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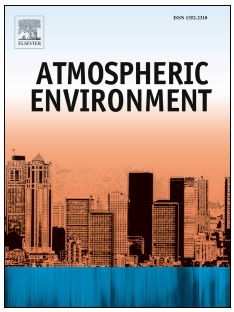

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A method for segregating the optical absorption properties and the mass concentration of winter time urban aerosol

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Abstract

A novel in-situ, real time method for the determination of inherent absorption properties of light absorbing carbonaceous particulate matter and its possible application for source apportionment are introduced here. The method is deduced from a two-week campaign under wintry urban conditions during which strong correlation was found between aerosol number size distribution and wavelength dependent optical absorption coefficient (*AOC(λ)*), measured by a Single Mobility Particle Sizer (SMPS) and a multi-wavelength photoacoustic absorption spectrometer, respectively, while wood burning and traffic (i.e. fossil fuel burning) activity were identified to be the dominant sources of carbonaceous particulate. Indeed, during the whole campaign, regardless of the actual emission strength of the aerosol sources, the measured number size distributions were always dominated by two unimodal modes with Count Mean Diameter (CMD) of 20 and 100 nm, which could be correlated to traffic and wood burning activities, respectively. AAE_{ff}, AAE_{wb} (i.e. the Aerosol Angström Exponent of traffic and wood burning aerosol, respectively), σ_{ff}*(266 nm)*, σ_{ff}*(1064 nm)*, σ_{wb}*(266 nm)* and σ_{ff}*(1064 nm)* (i.e. the segregated mass specific optical absorption coefficients at two of the measurement wavelengths) were found to be 1.17±0.18, 2.6±0.14, 7.3±0.3 m^2g^{-1}, 1.7±0.1 m^2g^{-1} 3.4±0.3 m^2g^{-1} and 0.31±0.08 m^2g^{-1}, respectively. Furthermore the introduced methodology can also disentangle and quantify the temporal variation of both the segregated optical absorptions and the segregated mass concentrations of traffic and wood burning aerosol. Accordingly, the contribution of wood burning to optical absorption of PM was found to be negligible at 1064 nm but increased gradually towards the

shorter wavelengths and became commensurable with the optical absorption of traffic at 266 nm during the whole measurement period. Furthermore, the contribution of wood burning mass to CM (mass of carbonaceous particulate matter) was dominant regardless of the strength of the emission activity of traffic and wood burning during the whole measurement period.

1. Introduction

The investigation of light absorbing carbonaceous particulate matter (LAC) is of utmost importance in the fields of atmospheric physics and chemistry. LAC is one of the most prominent drivers of global warming, while the uncertainties associated with radiative forcing calculations are also dominantly linked to its components (Penner et al., 2001; Schwartz, 2004; Lack et al., 2006; Solomon et al., 2007; Bond et al., 2013). On a regional scale, LAC is a major concern as an air pollutant, especially considering its adverse health effects and the public debate concerning its legal regulation.

It has been experimentally demonstrated in many recent studies, that LAC is dominantly composed of traffic and wood burning aerosol particularly under wintertime urban conditions, when the biological and photochemical activities are negligible (Sandradewi et al., 2008a, b; Favez et al., 2009). So far public policies have been committed to reducing the emission caused by traffic, industry and power plants while wood burning has been regarded as a renewable energy source with CO_2 neutral emission as an alternative for residential heating. Therefore, the relative amount of the wood burning aerosol fraction is expected to increase in the total mass of PM at an accelerating rate in the future.

Several methods have been introduced to quantitatively apportion aerosol fractions emitted by wood burning and traffic but most of them require time consuming and costly off-line chemical analysis. The recently developed semi continuous EC/OC ratio measurement based on the thermo-optical method using NIOSH 5040 protocol (Chow et al., 2001) or the real-time composition identification carried out by the aerosol mass spectrometer (AMS) are now also available for on-line source apportionment and open up novel possibilities in this field (Lanz et al., 2007; Kleinman et al., 2002). However, although they are extremely powerful tools for source apportionment, their widespread and regular application is limited mainly by instrument prices and laboratory costs.

As opposed to chemical features, the microphysical properties of airborne particles such as optical absorption, light scattering, and size distribution can be easily measured on-line, with high accuracy and sensitivity, especially under highly polluted urban conditions (Wehner and Wiedensohler, 2003; Kleinman et al., 2002; Scaire et al., 2008). Unfortunately in most of the cases there is a limited possibility of deducing chemical information from the measured microphysical data as most of them are not unique indicators of chemical composition (Utry et al., 2014). Nevertheless, some recent laboratory and field studies have demonstrated that although the optical absorption coefficient in itself does not, its wavelength dependency could correlate with chemical composition (Favez et al., 2009, 2010; Lewis et al., 2008; Chakrabatry et al., 2010; Flowers et al., 2010; Ajtai et al., 2011b). Therefore, the multi-wavelength measurement of optical absorption could yield a chemically

selective parameter in a real time source apportionment model (Moosmüller et al., 2011; Sandradewi et al., 2008a, b; Favez et al., 2010; Ajtai et al., 2011b).

