Aerosol and Air Quality Research, 16: 1–10, 2016

ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2015.05.0356

Real-Time Determination of Absorptivity of Ambient Particles in Urban Aerosol in Budapest, Hungary

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ABSTRACT

The absorption properties of ambient urban aerosols as well as their size distributions were measured in real-time using a dual wavelength optical particle spectrometer (DWOPS). A photoacoustic spectrometer (PAS) was simultaneously used to directly measure light absorptivity of the particles in question along with an Aethalometer and a commercial optical particle counter (OPC). The sizing performance of the DWOPS was compared with the commercial OPC unit showing very good agreement. The data from DWOPS, PAS and Aethalometer instruments are presented and show the potential of the DWOPS technique for a direct and simple assessment of the absorbing part of the complex refractive index of ambient particles as a function of particle size.

Keywords: Atmospheric particles; Light absorption; Refractive index; Soot aerosol.

INTRODUCTION AND MOTIVATION

Much of the concern over atmospheric aerosols derives from their possible effects on human health and environment, including global warming. Besides health concerns the determination of optical properties of soot is of utmost importance. Aerosol light absorption by airborne soot-like particles with its relation to black carbon (Andrae and Gelencsér, 2006) is suspected to greatly contribute to radiative forcing; as much as 50% of the CO_2 forcing (Ramanathan and Carmichael, 2008).

Quantification of the magnitude of these effects still poses considerable challenges due to the rather limited data and its understanding when it comes to the absorptivity of ambient aerosols. The significance of gaseous contributions to global warming is now well recognized and their impact can be accurately evaluated. Understanding of the impact of aerosols is a more challenging issue. Physical processes involving aerosols and their direct or indirect effects on the optical properties of the atmosphere are still an open field of research that is only roughly described in the models, hence demanding further reliable data collection. This issue becomes increasingly complex when absorbing particulate matter has to be taken into account. Incineration processes as well as the combustion of fossil and biomass fuels generate soot-like particles that may strongly differ in morphology and chemical composition. The latter can be determined by e.g., Raman spectroscopy, electron microscopy or the cavity ring-down technique (Mason *et al.*, 2012, Bulatov *et al.*, 2002), however these methods so far are impractical for routine, or field measurements of particle absorbing properties.

In principle, light absorption by airborne aerosols can also be determined by "difference methods", where absorption is determined from the difference in extinction and scattering measured simultaneously by an extinction cell and a nephelometer. While this method can be useful for the validation of absorption techniques, it may result in substantial errors when concluding the absorptivity from the difference of two large valued extinction and scattering coefficients (e.g. Schnaiter *et al.*, 2005).

Consequently, an assessment of the particle absorptivity while still airborne is essential but the variability of soot-like particles likely changes its optical properties making it rather difficult to be determined directly (Lee, 2014). Usually the optical properties determined relate to a particulate measuring technique. A major limitation is the absence of established standards or reference data for instruments' calibration that may be applied to measure the various properties of soot (Baumgardner *et al.*, 2012).

Various methods and instruments have been developed for measuring some of optical aerosol properties (Zhao *et al.*, 1997; Spindler *et al.*, 2007; Moosmüller et al, 2009), however to quantitatively determine absorptive properties of aerosol and the presence of carbonaceous particles is still a challenge because the question of which parameter

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best mirrors the actual environmental burden. Widely spread filter deposition measurements or collection and post-sampling evaluation is prone to various errors (Petzold *et al.*, 2004; Watson *et al.*, 2005; Subramanian *et al.*, 2007). Depending on the technique used and applied, correction errors of up to a factor of two can be expected when inferring the in-situ aerosol absorptivity from filter-based measurements (Andrae and Gelencsér, 2006).

In this contribution we present data on absorptive properties of urban aerosols based on real-time evaluation of scattered light using the custom-built Dual Wavelength Optical Particle Spectrometer (DWOPS) alongside data simultaneously collected by another custom-built instrument based on the photoacoustic measurement, the 4λ-PAS (photo-acoustic spectrometer) (Arnott *et al.*, 1999, Ajtai *et al.*, 2011). Moreover, we compare our methodology with two commercially available instruments, the Magee Scientific Aethalometer A31 and a conventional optical particle counter (PAS – Portable Aerosol Spectrometer Mod.1.109, Grimm Aerosol Technik; in order to avoid confusion regarding the acronym (PAS) the PAS Mod.1.109 will be abbreviated OPC (optical particle counter) in this paper). Conventional optical particle counters/spectrometers measure a single pulse resulting from the interaction of a particle with an illumination source to measure the particle size. A clear disadvantage of this method is that particle sizing depends on the complex refractive index and may vary greatly between different refractive indices. Nevertheless, particle counting (measurement of number concentrations) and in a limited sense particle sizing (measurement of optical equivalent diameter) with such instruments is feasible (e.g., Szymanski and Liu, 1986, Burkhard *et al.*, 2010, Görner, 2012; Belosi, 2013).

The DWOPS is a spectrometer with a design which uses two illumination sources with different wavelengths. This provides sufficient information to obtain unique data about the optical particle size and its complex index of refraction, especially the imaginary part representing the absorptivity (Szymanski *et al.*, 2002; Nagy *et al.*, 2007; Szymanski *et al.*, 2009). The real-time determination of ambient aerosols' absorbing properties may provide information regarding the origin of atmospheric particulate matter. The day time absorbing fraction is likely due to the traffic conditions, whereas the night time absorptivity could be linked to biomass burning, at least in the winter time (Sandradewi *et al.*, 2008; Favez *et al.*, 2009). Furthermore, knowledge of the particle number concentration and the value of the refractive index, especially for the absorbing portion of atmospheric aerosols can be used to more completely describe the ambient atmospheric situation to deliver much needed data to study radiative transfer through the atmosphere (Bergstrom, 2007).

