Photoacoustic and filter measurements related to aerosol light absorption during the Northern Front Range Air Quality Study (Colorado 1996/1997)

H. Moosmüller, W. P. Arnott, C. F. Rogers, J. C. Chow, and C. A. Frazier
Desert Research Institute, University of Nevada System
P.O. Box 60220, Reno, NV 89506

L. E. Sherman and D. L. Dietrich
Air Resource Specialists
1901 Sharp Point Drive, Suite E, Fort Collins, CO 80525

Abstract

A new photoacoustic instrument for the measurement of aerosol light absorption was collocated with conventional aerosol instrumentation during the 1996-1997 winter intensive of the Northern Front Range Air Quality Study. Measurements of the light absorption efficiency for black carbon were 5 m^2/g at 685 nm and 10 m^2/g at 532 nm, and for elemental carbon, 3.6 m^2/g at 685 nm. We show that these values together with previous photoacoustic measurements of aerosol light absorption elucidate the wavelength dependence of absorption efficiency for carbonaceous aerosol in the visible and near-visible region. Integrating plate type filter measurements of aerosol light absorption result in far larger values than those measured with the photoacoustic instrument. We demonstrate that a very recently published correction technique [*Horvath*, 199X] can yield improved agreement.

Introduction

Aerosol has a direct influence on the radiation balance by scattering and absorbing light. Aerosol light absorption is due mostly to black carbon (BC) particles resulting from the incomplete combustion of fossil and biomass fuels [e.g., *Charlson and Ogren*, 1982; *Japar et al.*, 1986; *Liousse et al.*, 1993]. BC particles are generally smaller than one micrometer in diameter. In addition, airborne soil particles can include light absorbing compounds such as iron oxides, which in some geographical regions contribute significantly to aerosol light absorption [e.g., *Foot and Kilsby*, 1989; *Huffman*, 1996; *Pinnick et al.*, 1993]. While the global contribution of aerosol light absorption to aerosol extinction has been estimated to be on the order of a few percent [*Tegen et al.*, 1997], the regional contribution in polluted areas can reach ten to thirty percent [*Adams et al.*, 1990; *Turpin et al.*, 1990; *Watson et al.*, 1988]. Even in cases where the contribution of aerosol light absorption to total aerosol light extinction is relatively small, it may still be important. While scattering spatially redistributes radiative energy, absorption converts radiative energy to thermal energy. Light absorption and scattering by aerosols have a very different influence on both radiative heating [*Charlock and Sellers*, 1980] and photochemistry [*Dickerson et al.*, 1997].

Routine measurement methods for ambient aerosol light absorption such as the integrating plate type methods or calculation of aerosol light absorption from elemental carbon concentrations often have a questionable absolute calibration. The Northern Front Range Air Quality Study (NFRAQS) offered the opportunity to compare these routine measurements not only with each other but also with the results of a more direct measurement of aerosol light absorption by the novel and highly sensitive Desert Research Institute (DRI) photoacoustic instrument. This instrument was operated during the winter intensive of NFRAQS at the “core” measurement site at Brighton, CO (39˚59’04”N, 104˚46’01”W, 1570 m Elevation), collocated (distance between sampling inlets about 10 m) with a number of other aerosol measurements including an aethalometer, nephelometers, and filter sampling for aerosol light absorption, elemental/organic carbon speciation, and elemental analysis. The Brighton, CO site is located in a semi-rural environment about 30 km northeast of downtown Denver, CO. This paper presents a comparison of photoacoustic measurements of aerosol light absorption with other, more routine measurements taken at the Brighton, CO site, and reports the wavelength dependency of aerosol light absorption from these and previous measurements.

Instrumentation

I. Filter Light Absorption: Aethalometer and Integrating Plate

Ambient aerosol light absorption is commonly determined by measuring the optical attenuation through a filter on which aerosol particles have been accumulated. This measurement can take place after sampling has been terminated (e.g., integrating plate technique [*Lin et al.*, 1973]), or in real time during the sampling process (i.e., aethalometer [*Hansen and Novakov*, 1990; *Hansen et al.*, 1984]). These methods are perhaps questionable due to the sampling process on the filter; the physical shape of particles (e.g., fractal chain aggregates [*Katrinak et al.*, 1993]) may be changed and the interaction of scattering and absorption by densely aggregated aerosols and the filter/plate system are not properly described by Beer's law. In addition, filter methods do not properly account for the influence of humidity on aerosol light absorption, as, even for real time methods such as the aethalometer, the humidity dependence of aerosol light absorption is likely to be modified by the filter substrate [*National Research Council*, 1996]. As a consequence, integrating plate type measurements require use of an empirical calibration factor.

At the NFRAQS Brighton site a commercial version of the aethalometer (Magee Scientific Mfg. Co. Aethalometer AE-10IM) was operated. This instrument measures light attenuation due to aerosol particles accumulated on a quartz filter. This attenuation is reported as mass density of BC using an attenuation efficiency of 19 m^2/g. A comparison between aethalometer BC and thermal EC (elemental carbon) measurements reports excellent agreement with a regression slope of 1.02 ± 0.04 and correlation coefficient $R^2 = 0.973$ [*Hansen and McMurry*, 1990]. However, the reverse process of converting BC mass density, as measured by an aethalometer, to atmospheric light absorption typically uses an absorption efficiency of 10 m^2/g for BC aerosol [e.g., *Bodhaine*, 1995].

Also available from the Brighton site are results from an integrating plate type measurement performed on fibrous PTFE membrane filters, on which aerosol particles were accumulated for six or twelve hour periods. Light attenuation through these filters is measured before and after sampling with a Tobias TBX-10 Densitometer. This instrument operates with “white” light, however, the effective spectral weighting (i.e., convolution of source and detector spectrum) is not known to us. Light attenuation due to aerosol particles accumulated on these filters is reported as an absorption coefficient with dimension of inverse distance without any calibration factor to compensate for deficiencies of the integrating plate method.

II. DRI Thermal/Optical Reflectance Carbon Analysis System (TOR)

Elemental carbon (EC) is defined by its thermally refractory nature and this property is used in a variety of thermal carbon analysis methods. During NFRAQS aerosol particles were accumulated for six or twelve hour periods on quartz-fiber filters which were subsequently analyzed using the DRI TOR method [*Chow et al.*, 1993]. While the operational definitions of EC and BC are completely different, i.e., based on refractory and light absorption properties, respectively, it is commonly assumed that EC and BC are identical [e.g., *Liousse et al.*, 1993]. The assumption is that EC may be considered to be the only aerosol component that strongly absorbs light.

III. DRI Photoacoustic Instrument

The disadvantages of the integrating plate type methods can be eliminated by measuring the aerosol light absorption *in situ*, i.e., with the particles suspended in their ambient state. This has been done by using photoacoustic detection [e.g., *Adams*, 1988; *Foot*, 1979; *Japar and Szkarlat*, 1981; *Roessler*, 1984; *Terhune and Anderson*, 1977; *Truex and Anderson*, 1979], where light from an intensity modulated source is absorbed by aerosol, resulting in periodic heating and subsequent expansion of the surrounding air. This expansion results in a sound wave at the modulation frequency which can be sensitively detected with a microphone [e.g., *Pao*, 1977].