Sandradewi et al. (2008) have been the first to propose a new method for the apportionment of wood burning and traffic aerosols based on the real time measurement of the multi-wavelength absorption spectra. Since all the early applications of this method have been based on transmission measurement on filter accumulated aerosol using a multi-wavelength Aethalometer, it is commonly called as the "Aethalometer model" in the literature. Although in some very recent studies both the applied method used for absorption measurement and the model itself have been criticized (Harrison et al., 2013), due to its simplicity and effectiveness this approach is likely to become more and more widely used. The original "Aethalometer model" is based on supplementary measurements of the chemical properties of the aerosol via filter sampling and off-line chemical analysis. However, due to the inherent problems associated with filter measurements, such as the limited sensitivity (which necessitates long time sampling) and the limited reliability of data interpretations (due to sampling and methodology artefact) as well as their off-line nature (Andreae and Gelencsér, 2006; Moosmüller et al., 2009; Schnaiter et al., 2005), various assumptions and simplifications are introduced into the Aethalometer model, which however further decrease its reliability.

In the present study we propose a novel type of source apportionment methodology based on in-situ, filter-free characterization of the ambient carbon fraction by simultaneous measurement of aerosol light absorption and size distribution using our state-of-the-art multi-wavelength photoacoustic spectrometer (4λ-PAS) and a Single Mobility Particle Sizer (SMPS), respectively. Measurements were carried out under wintertime ambient conditions, during which the sources of carbonaceous particulate matter were found to be traffic and wood burning practically exclusively.

2. Sampling site and instrumentation

2.1. Measurement site

The measurements reported here were carried out under wintertime urban ambient conditions near the city center of Szeged, Hungary (46.26°N, 20.14°E), from 12 to 26 of January, 2011. Szeged is among of the most populated cities of Hungary with more than 170000 inhabitants. According to the Hungarian Central Statistical Office (KSH) Szeged has the highest ratio of individual to district heating throughout the country, which is dominated by wood burning (Utry et al., 2013). Furthermore, during the campaign period, the city was also suffering from extremely high traffic activity, i.e. over 3000 trucks were passing through the city center daily, right next to the monitoring station. As a result of the high concentrations of typical wintertime urban carbonaceous particulate matter (traffic and residential heating aerosol) and the periodically varying ratio of these carbon constituents, this

measurement site was ideal to study the optical responses of typical wintertime urban ambience under continuously changing emission strengths of traffic and wood burning (Utry et al., 2013)

2.2. Instrumentations and sampling.

The absorption response of the ambient atmosphere was measured by our recently developed state-of-the-art multi-wavelength photoacoustic spectrometer (4λ-PAS). The filter free operation and the insensitivity to scattering, as well as the wide range of operational wavelengths from near-IR to UV provides absorption data with high reliability (Flowers et al., 2010; Utry et al., 2014). The principles of operation and the characteristic performance of this instrument both under laboratory and filed conditions are described in details elsewhere (Ajtai et al., 2010; 2011). The aerosol optical absorption coefficient (AOC) was determined at all operational wavelengths of the 4λ-PAS (266nm, 355nm, 532nm and 1064nm).The accuracy of this instrument was proved to be below 2-6% depending on the applied wavelengths due to the implemented wavelength independent gas-phase calibration (Ajtai et al., 2010).

Number and volume concentration as well as the size distribution of the atmospheric aerosol were measured by the Scanning Mobility Particle Sizer (SMPS, GRIMM system Aerosol Technik, Germany, type SMPS+C) and by the Optical Particle Counter (OPC, GRIMM, Aerosol Technik, Germany, type 1.108), respectively in the size range from 5 nm to 32 µm. SMPS consists of the Condensation Particle Counter (CPC Model #5.400) and the Classifier "Vienna"-Type Differential Mobility Analyzer (DMA, Model #5.500). The "short" DMA was used to measure the fine fraction of ambient particulate matter from 5 nm to 350 nm. First, the DMA separates particles in the equally charged aerosol stream according to their electrical mobility. Following that the separated particles are lead to the CPC unit where the size-segregated number density is measured. The shielding effect, which occurs when high concentrations of particles get into the DMA, is minimized by using the shelf-implemented coincidence correction protocol. The Optical Particle Counter (OPC, GRIMM Aerosol Technic, Germany, type 1.108) was used to measure the size distribution in the coarse fraction from 0.3nm to 32µm size range according to the light scattering intensity of the aerosol stream.