INSTRUMENTS AND SAMPLING PROCEDURE

Atmospheric particulate matter (PM) in size ranges corresponding to ambient monitoring standards was investigated. The monitoring of ambient number concentrations and their correlation with the absorbing particles' properties was endeavored using a custom-built dual wavelength optical particle spectrometer (DWOPS), custom-built photo-acoustic spectrometer (4λ-PAS) and the Aethalometer. This data was linked to ambient conditions such as wind direction, temperature, and relative humidity. As the DWOPS, despite being described in the literature, is not yet commercially available we compared the monitored number concentrations with that measured by the OPC (Mod.1.109). For this comparison we selected a particle size range from 0.6–5 μm, in which particles were measured by both the DWOPS and OPC instruments, with 100% efficiency.

All instruments were placed inside a temperature and humidity controlled mobile laboratory station equipped with an inlet for the total suspended particulates (TSP) located on the roof of the van in the height of about 3.5m above street level. The sampling lines were short and vertically directed from the inlet into the instruments.

The present data was measured during a field measurement campaign in March 2010 at the observation site of the Hungarian Meteorological Service (Gilice sq. Budapest, GPS position: 47°25'52.9"N 19°10'56.4"E) and at Astoria place (ELTE University Campus, GPS position: 47°29'35.5"N 19°03'39.4"E). The daily average air temperature during the campaign varied between –4 to 4°C. The Gilice sampling point is located between two major roads several hundred meters away. Astoria sampling point is located in downtown Budapest near a road with high traffic density.

Aethalometer

The Magee Scientific Aethalometer A31 acquires data through seven channels at wavelengths ranging from ultraviolet to near-infrared. An algorithm converts the incremental optical signals in each channel to a mass of material. If the aerosol consists of only extremely small pure black spheres, the signals in all channels would be interpreted identically as the same mass of material, and the seven data chart traces would be perfectly superimposed. Deviations from this spectral uniformity could be caused by several factors including: large particle size (more absorption per unit mass in the red, less in the blue wavelength range); presence of aromatic organic compounds (onset of enhanced absorbance in blue and ultraviolet channels); colored mineral dust (extra absorption in particular channels); and other effects due to the specifics of the aerosol composition.

There is an ongoing dispute regarding the data deviations from the absorbance law, nevertheless the data provides a certain insight toward the understanding of aerosols' optical particle properties (Sandradewi *et al.*, 2008).

OPC - Portable Aerosol Spectrometer Mod. 1.109

This OPC measures particle concentration as well as size and as specified by the manufacturer provides information over the particle size range of 0.25–32 μm in 30 channels. It uses a 683nm laser diode to illuminate the aerosol beam, and an off-axis wide-angle collecting optics to detect light pulses with a photo diode. The optical arrangement with the mean scattering angle 90° collects the elastically scattered light within 120° on one side and an additional 18° on the

opposing side using a parabolic mirror. We compare our measurements with that of this spectrometer because this instrument is also used for ambient air monitoring in Hungary, although some of its limitations have been described recently (Heim *et al.*, 2008; Burkhard *et al.*, 2010).

Photoacoustic Spectrometer – The 4λ-PAS

The photo-acoustic spectrometry method used is based on the idea of determining light absorption from the amplitude of a sound wave generated by the absorption of modulated light (Rosencwaig, 1980). The advantage of this technique is that it is not sensitive to the scattering of light, and gives a linear response in relation to the absorptivity of aerosols (Arnott *et al.*, 1999; Krämer *et al.*, 2001).

However, this technique may only be applied to rather polluted air due to its limited sensitivity (Lack *et al.*, 2006; Bozoki *et al.*, 2011). The 4λ-PAS system used in this study is described by Ajtai *et al.* (2011), where the measurement results are also presented. The limitation of this method as it was mentioned earlier is that it requires ambient concentrations of the absorbing species on the order of 1 $\mu g\ m^{-3}$.

Since the data obtained from the 4λ-PAS shows remarkable agreement with the data measured by the 7λ-Aethalometer presented in (Ajtai *et al.*, 2011), we refer only to the data measured by the Aethalometer in the manuscript.

Dual Wavelength Optical Particle Spectrometer – DWOPS

This measurement method was first introduced in 2002 (Szymanski *et al.*, 2002) and a laboratory prototype was built in 2007. At this time comprehensive numerical performance test together with the first laboratory measurement results were published (Nagy *et al.*, 2007; Szymanski *et al.*, 2009). A mobile prototype instrument was developed and built to study the method during field measurement campaigns and in order to compare its results with that of other devices.

Although the described prototype operates in the size range from 0.6–5 μm it attests the feasibility of the method for measuring the particle size and the absorptivity of investigated particles. The current instrumental limits are not of the principal nature but are dictated by current opto-mechanical restrains of the prototype.

Due to its small size and weight as well as real-time capacity this instrument is very suitable for field measurements determining the absorbing properties of ambient aerosols, monitoring particles generated by industrial activities, and for laboratory studies.

For a spherical particle, light scattered into a given angular range is determined by three quantities - the particle size, and the real and the imaginary part of the complex refractive index. Common optical configurations of OPCs consist of a single light source and a detection system encompassing a given angular range in which light flux scattered from a single particle is measured. From such optical systems no additional information can be obtained about the particle in question, except its size, with certain limitations.