However, practical use of this method was limited due to relatively low sensitivity and the use of inefficient light sources. The recent development of highly efficient all-solid-state lasers has led to the development of more compact and practical systems [*Arnott et al.*, submitted; *Petzold and Niessner*, 1996], and better acoustic resonator design has improved instrument sensitivity to about 0.5 Mm^{-1} for the DRI photoacoustic instrument [*Arnott et al.*, submitted]. The DRI photoacoustic instrument was operated at the Brighton site. During part of the study a diode-pumped frequency doubled Nd:YAG laser operating in the green at 532 nm was used as light source, while during other parts an AlGaInP diode laser operating in the red at 685 nm was utilized. At both wavelengths, the contribution of gaseous light absorption can be neglected for atmospheric measurements [*Arnott et al.*, submitted].

NFRAQS Data

An initial question regarding the use of real time instrumentation for the measurement of aerosol light absorption such as the photoacoustic instrument or the aethalometer is: does the real time (10 min time resolution) measurement of aerosol light absorption produce any new information or are real time nephelometer measurements in combination with the batch analysis of filter samples sufficient? This question is equivalent to asking if the aerosol single scattering albedo changes significantly on time scales below a few hours typical for filter samples. As an example, aerosol light absorption coefficients (B_{abs}) measured with the green laser are shown together with aerosol scattering coefficients measured with a nephelometer (Optec NGN-2) in Fig. 1 for December 18, 1996, with gaseous scattering subtracted out. Note that some, but not all peaks in aerosol light absorption occur simultaneously with peaks in light scattering and *vice versa*. This behavior corresponds to an aerosol single scattering albedo which can change significantly on a 10-min time scale, as shown in Fig. 2 for December 18.

Comparison of Photoacoustic Measurements with Analysis of Filter Samples

Three daily filter sampling periods (06:00 - 12:00, 12:00 - 18:00, and 18:00 - 06:00) were used during NFRAQS. For each period a quartz filter was analyzed by the DRI TOR method and a fibrous PTFE membrane filter with an integrating plate type method. Results include EC concentration and particle light absorption B_{abs} for each sampling period. Measurements of aerosol light absorption with the DRI photoacoustic instrument were averaged over each sampling period for comparison purposes. Results of linear regression analysis are summarized in Table 1 and will be discussed below.

The relation between DRI TOR measurements of EC and those of aerosol light absorption B_{abs} with the DRI photoacoustic instrument operating at 685 nm is shown in Fig. 3 for January 3, and 6 through 9, 1997. Linear regression indicates relatively good correlation between EC and photoacoustic B_{abs} with a correlation coefficient of $R^2 = 0.85$, a slope of 3.58 m^2/g and a zero-offset of 0.17 Mm^{-1}. The slope corresponds to the aerosol absorption efficiency.

Figure 4 shows the relation between photoacoustic measurement of aerosol light absorption at 685 nm and integrating plate type measurements of filter B_{abs}. Linear regression indicates fair correlation between photoacoustic and filter B_{abs} with a correlation coefficient of $R^2 = 0.72$, a slope of 3.00 and a zero-offset of 3.43. Note that the correlation would be substantially improved (to 0.86) if a single filter measurement (i.e., $B_{abs} = 0.0$) would be excluded. Both slope and zero-offset are quite large.

A very recent publication by Horvath indicates that integrating plate type measurements always give too high a value for the particle light absorption coefficient B_{abs}, based on theoretical arguments and corroborated by experimental measurements [*Horvath*, 199X]. Horvath also gives a simple correction table for this kind of measurement where the correction factor is based on the particle albedo. We have applied these correction factors to the filter data shown in Fig. 4, with particle albedo calculated from photoacoustic and nephelometer measurements. No inter- or extrapolation of Horvath's table values took place, we just chose the closest value included in the table. The correlation between the corrected filter measurements of B_{abs}, and the photoacoustic measurement of aerosol light absorption at 685 nm is shown in Fig. 5. Linear regression indicates relatively good correlation between photoacoustic and corrected filter B_{abs} with a correlation coefficient of $R^2 = 0.83$, a slope of 1.68 and a zero-offset of -0.38. Besides the increase in correlation coefficient (from 0.72 to 0.83), the correction has also reduced the zero-offset substantially (from 3.43 to -0.38).

As the photoacoustic instrument was operated for only two days at 532 nm and for these two days some photoacoustic and some filter data were missing, only three sampling periods had both photoacoustic and filter data. Due to this lack of data, the correlation between 532-nm photoacoustic data and filter data was not investigated.

Comparison of Photoacoustic and Aethalometer Measurements

Simultaneous measurements with the DRI photoacoustic instrument and the aethalometer were made for several days in December 1996 and January 1997. During December 17 and 18, 1996, the photoacoustic instrument was operated with a green laser as light source emitting at 532 nm. The correlation between aethalometer and photoacoustic measurements is shown in Fig. 6. Linear regression analysis indicates good correlation with an absorption efficiency for BC of 10 m^2/g, a zero-offset of 0.87 Mm^{-1} and a correlation coefficient of $R^2 = 0.87$. Figure 7 shows a comparison between the time evolution of photoacoustic B_{abs} and aethalometer BC. The two different y-axes have been scaled based on the absorption efficiency of about 10 m^2/g obtained from the regression analysis. This time series documents generally good agreement between the two instruments, although at certain times results can be different up to a factor of two. Disagreements are more frequent during short spikes which often encompass only a single data point. This could be due to spatial inhomogeneities in aerosol properties (the sampling inlets are about 12 m apart), temporal changes in aerosol absorption efficiency, and/or inaccurate measurements by one or both of the instruments. Results for the regression analysis for the two individual days are given in Table 1.

Correlations between the DRI photoacoustic instrument operating at 685 nm and the aethalometer were analyzed for January 3 and 6 through 9, 1997, a total of five days. As examples, the correlation for January 7, 1997 is shown in Fig. 8, the time series for January 6 through 9, 1997 in Fig. 9, and Table 1 summarizes the results of regression analysis for the five individual days. The results for the photoacoustic instrument operating at 685 nm are quite similar to those at 532 nm with good correlations (R^2 around 0.9) and good agreement between the two time series, with the exception of the absorption efficiency (i.e., slope). For the photoacoustic measurement at 685 nm the average absorption efficiency was about 5 m^2/g, a factor of two smaller than the value determined during the two days of operation at 532 nm (i.e., 10 m^2/g).

Discussion

Comparisons of DRI photoacoustic measurements of aerosol light absorption and aethalometer measurements of BC concentrations have resulted in an absorption efficiency for BC of about 5 m^2/g at 685 nm and about 10 m^2/g at 532 nm. In addition, the absorption efficiency for EC has been determined to be about 3.6 m^2/g at 685 nm, somewhat smaller than the BC efficiency at the same wavelength. Recent reviews of aerosol light absorption have summarized values for the absorption efficiency ranging from 2 m^2/g to 17 m^2/g [*Horvath*, 1993; *Liousse et al.*, 1993]. This large range has been attributed mainly to the diversity of methods used for measuring aerosol light absorption and the variability of the aerosols encountered. One can obtain a somewhat more clear picture by including only measurements of aerosol light absorption by the photoacoustic method and by considering the wavelength at which the photoacoustic instruments operate. The results of a limited survey for the visible and near-visible spectral region is shown in Fig. 10. At each wavelength the aerosol absorption efficiencies for different aerosols are quite close to each other. The absorption efficiency decreases toward longer wavelengths. The values determined in this work fit nicely with previously published values. Fitting a simple power law to the experimental data indicates that the absorption efficiency is proportional to $\lambda^{-2.7}$ with a correlation coefficient of $R^2 = 0.95$, where λ is the wavelength used for the photoacoustic measurement of

B_{abs}. A meaningful evaluation that seeks closure of measured and theoretical light absorption is currently impossible as refractive indices are only poorly known at these wavelengths, aerosol size distribution is generally unknown, and aerosol shape is generally non-spherical.