3. The apportionment method

3.1. The assumptions behind the extrapolation method

The applicability of the above described extrapolation method (and the validity of Equation 3-6) depends on whether the following assumptions are fulfilled throughout the studied measurement period, independently of the actual intensity of the traffic and wood burning sources:

- Optical absorption coefficient *(AOC(λ))* can be written as the sum of the optical absorption coefficient of wood burning aerosol *($AOC_{ff}(\lambda)$)* and the optical absorption coefficient of traffic aerosol *($AOC_{wb}(\lambda)$)* at any of the used measurement wavelengths (see Equation 3 and 4).
- Both the traffic and the wood burning aerosol fraction can be characterized with a well distinguishable characteristic unimodal mode in the number size distribution and the concentration of light absorbing carbonaceous aerosol can be written with a good accuracy as the sum of wood burning (N_{wb}) and traffic (N_{ff}) aerosol number concentration within these characteristic modes. This and the previous assumption state the dominance of these two aerosol types in the light absorbing fraction of the atmospheric aerosol.
- The wood burning and the traffic aerosol can be characterized unambiguously by single particle optical absorption coefficients ($SPAOC_{wb}(\lambda)$ and (SP$AOC_{ff}(\lambda)$, respectively) at any of the used measurement wavelengths. The applicability of Equation 5 and 6 depends on this and the previous assumptions.

The first two assumptions will be discussed in details in 3.2.1. The presented results prove that the third assumption is valid at least during the period of this relatively short campaign. Its long term validity will be a subject of further field campaigns.

3.2. The extrapolation method for the determination of the segregated AAEs

The introduced method consists of four subsequent steps (see below) and it is based on the following Equations:

$$\frac{AOC_{ff}(\lambda_1)}{AOC_{ff}(\lambda_2)} = \left[\frac{\lambda_1}{\lambda_2}\right]^{-AAE_{ff}} \quad (1)$$

$$\frac{AOC_{wb}(\lambda_1)}{AOC_{wb}(\lambda_2)} = \left[\frac{\lambda_1}{\lambda_2}\right]^{-AAE_{wb}} \quad (2)$$

$$AOC(\lambda_1) = AOC_{ff}(\lambda_1) + AOC_{wb}(\lambda_1) \quad (3)$$

$$AOC(\lambda_2) = AOC_{ff}(\lambda_2) + AOC_{wb}(\lambda_2) \quad (4)$$

$$AOC(\lambda_1) = N_{ff} \times SPAOC_{ff}(\lambda_1) + N_{wb} \times SPAOC_{wb}(\lambda_1) \quad (5)$$

$$AOC(\lambda_2) = N_{ff} \times SPAOC_{ff}(\lambda_2) + N_{wb} \times SPAOC_{wb}(\lambda_2) \quad (6)$$

$$CM(PM_1) = \sigma_{ff}(\lambda_1)^{-1} \times AOC_{ff}(\lambda_1) + \sigma_{wb}(\lambda_1)^{-1} \times AOC_{wb}(\lambda_1) \quad (7)$$

$$CM(PM_1) = \sigma_{ff}(\lambda_2)^{-1} \times AOC_{ff}(\lambda_2) + \sigma_{wb}(\lambda_2)^{-1} \times AOC_{wb}(\lambda_2) \quad (8)$$

$$CM_{wb}(PM1) = AOC_{wb}(\lambda) \times \sigma_{wb}(\lambda) \quad (9)$$

$$CM_{ff}(PM1) = AOC_{ff}(\lambda) \times \sigma_{ff}(\lambda) \quad (10),$$

where λ is wavelength, $AOC_{ff}(\lambda)$ and $AOC_{wb}(\lambda)$ are the optical absorption coefficient of fossil fuel and wood burning aerosol at the given wavelength, $AOC(\lambda)$is optical absorption coefficient measured at the given wavelength, N_{ff} and N_{wb} are total number concentration of fossil fuel and wood burning aerosol, $SPAOC_{ff}(\lambda)$ and $SPAOC_{wb}(\lambda)$ are single particle optical absorption coefficient of fossil fuel and wood burning aerosol at the given wavelength, $CM(PM_1)$ is mass concentration of carbonaceous particulate matter, PM1, $\sigma_{ff}(\lambda)$ and $\sigma_{wb}(\lambda)$ are mass specific optical absorption coefficient of fossil fuel and wood burning aerosol at the given wavelength, while $CM_{ff}(PM1)$ and $CM_{wb}(PM1)$ is mass concentration of fossil fuel and wood burning aerosol respectively.

1. As a first step the segregated Absorption Angstrom Exponents (defined by Eq. 1 and 2) of traffic and wood burning aerosol (AAE_{ff} and AAE_{wb}, respectively) is determined with the help of an extrapolation method which is based on Eq. 3-6 and described in details in this section. The applicability of the extrapolation method depends on the validity of a few assumptions listed in this section which are verified in 3.2.1.
2. In the next step the temporal variations of the segregated aerosol optical absorption coefficients (AOC) for traffic and wood burning components ($AOC_{ff}(\lambda)$ and $AOC_{wb}(\lambda)$, respectively) throughout the campaign are calculated with the help of Eq. 1-4. Indeed once the AAE values are known from the previous step, these four equations only contain four unknown quantities, i.e. for each measurement point they can be solved algebraically.
3. Segregated mass specific optical absorption coefficients of traffic and wood burning aerosol ($\sigma_{ff}(\lambda)$ and $\sigma_{wb}(\lambda)$, respectively) is determined with the help of Eq. 7 and 8, where $CM(PM_1)$ means the mass concentration of sub-micron sized carbonaceous particulate matter measured by an independent method. See section 3.3 for details.
4. Finally by dividing the segregated AOC values with the corresponding σ values the temporal variation of the segregated mass concentrations can be determined as given in Eq. 9 and 10.