The DWOPS allows to approach the *a priori* ill-posed inverse measuring problem (Bohren and Huffman, 1983) linked with any optical particle measurement. It measures and evaluates light scattered from an individual particle into four different angular ranges. Individual particles in the sensing volume of the device are illuminated by two fairly linearly polarized (10:1) laser diodes (red, with $\lambda = 660$ nm and infrared, with $\lambda = 808$ nm). Light is scattered from particles into angular ranges (10°–30° and 150°–170°) in the forward and backward direction with respect to the direction of propagation of each laser. This scattered light is then separated by the dichroic beam splitters according to its wavelength, measured by an appropriate detector and converted into a voltage pulse with a magnitude proportional to the amount of scattered light. This procedure yields a quadruple set of independent voltage pulses from each single particle, in contrast to conventional OPCs measuring a single pulse from every detected particle. Knowing exactly the scattering geometry of the apparatus, as well as the properties of illumination sources and detectors, one can calculate by means of the MIE theory the expected electrical response of each sensor. Combining this information with the measured data simultaneously provides information about equivalent optical particle size and particle's complex refractive index (real and imaginary part).

Refractive indices are in general wavelength dependent. Due to the rather narrow interval of wavelengths used in DWOPS this dependence is not significant.

In this contribution we focus on the absorptivity of aerosols. For completeness it needs to be mentioned that the real part of refractive index was also measured with values typically within the interval 1.5–1.8.

RESULTS AND DISCUSSION

The variation of ambient number concentrations as a function of time shows remarkable correlation for both devices (DWOPS and OPC) in the corresponding size ranges collected over a period of 330 hours, at Gilice Square (Fig. 1). The agreement between the commercial OPC and the custom-built DWOPS data for ambient particle counting is evident.

The pronounced peaks (at the 40^{th}, 64^{th} and 175^{th} hours of sampling correspond well to the rapid change in environmental conditions (wind and humidity) evident from Fig. 7, which will be discussed later in this contribution.

Besides particle counting and particle sizing, in contrast to currently available spectrometers and optical particle counters, the DWOPS allows the assessment of the complex refractive index of single, detected particles. In this contribution we focus especially on the absorbing properties of ambient aerosols. Figs. 2 and 3 show the fractions of detected particles which are absorbing (the imaginary part of the complex refractive index $m_{IM} > 0.1$) which was measured by DWOPS at the sampling locations as described above (Gilice Square and Astoria). This data was subdivided into the sub- and super-micron size range. The latter one was further subdivided using as boundary the particle diameter of 2.5 μm, which corresponds to the $PM_{2.5}$ value used in air quality studies. The trends in the absorbing fraction in the different size ranges show characteristic differences between the two locations. While the absorbing fractions of the

super-micron particles are in the 10–20% range in both locations, the absorbing fractions of the sub-micron particles are higher at Gilice sq. and lower at Astoria. In the first three days of the sampling at Gilice square, when the weather conditions (wind direction, temperature, rel. humidity) were different from the weather conditions of the rest of the

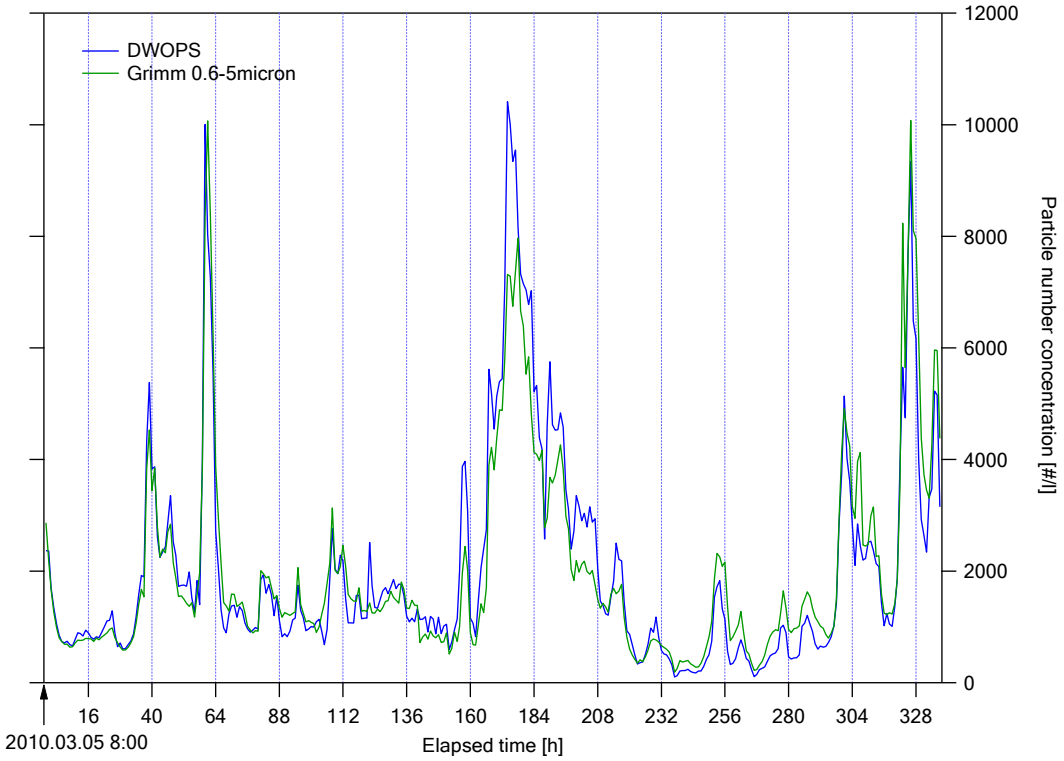

Fig. 1. Comparison of concentrations measured by DWOPS and OPC Mod.1.109 in the 0.6–5 micron size range.