Correlation of aerosol light absorption measured with the photoacoustic instrument and an integrating plate type filter measurement is fair with a slope of about 3, a zero-offset of about 3.4 Mm^{-1}, and a correlation coefficient $R^2 = 0.72$. Previous suggestions that integration plate type measurements overestimate the aerosol light absorption by some factor could bring the slope closer to one but would not eliminate the sizable zero-offset or improve the correlation. In contrast, application of a novel correction technique based on aerosol albedo [*Horvath*, 199X] has reduced slope and zero-offset to 1.68 and -0.38 Mm^{-1}, respectively, and also improved the correlation coefficient to $R^2 = 0.83$. The slope is still not equal one, however this may be due in part to an effective wavelength for the densitometer of less then 685 nm. For example, assuming a wavelength dependence for B_{abs} of $\lambda^{-2.7}$, as indicated in Fig. 10, would change the wavelength-corrected slope to one if the densitometer has an effective wavelength of 566 nm. While, the results testing Horvath's correction procedure [*Horvath*, 199X] are based on a limited number of samples and a very primitive implementation of Horvath's ideas, they suggest that an improved implementation and more extended testing of these ideas would be a worthwhile endeavor.

Conclusions

Comparison of photoacoustic measurements of aerosol light absorption with aethalometer measurements of BC (black carbon) results in values of light absorption efficiency for black carbon of 5 m^2/g at 685 nm and 10 m^2/g at 532 nm. Comparison of photoacoustic measurements with thermal optical reflectance(TOR) measurements of elemental carbon (EC) yields a somewhat smaller absorption efficiency of 3.6 m^2/g for EC at 685 nm. These values together with previous photoacoustic measurements of aerosol light absorption indicate a wavelength dependency of absorption efficiencies for light absorbing aerosol of $\lambda^{-2.7}$ in the visible and near-visible region.

Integrating plate type filter measurements of aerosol light absorption result in far larger values than those measured with the photoacoustic instrument. A very recently published correction technique [*Horvath*, 199X] can yield improved agreement and the remaining disagreement is possibly due to different measurement wavelengths.

Time series of photoacoustic and aethalometer measurements show good agreement. However, the photoacoustic method is a direct measurement of aerosol light absorption while the aethalometer is not. Therefore, the photoacoustic method is useful to calibrate indirect methods for measuring aerosol light absorption such as the aethalometer. It should be kept in mind that results presented here are based on a limited number of samples at a single location and may not represent aerosol types found at other times or locations. Increased future utilization of the photoacoustic method for ambient measurements of aerosol light absorption is therefore strongly encouraged.

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Table 1: Results of linear regression analyses

date(s)	x-axis	y-axis	slope	zero-offset	R^2
3, 6-9 January 97	TOR EC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	3.58 m^2/g	0.17 Mm^{-1}	0.85
3, 6-9 January 97	PA B_{abs} @ 685 nm (Mm^{-1})	Filter B_{abs} (Mm^{-1})	3.00	3.43 Mm^{-1}	0.72
3, 6-9 January 97	PA B_{abs} @ 685 nm (Mm^{-1})	Corrected Filter B_{abs} (Mm^{-1})	1.68	-0.38 Mm^{-1}	0.83
17 December 96	Aethalometer BC (µg/m^3)	PA B_{abs} @ 532 nm (Mm^{-1})	12.10 m^2/g	0.19 Mm^{-1}	0.90
18 December 96	Aethalometer BC (µg/m^3)	PA B_{abs} @ 532 nm (Mm^{-1})	8.93 m^2/g	1.42 Mm^{-1}	0.87
Av. (17, 18) December 96	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	10.51 m^2/g	0.80 Mm^{-1}	0.88
3 January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	4.61 m^2/g	-0.31 Mm^{-1}	0.85
6 January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	5.43 m^2/g	-0.22 Mm^{-1}	0.88
7 January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	5.44 m^2/g	0.14 Mm^{-1}	0.93
8 January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	4.14 m^2/g	0.08 Mm^{-1}	0.91
9 January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	5.03 m^2/g	0.09 Mm^{-1}	0.91
Av. (3, 6-9) January 97	Aethalometer BC (µg/m^3)	PA B_{abs} @ 685 nm (Mm^{-1})	4.93 m^2/g	-0.05 Mm^{-1}	0.90

Figure Captions

Fig. 1: Aerosol light absorption and scattering coefficients during December 18, 1996

Fig. 2: Aerosol albedo during December 18, 1996

Fig. 3: Correlation between EC and photoacoustic measurements at 685 nm for January 3 and 6 through 9, 1997

Fig. 4: Correlation between photoacoustic measurements at 685 nm and filter absorption measurements for January 3 and 6 through 9, 1997

Fig. 5: Correlation between photoacoustic measurements at 685 nm and corrected filter absorption measurements for January 3 and 6 through 9, 1997

Fig. 6: Correlation between aethalometer BC measurements and photoacoustic measurements at 532 nm for December 17,18, 1996

Fig. 7: Comparison of photoacoustic measurements of aerosol light absorption at 532 nm and aethalometer measurements of BC concentration

Fig. 8: Correlation between aethalometer BC measurements and photoacoustic measurements at 685 nm for January 7, 1997

Fig. 9: Comparison of photoacoustic measurements of aerosol light absorption at 685 nm and aethalometer measurements of BC concentration

Fig. 10: Absorption efficiency of light absorbing aerosols as function of wavelength. Values for laboratory generated aerosols are shown as open symbols, for ambient aerosols as solid symbols

Refs. Fig. 10: [*Adams et al.*, 1989; *Petzold and Niessner*, 1996; *Roessler and Faxvog*, 1979; *Truex and Anderson*, 1979; *Turpin et al.*, 1990]