The ultimate operational wavelengths (266 and 1064 nm) were selected here to demonstrate the applicability of the introduced method, because the contribution of the traffic and wood burning aerosol particles to the optical absorption was expected to be the most divers at these wavelengths, resulting in higher apportionment selectivity. By writing the two actual measurement wavelengths

(266 nm and 1064 nm) into Eq. 5 and Eq. 6, respectively and dividing Eq. 5 by Eq. 6, one gets the following equation:

$$\frac{AOC(266nm)}{AOC(1064nm)} = \frac{SPAOC_{ff}(266nm)+\frac{N_{100}}{N_{20}}\cdot SPAOC_{wb}(266nm)}{SPAOC_{ff}(1064nm)+\frac{N_{100}}{N_{20}}\cdot SPAOC_{wb}(1064nm)} \quad (9)$$

N_{100} and N_{20} stands for the number concentration of particle associated to the identified modes with count median diameter of 100 nm and 20 nm, respectively.

Figure 1 shows the ratio of the measured AOCs as a function of N_{100}/N_{20}. In case N_{100}/N_{20} converges to either 0 or infinity, Eq. 9 can be rewritten as:

$$\lim_{\frac{N_{100}}{N_{20}}\to 0} \frac{AOC(266nm)}{AOC(1064nm)} = \frac{SPAOC_{wb}(266nm)}{SPAOC_{wb}(1064nm)} = \left(\frac{266}{1064}\right)^{-AAE_{ff}} \quad (10)$$

$$\lim_{\frac{N_{100}}{N_{20}}\to \infty} \frac{AOC(266nm)}{AOC(1064nm)} = \frac{SPAOC_{ff}(266nm)}{SPAOC_{ff}(1064nm)} = \left(\frac{266}{1064}\right)^{-AAE_{wb}} \quad (11)$$

i.e. in these limits, which correspond to cases when the light absorption is totally dominated either by traffic or wood burning aerosol, respectively, the segregated AAE values can be determined. The segregated AAE values can be determined by fitting data points shown on Figure 1 the function shown on Fig. 1. and calculating the asymptotic value of the fitted curve (see section 4 for results).

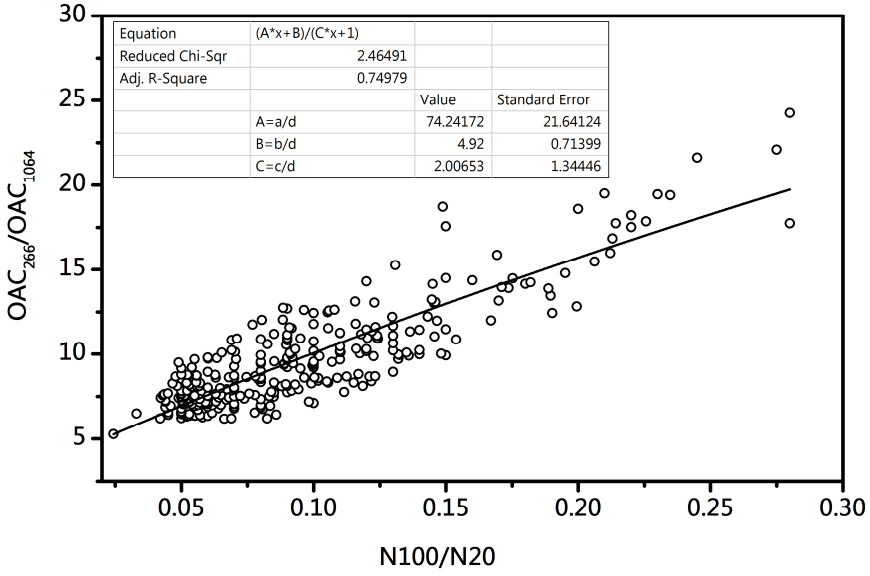


Figure 1. Measured optical absorption coefficient ratios as a function of the ratio of the number concentrations in the two aerosol modes (see text for details)