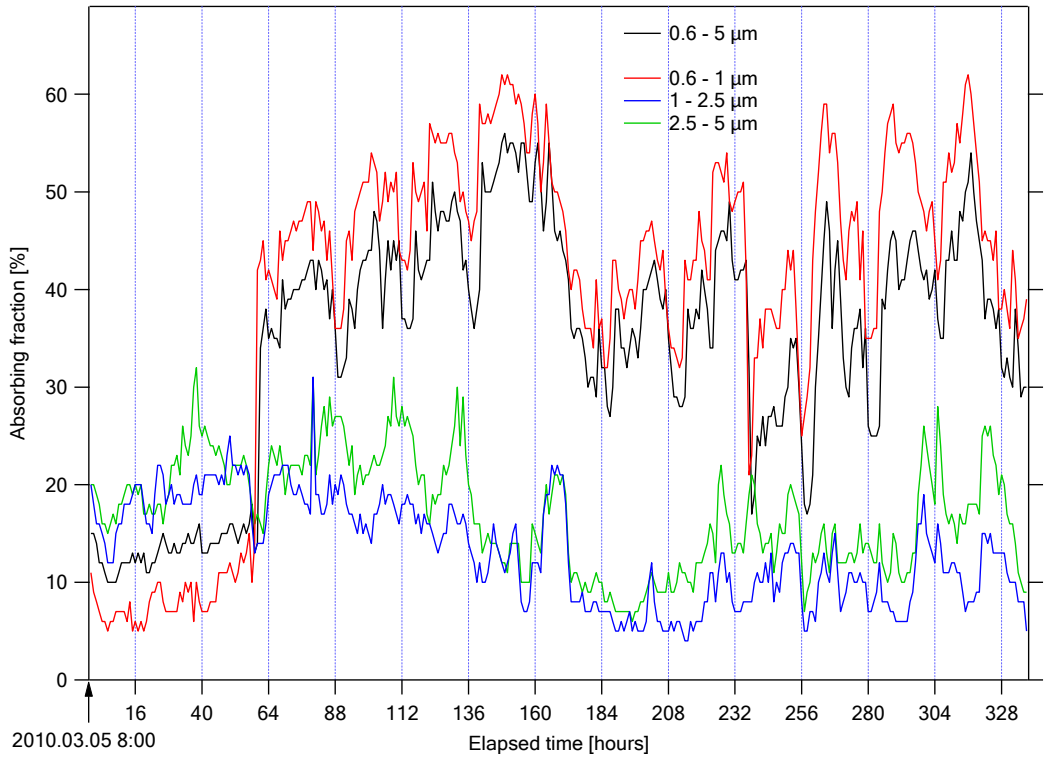

Fig. 2. Fraction of detected particles having non-zero absorptivity as a function of size interval measured by the DWOPS at Gilice square.

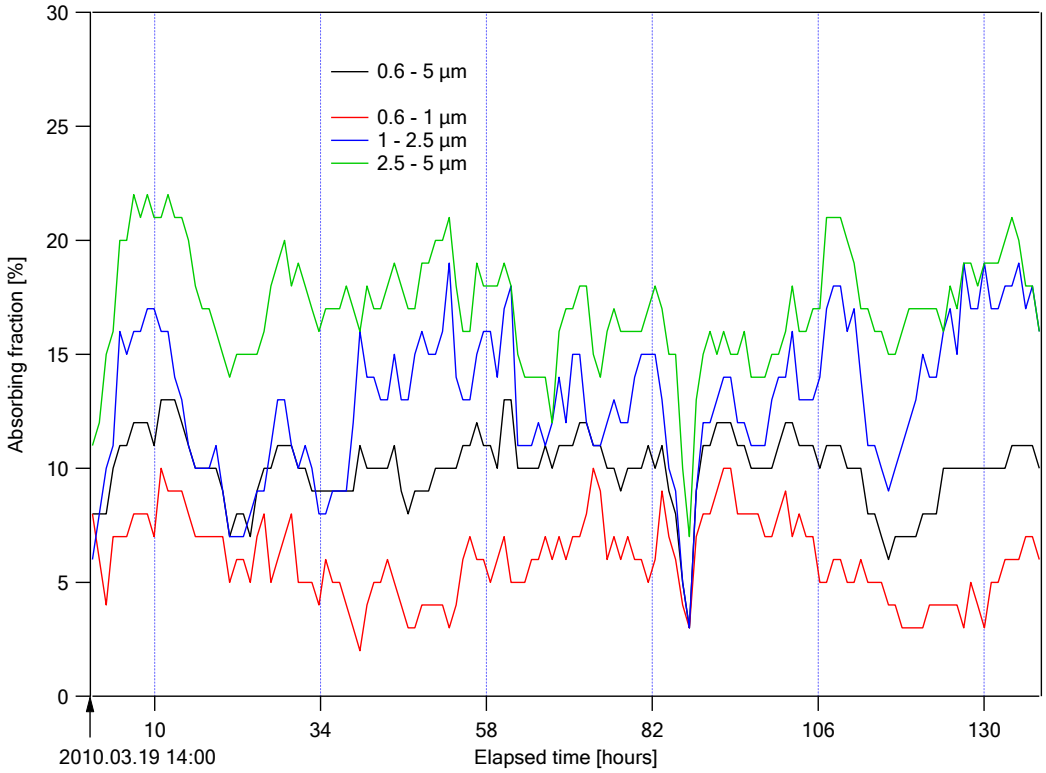

Fig. 3. Fraction of detected particles having non-zero absorptivity as a function of size interval measured by the DWOPS at Astoria.

measurement campaign, the absorbing fractions in the different size ranges showed similar behavior to the results at Astoria. Although, further investigations are necessary to explore the reasons for this type of behavior, aerosol aging (coagulation, aggregation, condensation, etc.) might be a possible reason. Traffic generates absorbing particles typically below the detection capabilities of the DWOPS. Consequently, they cannot be detected near the traffic route, but after a certain distance, (100–200 m away from the route) their size increases due to different processes, and they may contribute to larger particle fractions.