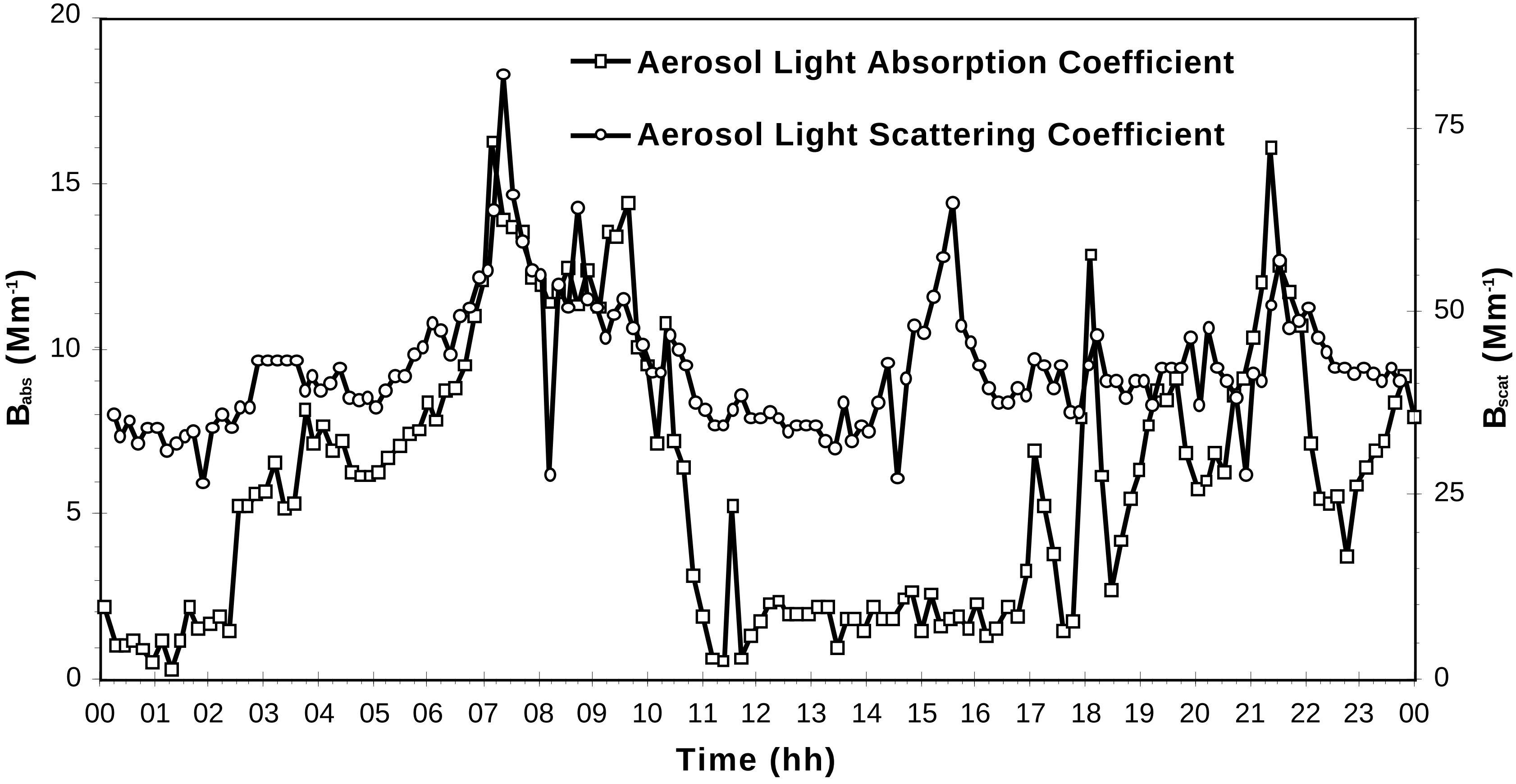

Fig. 1: Aerosol light absorption and scattering coefficients during December 18, 1996