3.2.1. Segregated number size distributions

Throughout the reported measurement campaign two characteristic mode with a Count Median Diameter of 20 nm (CMD_{20}) and 100 nm (CMD_{100}), respectively could always be identified in the measured number size distributions. Their relative weight varied in time but nevertheless almost all measured particles were found to belong to either mode. In our previous study (Utry et al., 2014) the CMD_{20} and CMD_{100} modes were attributed to traffic and residential wood burning for household heating, respectively. This assignment is justified both by the results of off-line chemical analysis and by the daily variation of the ratio of segregated number size concentrations in the CMD_{100} and CMD_{20} modes (i.e. N_{ff}/N_{wb}) which reached its maximum at 18:00 and at around 8:00 (i.e. during rush hours), while having a local and an absolute minima at around 12:00 and 3:00, respectively. The average daily variation (taking into account only working days, i.e. excluding week-ends) of N_{ff} and N_{wb} are shown in Figure 2. Furthermore, we also found that the two modes ad up to the original size distribution during any period of the day with good accuracy (Figure 3). These findings are in line with the widely adopted assumption that under typical wintry urban conditions whenever individual heating (i.e. wood burning) dominates, the fine fraction of ambient aerosol (<PM1) does almost exclusively include carbonaceous particulate matter from traffic (CMD20) and from residential heating (CMD100) (Schneider et al., 2005; Bond et al., 2002; Wehner and Wiedensohler, 2003; Rissle et al., 2006). Based on the presented results the validity of Eq. 5 and 6 can be indeed assumed. However it should be noted that the separation of the size distribution into two modes is possible only if number representation is used while e.g. the volumetric representation cannot be applied as explained elsewhere (Schneider et al., 2005; Hedberg et al., 2002).

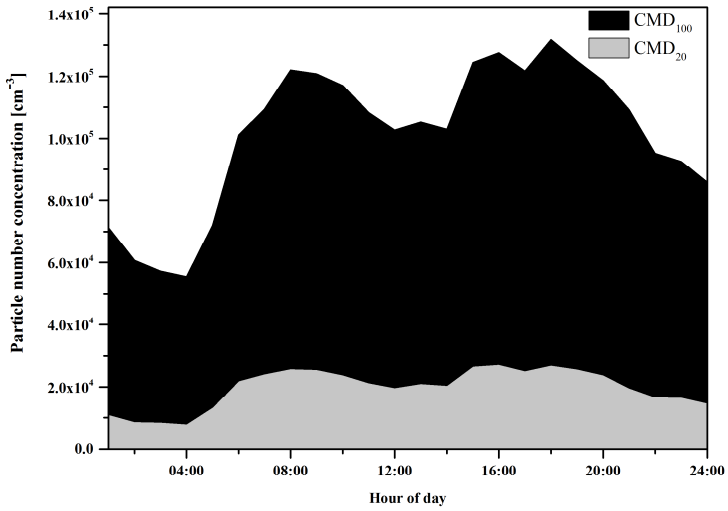
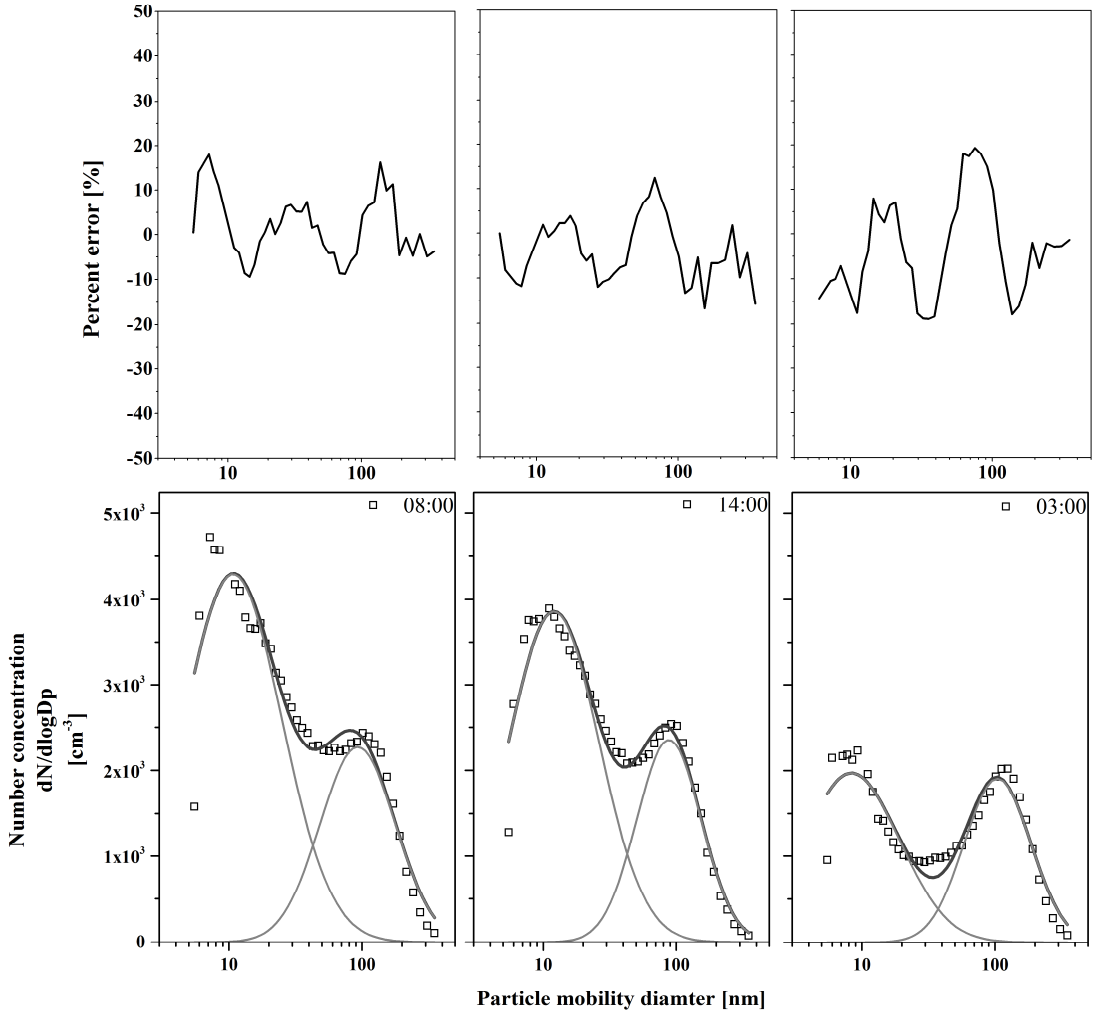


