervals the difference is close to zero.

In order to better understand the reason for the periodic deviation between the readings of the two systems, Ångström exponents were fitted to each measured wavelength dependency of the OAC for the Aethalometer and the PAS separately. (Note that in this case all available wavelengths of each instrument were used in the numerical fittings.) The result can be seen in Fig. 3. A characteristic diurnal variation of the Ångström exponents can be observed. In case of Ångström exponents close to one the numerical fittings for both systems give similar Ångström exponents; however when the Ångström exponent is markedly higher than one, the PAS clearly indicates larger variations in the Ångström exponent. It has to be noted that the measurement of the OAC of LAC presented here was also performed at the same location under summer



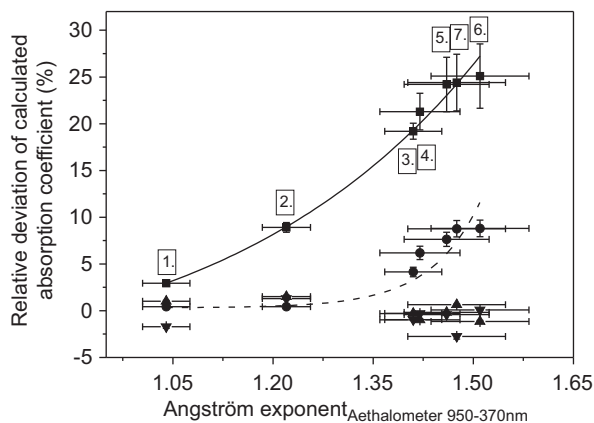
**Fig. 2.** The optical absorption coefficients measured by the  $4\lambda$ -PAS (black line) and inter/extrapolated from the readings of the  $7\lambda$ -Aethalometer (gray line) instruments (a), and the calculated differences between the readings of the two instruments (b).



**Fig. 3.** The temporal variation of the Ångström exponent determined from the measurements of the  $4\lambda$ -PAS (black line) and the  $7\lambda$ -Aethalometer (gray line) instruments.

weather conditions, and the Ångström exponent was found to be constant (about 0.84) for the entire wavelength coverage of the systems and independent of the time of day.

Next the measurement campaigns were divided into shorter (6 hour long) periods. The division is based on Fig. 3, i.e. intervals were selected in which the measured Ångström exponents were relatively constant. The selected intervals are



**Fig. 4.** The average relative deviations between the OAC readings of the two systems at the four wavelengths of PAS as a function of the average Ångström exponents fitted for the entire wavelength coverage of the Aethalometer.

**Table 1**

Ångström exponent calculated from the measured data in winter day-time and night-time periods in case of 4λ-PAS and the 7λ-Aethalometer, respectively.

|                         | Instrument |            |             |                 |             |
|-------------------------|------------|------------|-------------|-----------------|-------------|
|                         | 4λ-PAS     |            |             | 7λ-Aethalometer |             |
| Fitted wavelength range | 266–355 nm | 355–532 nm | 532–1064 nm | 370–520 nm      | 520–950 nm  |
| Interval 1              | 1.76       | 1.09       | 1.08        | 1.06 ± 0.15     | 1.03 ± 0.1  |
| Interval 2–7 (averaged) | 2.1        | 1.37       | 1.14        | 1.4 ± 0.2       | 1.17 ± 0.14 |

marked by the shaded areas in Figs. 2 and 3, and with numbers, too. Note that the intervals with the lowest Ångström exponent are uniformly numbered as interval 1.

For the selected intervals the average Ångström exponents for the entire wavelength coverage of the Aethalometer and the average deviations between the OAC readings of the two systems at the four wavelengths of PAS are calculated and plotted in one graph as shown in Fig. 4. (All intervals marked with 1 are averaged into a single value.) An exponential dependence of the deviation on the Ångström exponent was observed for 266 nm, a weaker one for 355 nm, while at 532 nm and 1064 nm the deviation seems negligible. Similar exponential dependences were found when the average deviations between the OAC readings of the two systems at the four wavelengths of PAS were plotted against either the Ångström exponent fitted to the shorter wavelength portion of the Aethalometer (i.e. 370–520 nm) or to any wavelengths of the 4λ-PAS. (Note: in another measurement under summery conditions, the Ångström exponents were found to be around 0.9 for all wavelength ranges and for both instruments. Though the measurement points corresponding to this measurement are not shown in Fig. 4, they are actually in good agreement with the presented results as for this low Ångström exponent no deviation between the calculated absorption coefficients was found.)

In Table 1 the Ångström exponents calculated for various wavelength portions of the systems and for the different intervals (with the results for intervals 2–7 are averaged together) are listed. It can be seen that for the longer wavelengths the calculated Ångström exponents agree well, but deviate considerably from each other in the shorter wavelength range.

#### 4. Conclusions and outlook

OAC measurements of LAC at a sub-urban measurement site were performed under wintry conditions with a four-wavelength photoacoustic spectrometer (4λ-PAS) and with a seven-wavelength Aethalometer. A strong diurnal variation in the Ångström exponents fitted to the optical absorption coefficients measured either by a 4λ-PAS or a seven-wavelength Aethalometer at a sub-urban measurement site under wintry conditions was observed. This finding indicates that the composition of atmospheric aerosols varied throughout the day, and was most probably dominated by traffic aerosols during daytime, and by organic carbonaceous aerosols originating from biomass burning during evening and night (Sandradewi et al., 2008a, b; Favez et al., 2009).

OACs measured by the Aethalometer was found to agree well with the readings of the 4λ-PAS in the visible, but in certain cases the measured (4λ-PAS) and the extrapolated (7λ-Aethalometer) OACs deviated considerably in the UV range. An exponential dependence between the Ångström exponent fitted to the wavelengths of the Aethalometer and the above mentioned deviation were found, which can be used to correct the readings of the Aethalometer extrapolated into the UV

during the presented measurements. We plan to examine in detail whether this dependence is generally valid for the present measurement site, and to verify whether this dependence is valid for other sub-urban sites as well.

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