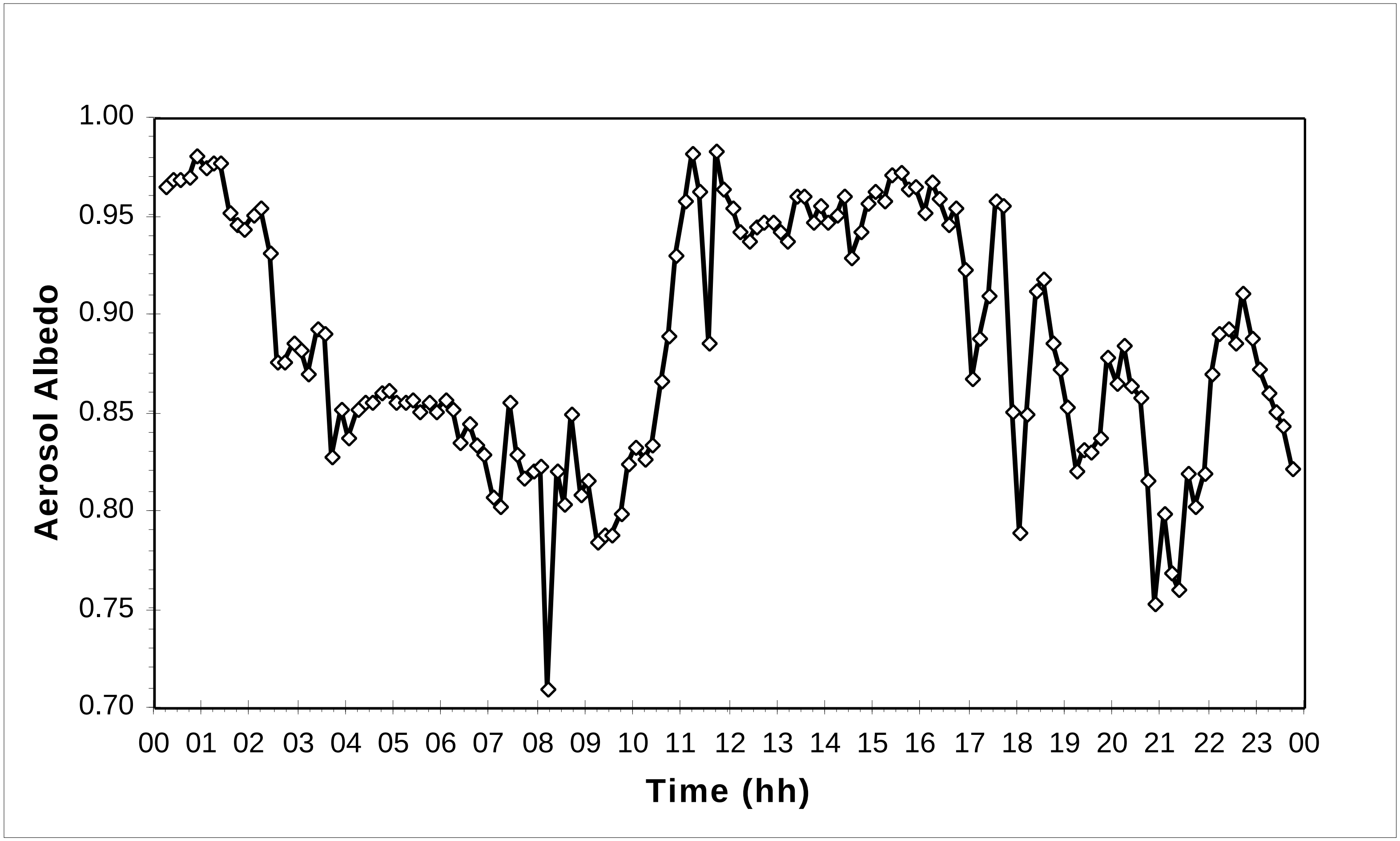

Fig. 2: Aerosol albedo during December 18, 1996

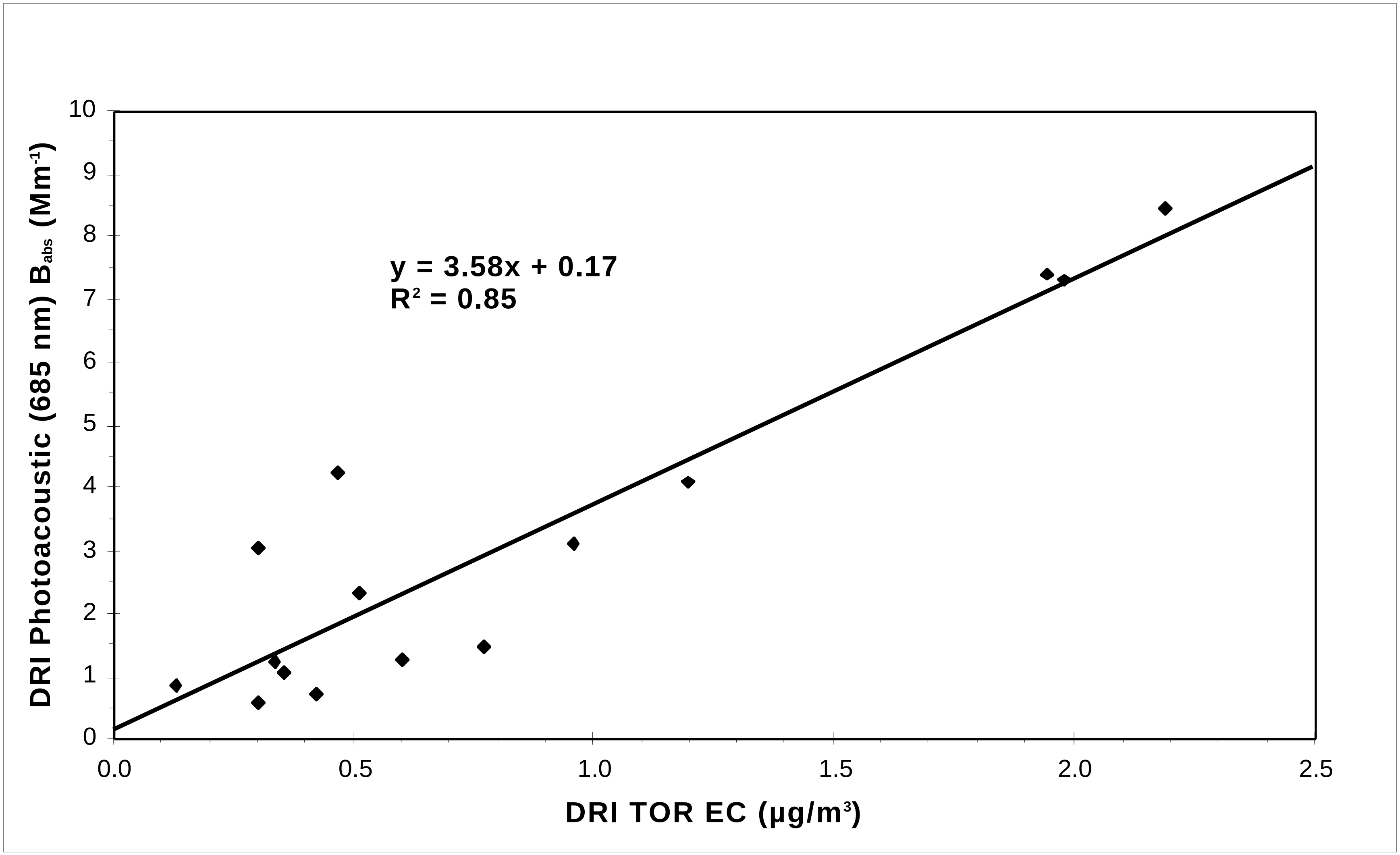

Fig. 3: Correlation between EC and photoacoustic measurements at 685 nm for January 3 and 6 through 9, 1997

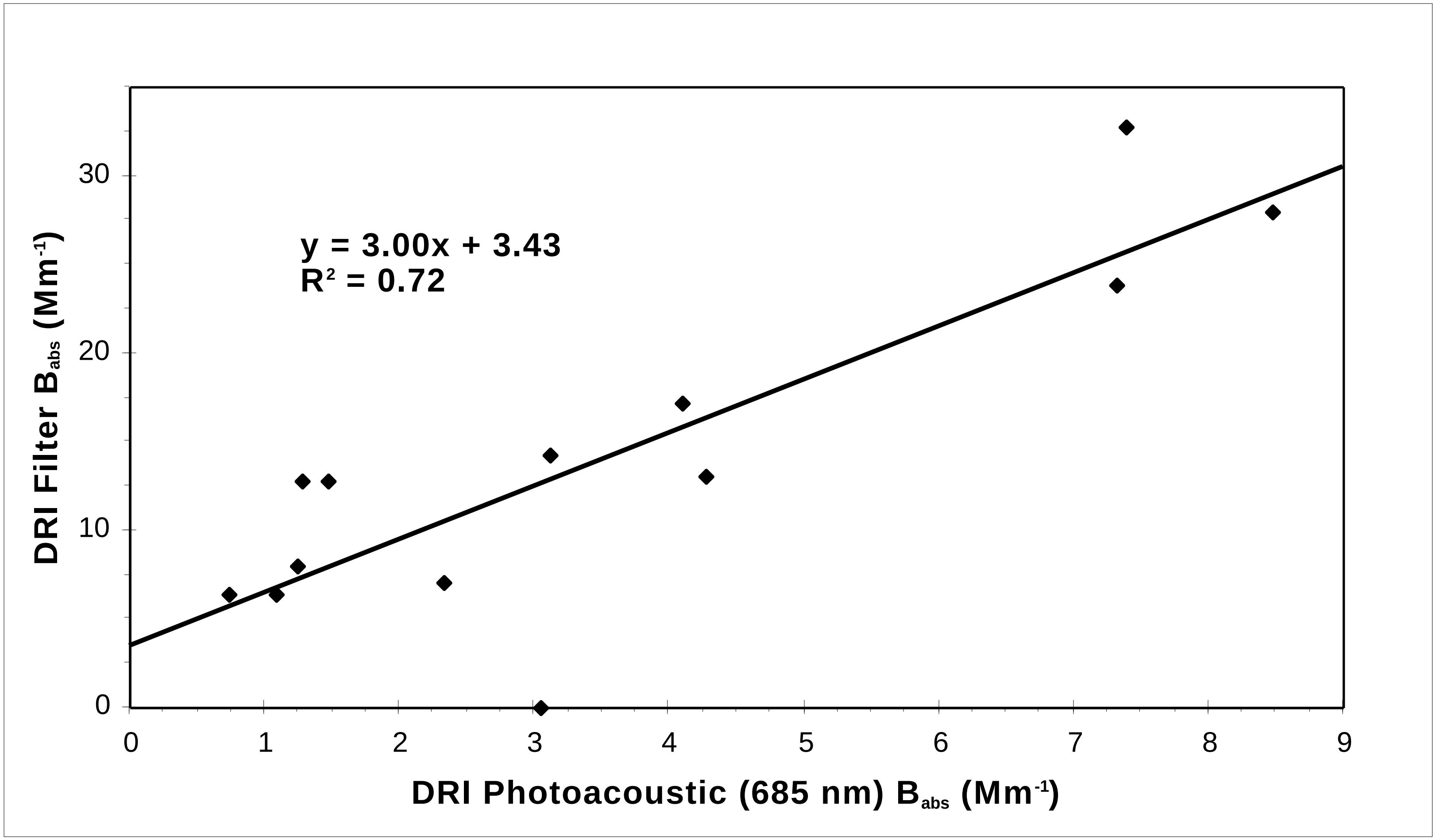

Fig. 4: Correlation between photoacoustic measurements at 685 nm and filter absorption measurements for January 3 and 6 through 9, 1997