Figure 2. The daily variation of the segregated number-concentration in the CMD=20nm and CMD=100nm modes.



243

244 *Figure 3. The lower panel contains the segregated (unimodal) number-size distributions (grey lines),*
245 *the sum of these modes (black lines) and the originally measured number size distributions (dots) at*
246 *three characteristic time of the day. The upper panel contains the percent errors between the*
247 *measured and the calculated (from the segregated) number size distributions.*

248

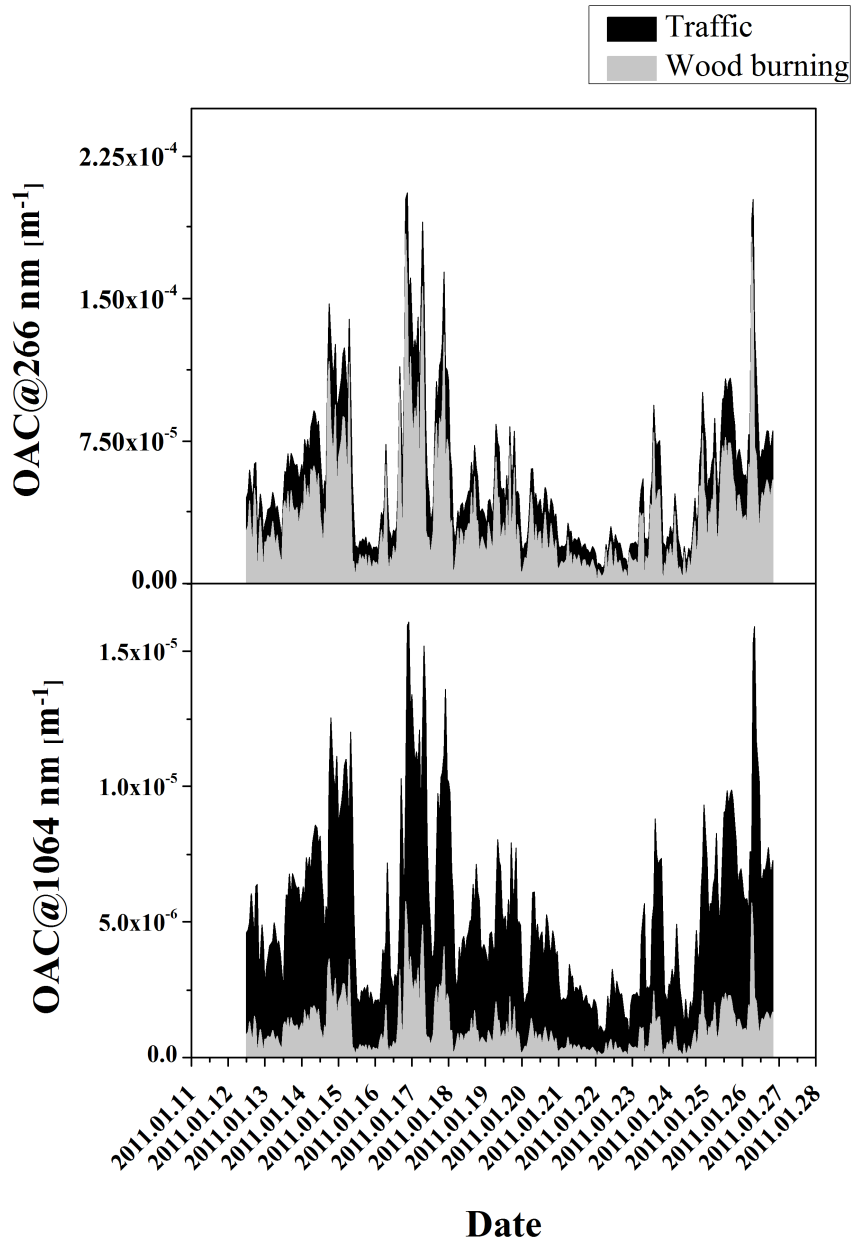
249 *3.3 The determination of the segregated mass specific optical absorption coefficients*