Concurrently with the spectrometers' measurement the 4λ-PAS and the Aethalometer were operated to obtain information related to soot, specifically for the black carbon (BC) in the urban aerosol (Andrae and Gelencsér, 2011). Data evaluated at the wavelength of 950nm should correspond well to the mass concentrations of the absorbing species. Fig. 4 shows the Aethalometer determined mass concentration of the BC vs. time in ambient aerosol at Gilice Square. Because in our measurements we aimed only at time correlations, we did not apply any loading corrections. Also, due to rather clean ambient conditions the usual artifacts linked with Aethalometer data were not relevant (Weingartner *et al.*, 2003). The mass concentration varies around 2000 ng m^{-3} with pronounced peaks due to wind direction changes as discussed below (Fig. 8). Additionally, the number concentration values obtained by the OPC (Mod.1.109) were divided into size ranges: 0.25–1, 1–2.5 and 2.5–10 µm in terms of particle (optical) diameter and are shown as a function of time. It is evident that the morphology of the Aethalometer data (mass concentration variation vs. time) corresponds well to the OPC data allowing the observation that the sub-micrometer particle component contains substantial amounts of the absorbing material.

The absorptivity of ambient particles cannot be deduced from data obtained with conventional OPCs such as the Mod.1.109, or similar instruments. The DWOPS principle conversely allows to determine besides the ambient particle size distribution, the absorptivity of particles in question. Figs. 5 and 6 show data from Gilice Sq. and Astoria, respectively. Both figures show the average absorbing part of the complex refractive index of measured particles. The data is again split into size ranges: 0.6–1, 1–2.5 and 2.5–5 µm (corresponding to the operating range of the current version of DWOPS). These data show clearly that the sub-micrometer fraction of ambient particles are composed of strongly absorbing material with value of about 0.7, indicating very strongly relationship to atmospheric soot particles. As the absorption decreases with the increasing particle size one could hypothesize that in the case of sub-micrometer particles the external mixture of aerosol dominates, whereas for the larger aerosols the internal mixture dominates (Levoni *et al.*, 1997).

Fig. 7 combines environmental data (wind direction, ambient relative humidity and temperature) with the aerosol data – mass concentration of BC particles (Aethalometer), number concentration of airborne particles (DWOPS), and the fraction of ambient particles which are optically absorbing (DWOPS). Clearly the wind direction changes are visible in all data sets. It is shown that the BC mass concentration

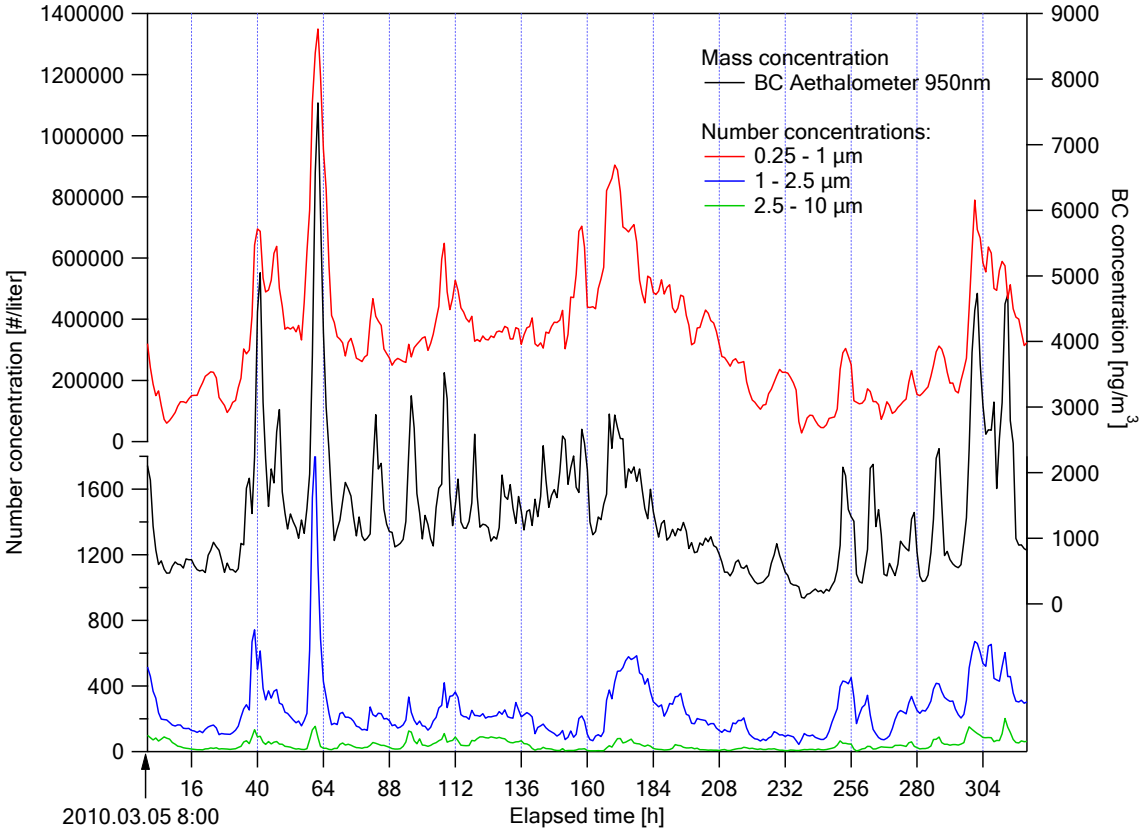

Fig. 4. Measured number and BC mass concentrations at Gilice square.