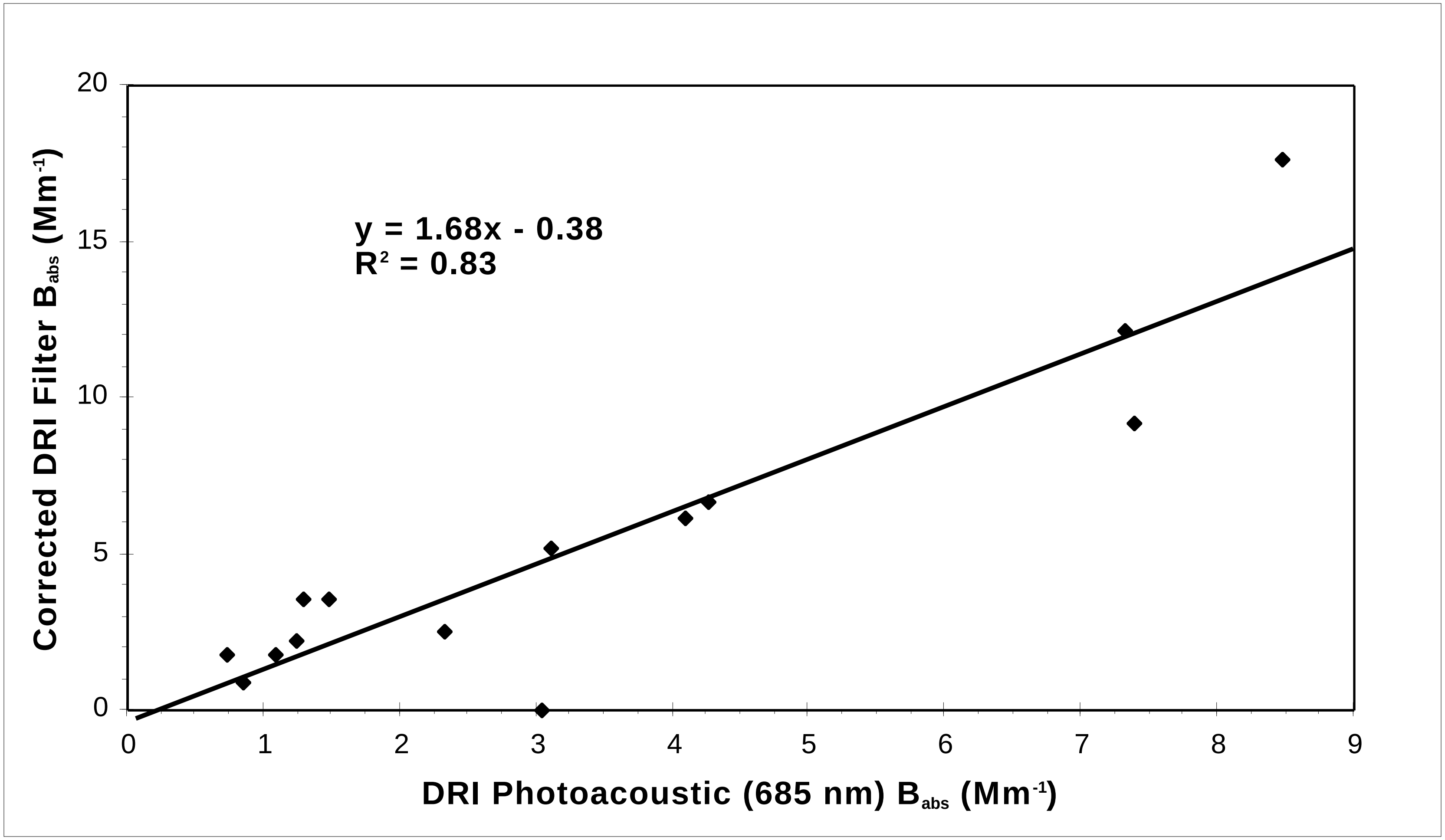

Fig. 5: Correlation between photoacoustic measurements at 685 nm and corrected filter absorption measurements for January 3 and 6 through 9, 1997

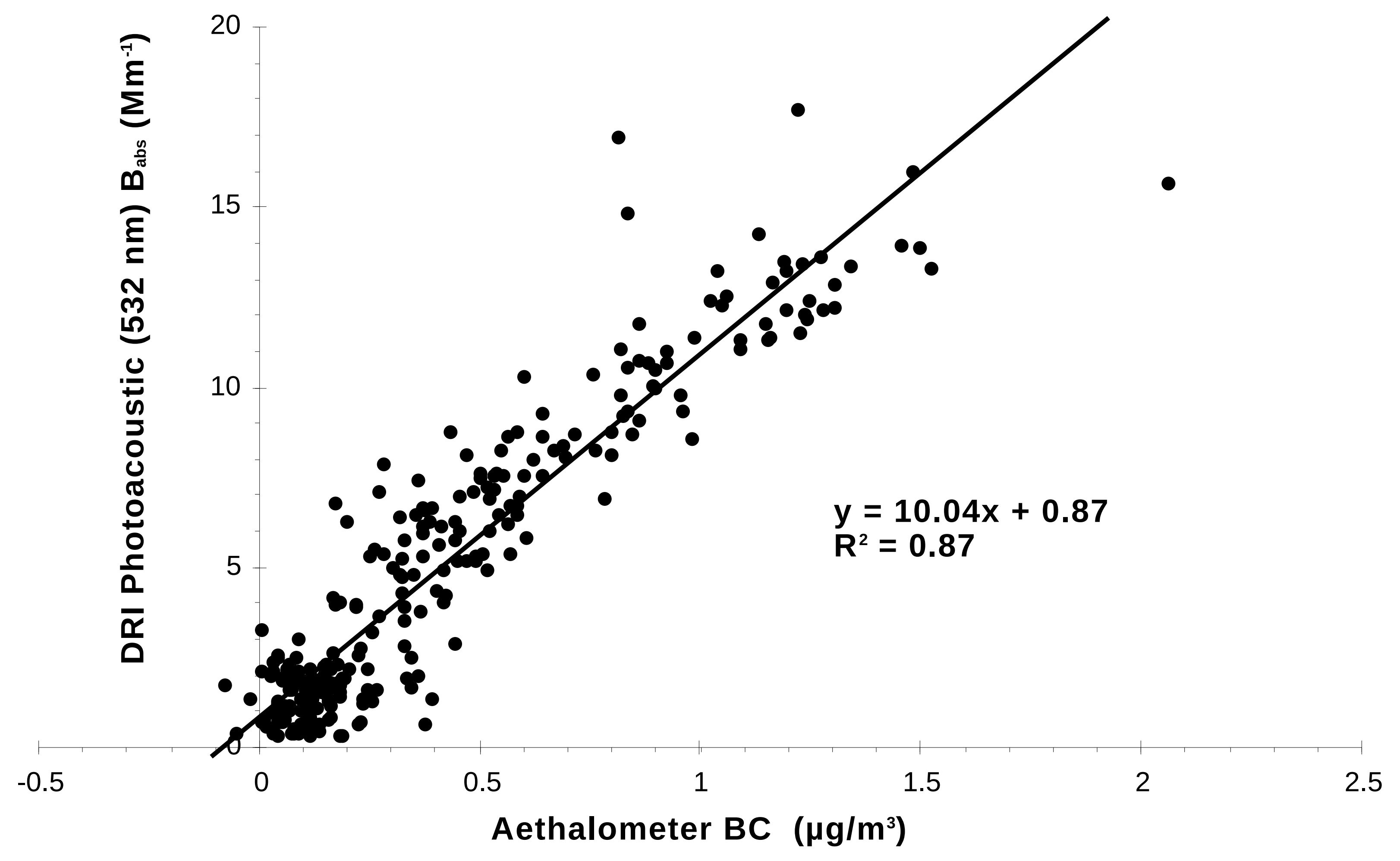

Fig. 6: Correlation between aethalometer BC measurements and photoacoustic measurements at 532 nm for December 17,18, 1996