250 The extrapolation method discussed above yields valuable information on the segregated
251 optical properties of the aerosol ambience. The segregated mass concentrations can also be calculated
252 by using Eq. 7 and 8 and the measured AOC values whenever the temporal variation of the total mass
253 concentration in the PM1 mode (CM(PM1)) is available. In case of the measurement campaign
254 analyzed in this publication, the mass concentration was calculated from the volume-concentration
255 measured by the SMPS assuming an average carbonaceous particle density of 1.5µg/m^3 which is one
256 of the most cited and accepted average values of ambient carbonaceous particulate in the literature
257 (McCurry et al., 2010; Turpin and Lim, 2001). Unfortunately as the particle density of traffic and
258 wood burning aerosol varies in a wide range between 1.3-2.2 µg/m^3 and depends on the operation

conditions of combustion as well as the type of fuel (Burtscher 2004; Park et al., 2004; Dinar et al., 2006), one can question the reliability of the calculated segregated mass concentrations. On the other hand in future applications of the apportionment method one can always use e.g. a TEOM or a beta attenuation monitor possibly combined with a PM1 impactor for the independent determination of CM(PM1) so this limitation should not apply in general.

4. Results

By using the extrapolation method described in details in section 3.2.1, AAE_{ff} and AAE_{wb} found to be 1.17±0.18 and 2.6±0.14, respectively. The correlation coefficient and the standard deviation of the fitting were found to be 0.74 and 0.05 respectively. The time series of one hour averaged data of AOC_{ff} and AOC_{wb} determined at 266 and 1064nm wavelengths are plotted in Figure 4. The mass specific absorption coefficient of traffic and wood burning aerosol was calculated to be σ_{ff}(266nm) = 7.3 ± 0.4 m^2/g, σ_{ff}(1064nm) = 1.7 ± 0.23 m^2/g and σ_{wb}(266nm)= 3.4 ± 0.18 m^2/g, and σ_{wb}(1064nm) = 0.31 ± 0.1 m^2/g using Eq. 7 and 8. The calculated temporal variation of the mass concentration of traffic and wood burning aerosol are plotted in Fig.5.



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275 *Figure 4. Temporal variation of the segregated AOC values at the two wavelengths of the 4λ-*
276 *PAS instrument*

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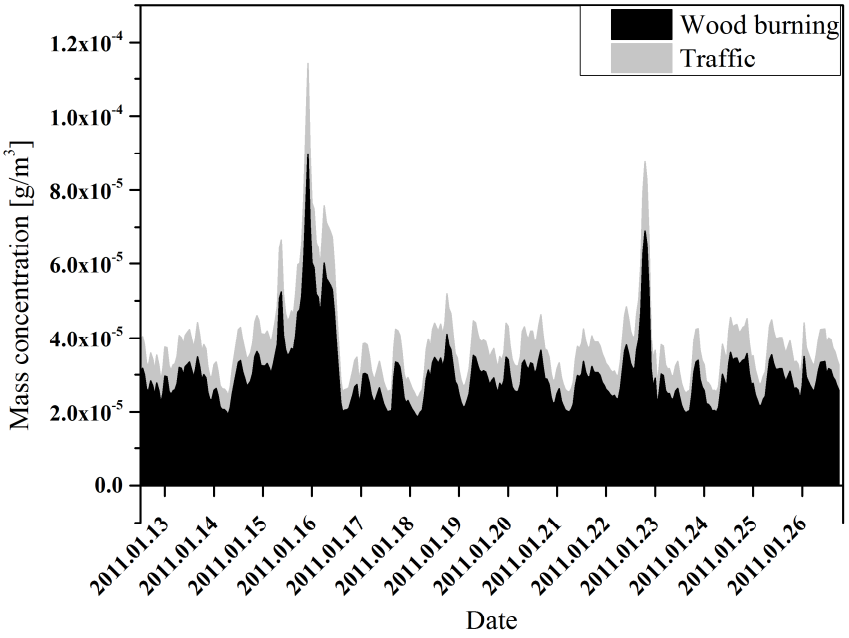

Figure. 5. Segregated mass concentration of wood burning and traffic aerosol determined by the introduced method

5. Discussion

The correlation coefficients and the standard deviation of the fitting parameters of the extrapolation method for the determination of the segregated AAE values indicate strong and reliable relationship between the measured parameters. The deduced AAE_{ff} value is very similar to both theoretical predictions (AAE_{ff} is expected to be approximately 1) from the literature which are based on the corpuscular approximation and also with results from other experiments made both under laboratory and field conditions (Lan et al., 2013; Russel et al., 2013; Favez et al., 2009). The calculated AAE_{wb} value is also analogous with AAE_{wb} values determined during other field studies (Sandradewi et al., 2008b; Lewis et al., 2008; Moosmüller et al., 2011). Indeed AAE_{wb} is known to vary in a wide range from 1.3 up to around 3 while it also depends on the type of wood and the condition of combustion (Bond, 2001; Kirchstetter et al., 2004; Schnaiter et al., 2003).