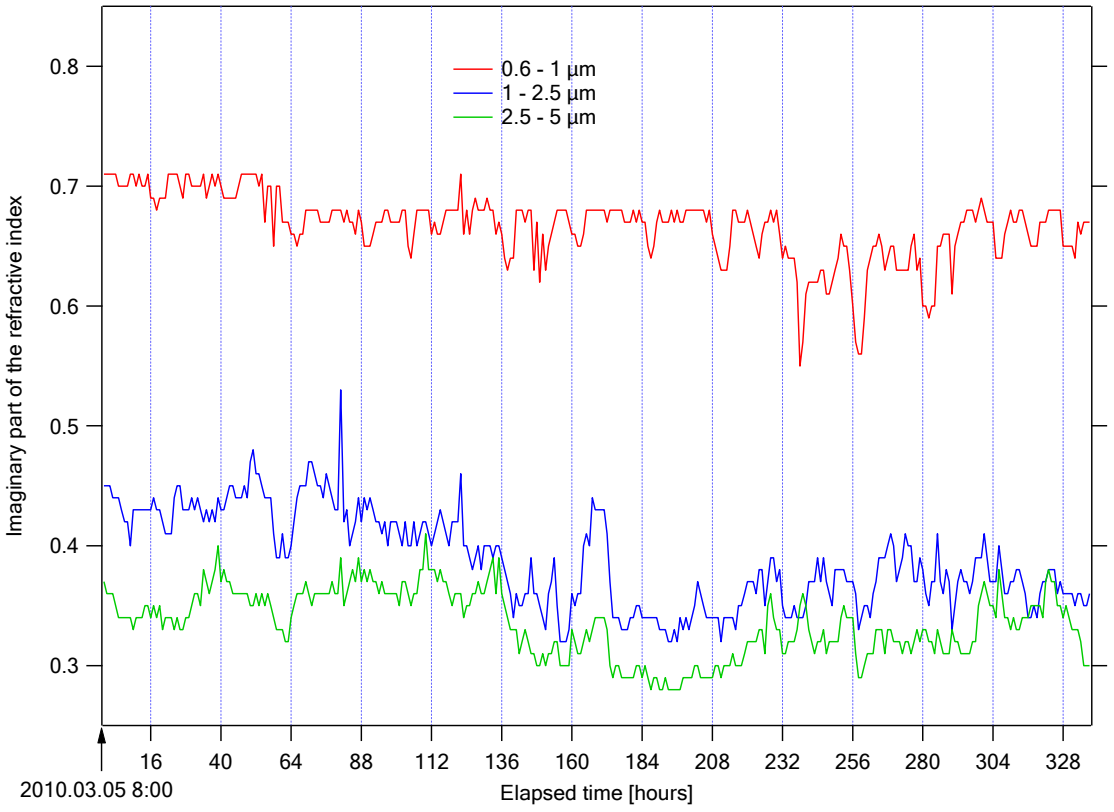

Fig. 5. Absorptivity (hourly average of the imaginary part of the refractive index) of ambient particles in various size ranges as a function of time at the location Gilice square.