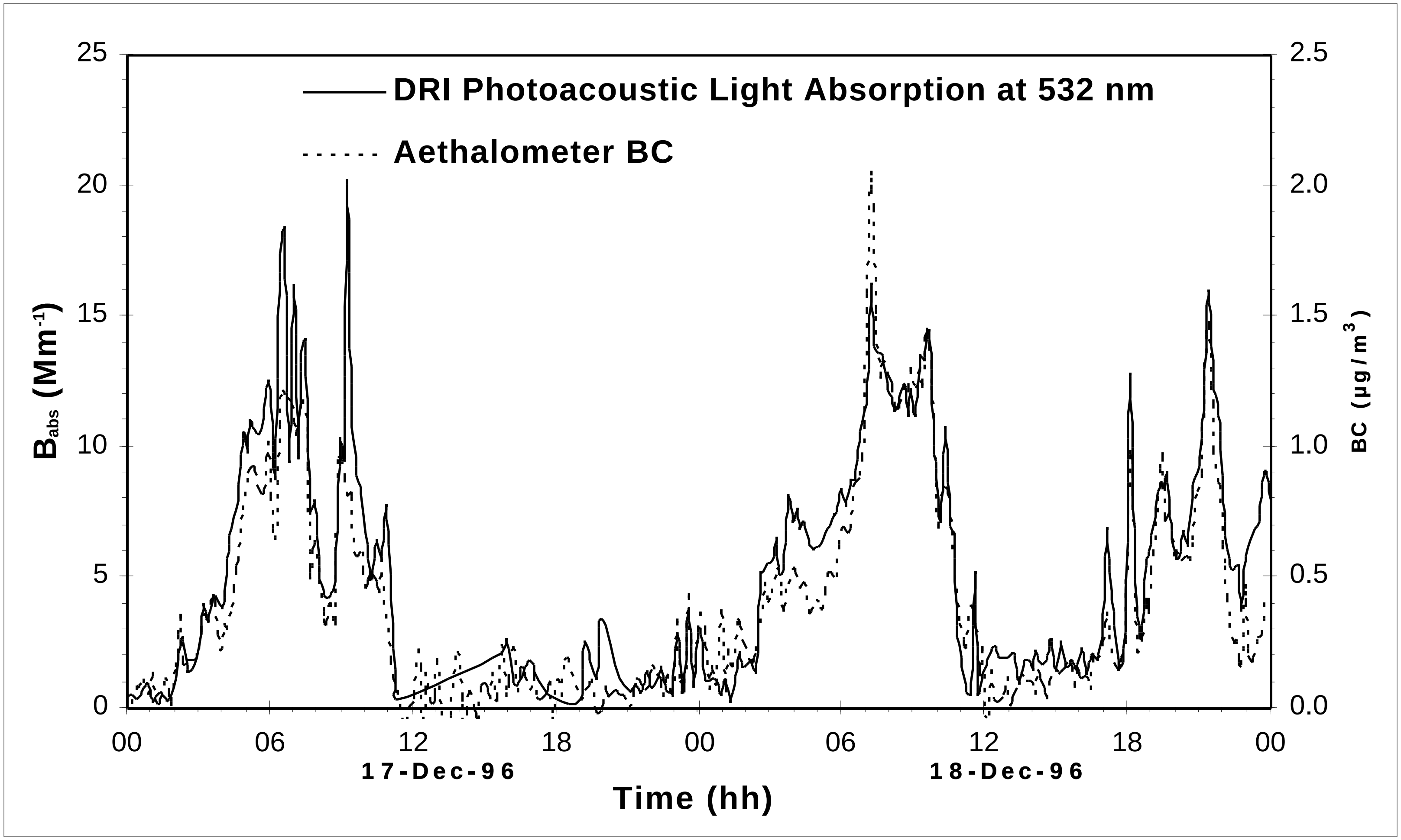

Fig. 7: Comparison of photoacoustic measurements of aerosol light absorption at 532 nm and aethalometer measurements of BC concentration

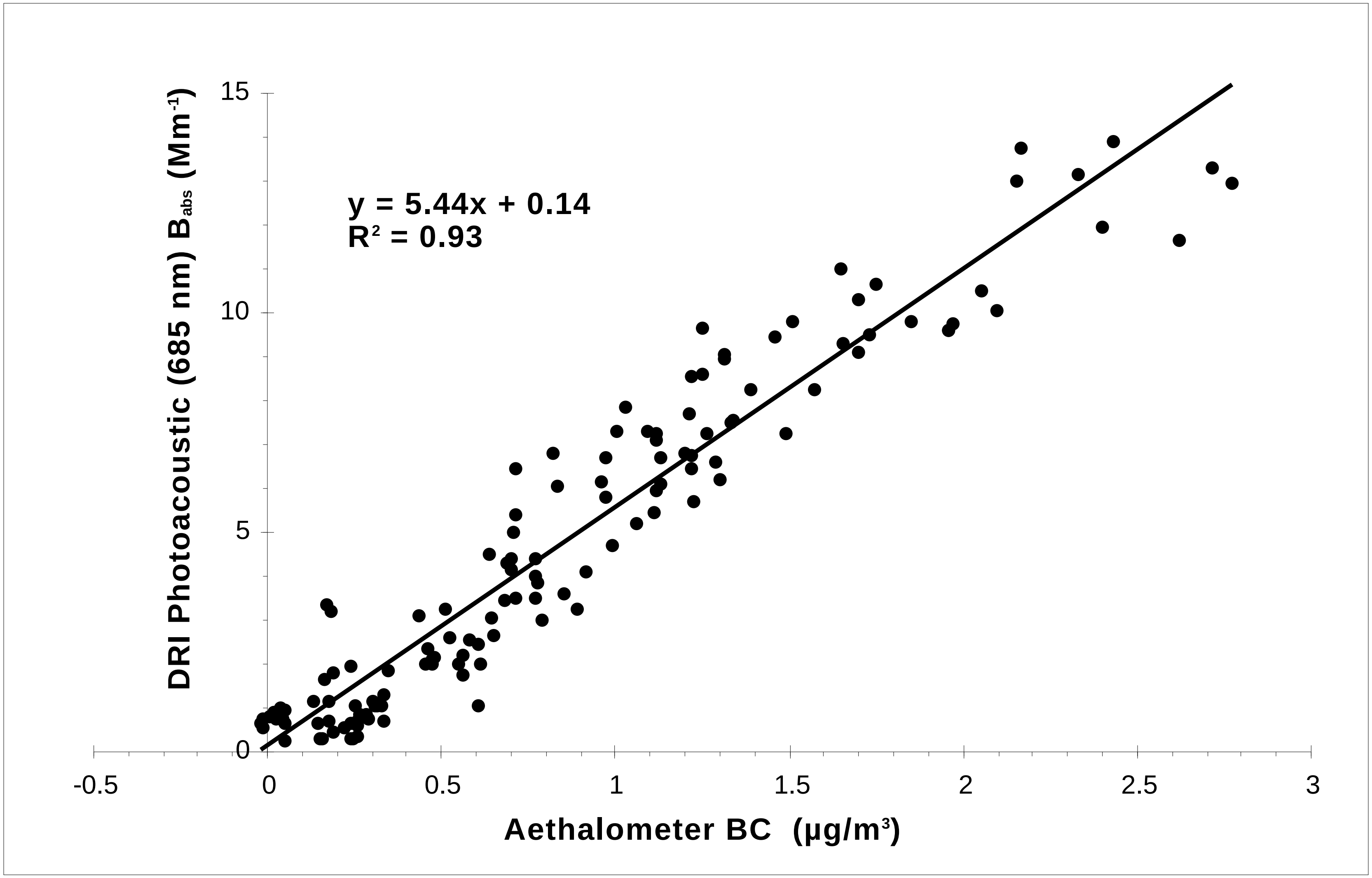

Fig. 8: Correlation between aethalometer BC measurements and photoacoustic measurements at 685 nm for January 7, 1997