In the extrapolation method only two out of the four available wavelengths of the 4λ-PAS instrument is used. Indeed it can be seen (Fig. 4) that the temporally varying AOC is dominated by AOC_{ff} and AAE_{wb} in the near-infrared and in the UV, respectively. These findings can be well explained by the inherent optical properties of these aerosols, since wood smoke is likely to contain high abundant organic compositions such as HULIS (Humic-Like Substances), PAH (Polycyclic Aromatic Hydrocarbons) which has gradually increasing light absorption towards the shorter

wavelengths. Furthermore both the dynamical variation and the relative magnitude of the segregated AOC values are in good agreement with the results of former field experiments made using the Aethalometer model (Sandradewi et al., 2008a; Favez et al., 2010; Castanho and Artaxo 2001). On the other hand it is noteworthy that AOC_{wb} is actually not negligible at 1064nm but its averaged relative abundance in the total AOC at this wavelength was more than 20% throughout the campaign. Based on the deduced segregated AAE values the segregation of the AOC was made at the other two wavelengths too and the average relative abundance of AOC_{wb} in the total AOC was found to increase gradually toward the shorter wavelengths: 20.5%, 45.1%, 65.8% and 82.2% at 1064nm, 532nm, 355nm and 266nm respectively.

The deduced segregated mass specific optical absorption values are highly plausible and are in good agreement with most of the similar data previously published in literature (Moosmüller et al., 2009; Petzold et al., 2004). Furthermore AAEff and AAEwb values can also be derived from these values substituting sigmas in AOC in eq. 3 and 4. They were found to be 1.1±0.2 and 2.62±0.4 respectively, which are in a reasonable agreement with values deduced by the extrapolation method, which further confirms the reliability of this approach.

It is important to emphasize that neither the AAE_{wb} nor the σ_{wb} values are representative to a specific wood type or burning condition but rather they correspond to a mixture of wood types and burning conditions as well as to ambient conditions i.e. most probably they can be used only for the analyzed measurement campaign under the actual local circumstances. The same applies for the traffic related segregated parameters, i.e. they also only represent local averages.

Finally as far as the segregated mass concentrations are concerned the calculated results show that, during the whole campaign, wood burning aerosol was dominant while traffic aerosol played only inferior role in CM(PM1). These findings are in line with other field experiments when the apportionment of wood burning emission was determined by Aethalometer model in urban or rural sites under wintry condition where dominantly wood burning contributed to residential heating. Furthermore, the average relative strength of wood burning to traffic aerosol was found to be around 75% and the average cumulative mass of carbonaceous aerosol (wood burning plus traffic) about 50% of the total PM2.5 (measured nearby in the official monitoring station of Hungarian Air Quality Network (HAQN)) which are also very typical under such conditions (Flowers et al., 2010; Sandradewi et al., 2008a; Bressi et al., 2014; Fuller et al., 2014)

6. Summary and conclusion

A carbonaceous particulate selective source apportionment study was performed for ambient particulate matter in the city center of Szeged, Hungary where the dominance of traffic and wood burning aerosol has been experimentally demonstrated earlier (Utry et al., 2014). The proposed model is based on the parallel, in-situ measurement of optical absorption and size distribution. AAE_{ff} and

AAE_{wb} were deduced from the measured data using the defined correlation between the AOC(1064nm)/AOC(266nm) and N_{100}/N_{20} ratios. $\sigma_{ff}(\lambda)$ and $\sigma_{wb}(\lambda)$ were determined with the help of the independently measured temporal mass concentrations in the PM1 mode. Furthermore, the proposed optical source apportionment is based on the assumption that the light absorbing fraction of PM is exclusively related to traffic and wood burning. This assumption is indirectly confirmed here by the fact that the measured size distribution is composed of two unimodal size distributions identified to correspond to traffic and wood burning aerosols. The method offers the possibility of replacing laborious chemical analysis with simple in-situ measurement of aerosol size distribution data. The results by the proposed novel optical absorption based source apportionment method prove its applicability whenever measurements are performed at an urban site where traffic and wood burning are the dominant carbonaceous sources of emission. Obviously, the effects caused by the activities of biogenic compounds or photochemistry as well as the changes in meteorological condition on the results of the extrapolation method should be further investigated in the future.

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- We introduce a new method for the real time source apportionment of atmospheric aerosol
- The method is deduced from a field measurement campaign
- The method requires the measurement of size distribution and optical absorption
- The applied instruments are a SMPS and a multi-wavelength photoacoustic absorption spectrometer