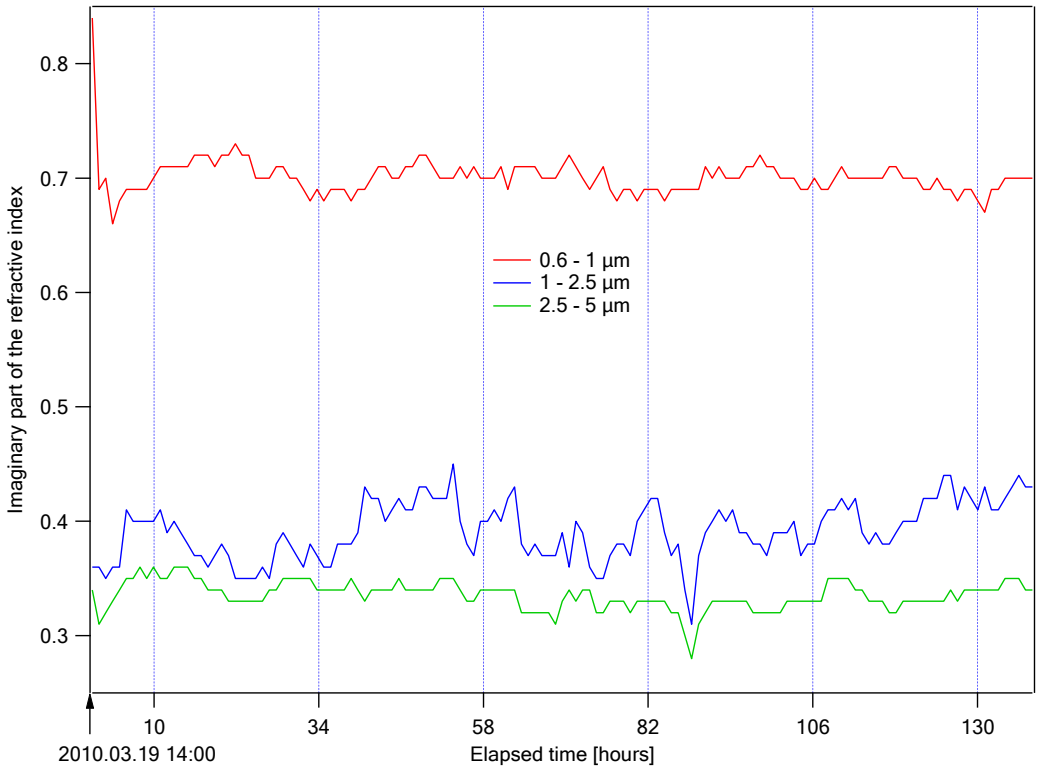

Fig. 6. Absorptivity (hourly average of the imaginary part of the refractive index) of ambient particles in various size ranges as a function of time at the location Astoria.

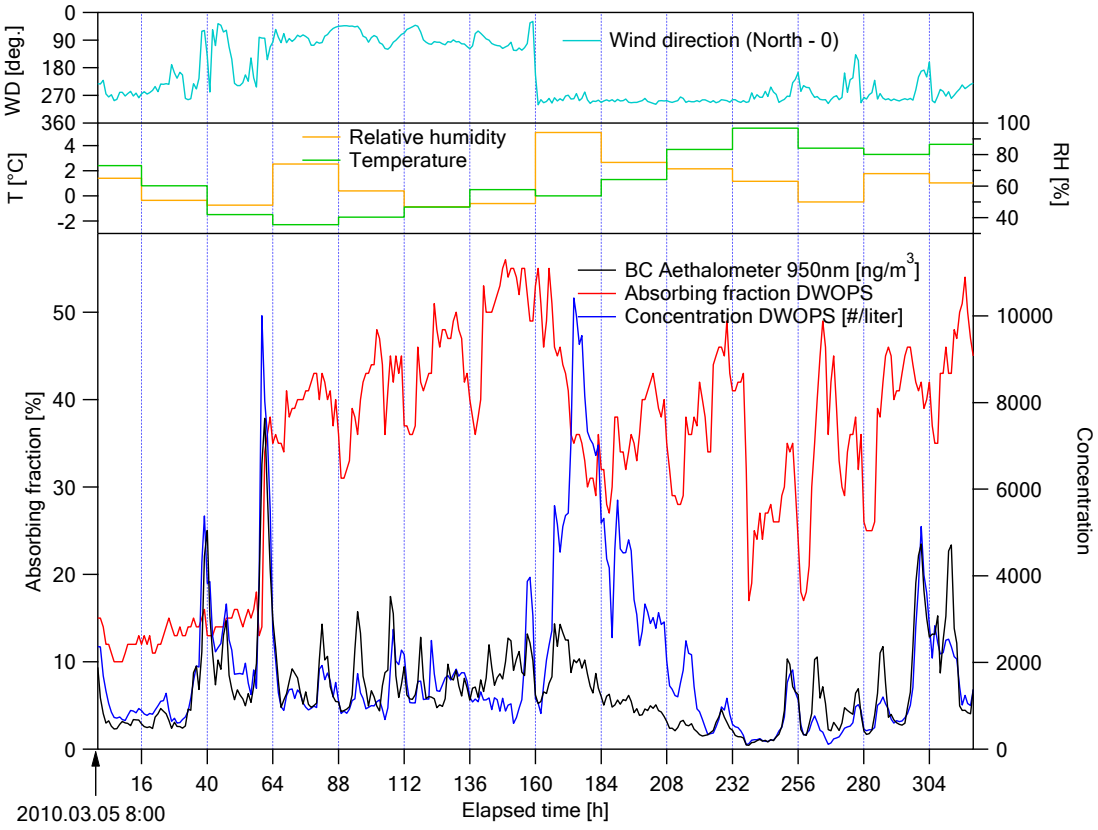

Fig. 7. Summary of the aerosol data – mass concentration of BC particles (Aethalometer), number concentration of airborne particles (DWOPS) and the fraction of ambient particles which is optically absorbing (DWOPS) and environmental data (wind direction, ambient relative humidity and temperature) measured at Gilice square.

values correlate rather well with the number concentration of particles which is shown in Fig.4.

The unique property of the DWOPS is that it simultaneously measures particle size and its complex refractive index allowing extraction from the data the number concentration of absorbing particles in the measured size interval as a function of particle absorption (Im (m) imaginary part of the complex refractive index m).

The diagrams in Figs. 8 and 9 show the data in both sampling locations. It is evident that a substantial part of measured particles is non-absorbing, or moderately absorbing (Im (m) $\in$ [0, 0.2]. The refractive index of BC is usually estimated to be around m $\approx$ 2 – 0.8i (Bond and Bergstrom, 2007).