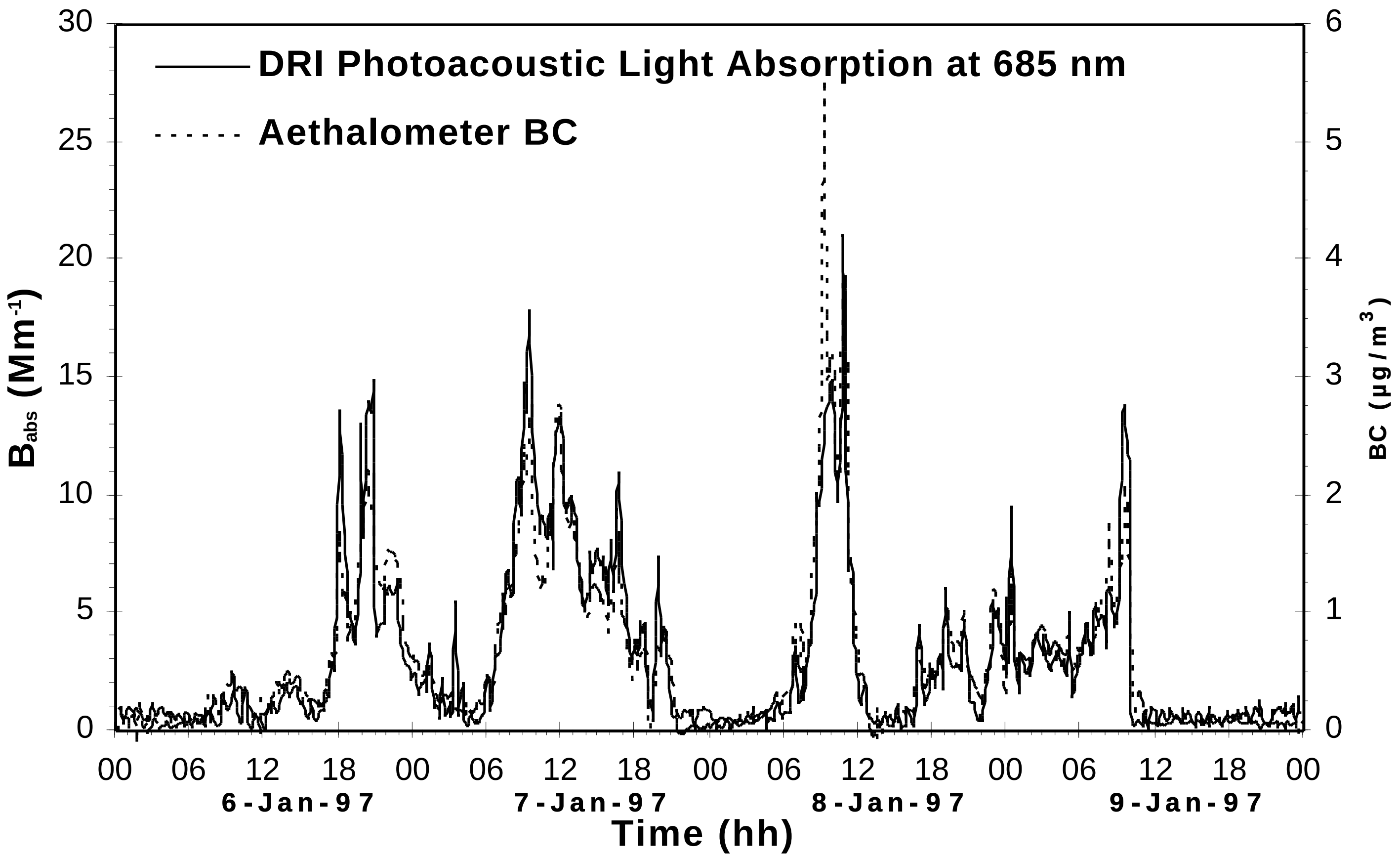

Fig. 9: Comparison of photoacoustic measurements of aerosol light absorption at 685 nm and aethalometer measurements of BC concentration

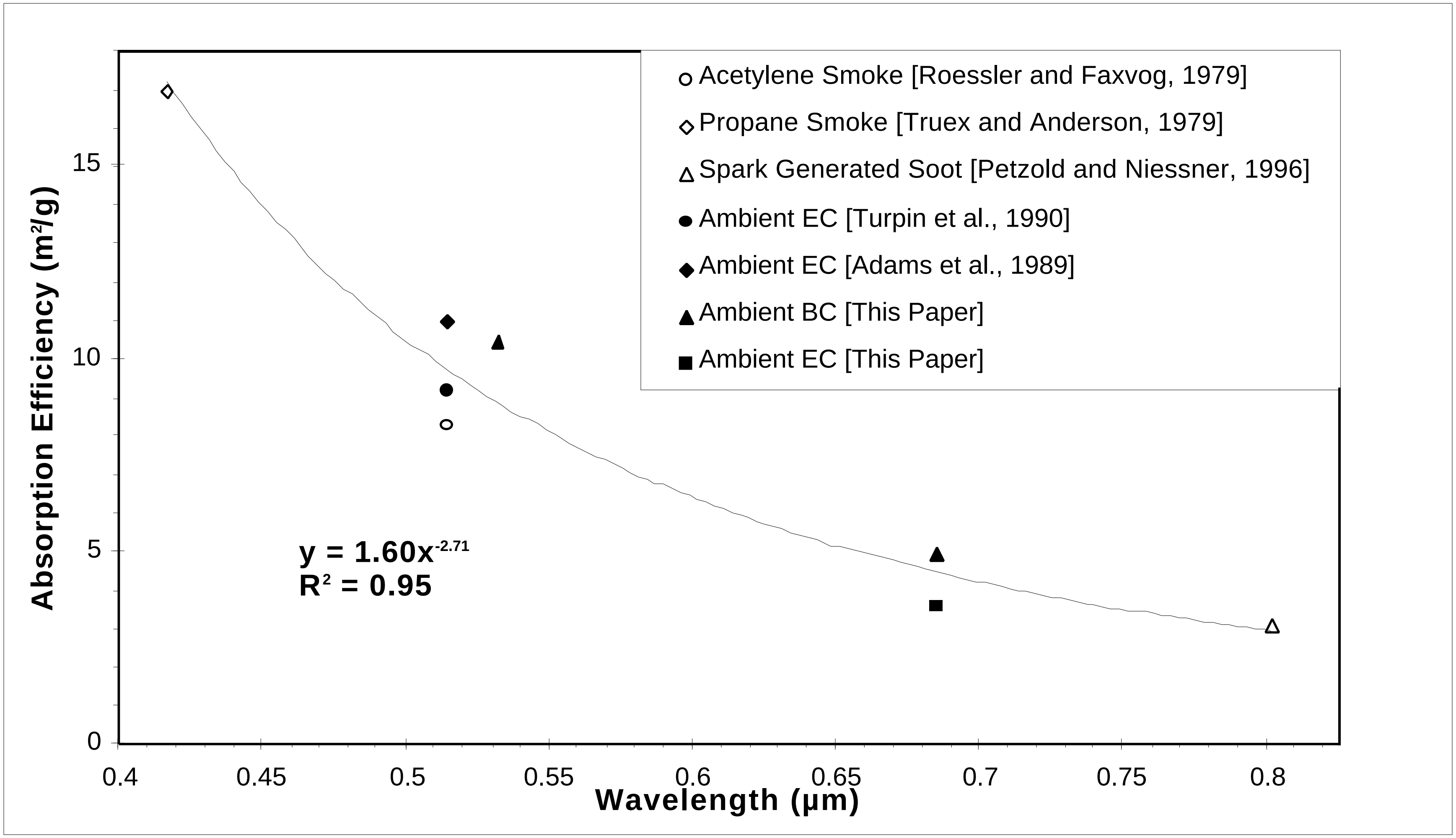

Fig. 10: Absorption efficiency of light absorbing aerosols as function of wavelength. Values for laboratory generated aerosols are shown as open symbols, for ambient aerosols as solid symbols