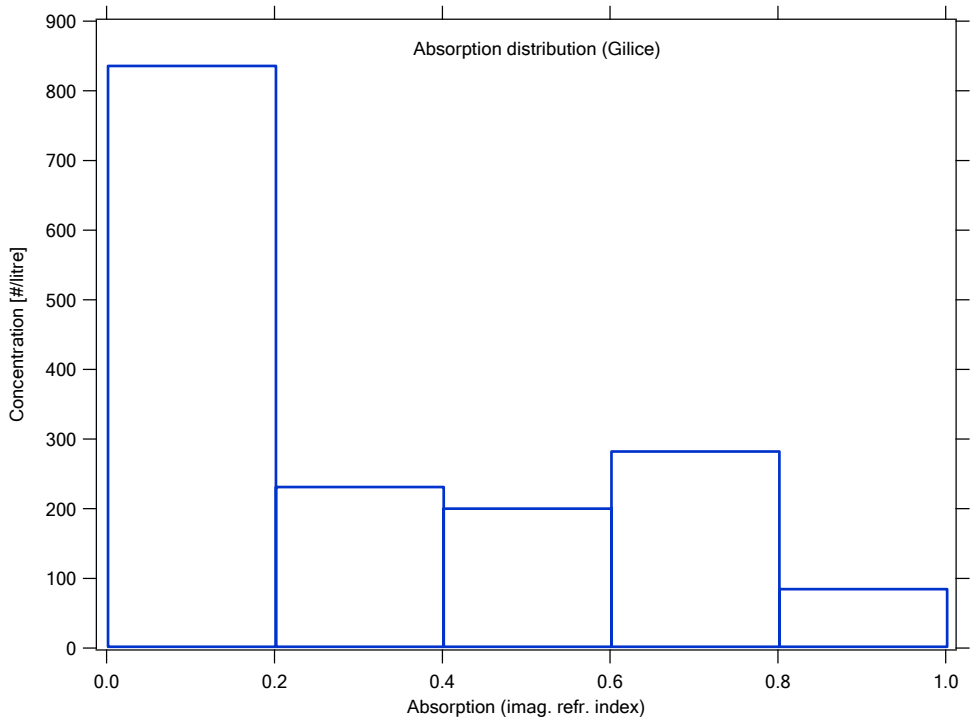

Fig. 8. Average absorptivity (imaginary part of refractive index) distribution of ambient particles at Gilice sqare.

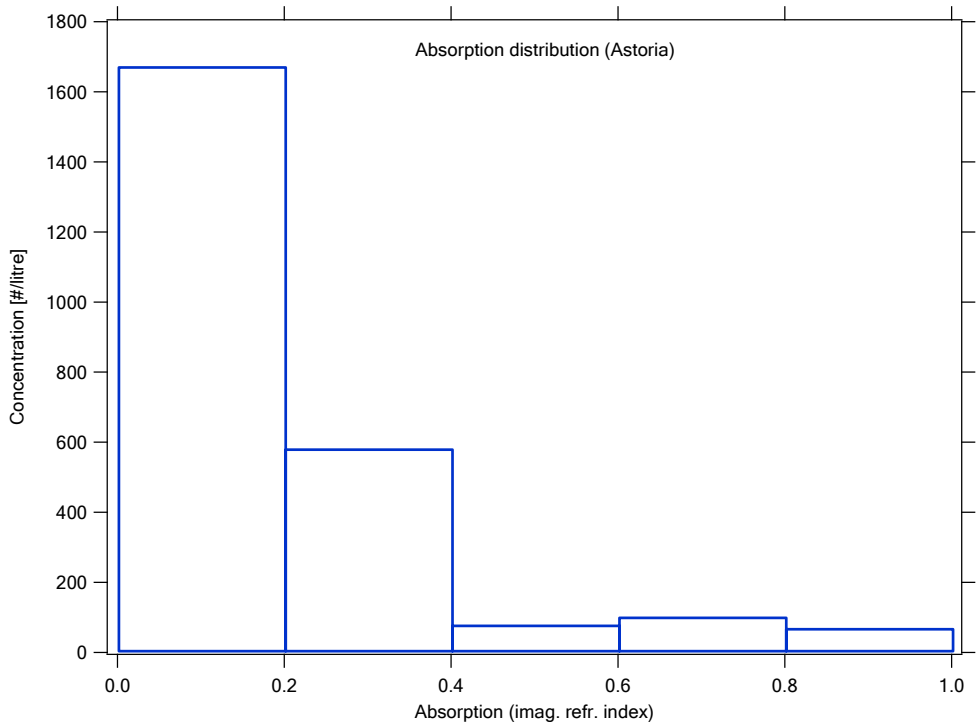

Fig. 9. Average absorptivity (imaginary part of refractive index) distribution of ambient particles at Astoria.

CONCLUSIONS

Measurements of ambient urban aerosols were conducted by simultaneously using optical particle spectrometry based on elastic scattering (DWOPS and OPC Mod.1.109) as well as acoustic spectrometry (PAS) and filter deposition technique (Aethalometer). Of all methods used in this study, only the DWOPS technique allows the direct assessment of the complex refractive index as well as the corresponding particle size. The imaginary part of the refractive index reflects the absorbing properties of particles in question.

The fact that DWOPS determines the size and the complex refractive index of each detected particle allows inferring the number concentrations of absorbing ambient aerosols as a function of their absorptivity. This is a unique capability of this instrument which is to our best knowledge is not achievable with any other real-time measuring technique practically suited for field measurements.

This novel capability opens new possibilities for a more comprehensive description of airborne particles and could contribute to defining a better set of reference materials for absorption. Consequently steps could be taken to standardize various methods used to determine the absorptivity of aerosols. It is a step in the right direction toward a better understanding of air pollution and its impact on the human and natural environment.

ACKNOWLEDGEMENTS

This work was supported by the Hungarian TéT_10-1-2011-0725 and the Austrian WTZ Hu12/2012 project.

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Received for review, May 22, 2015
Revised, September 14, 2015
Accepted, November 10, 2015