Atmospheric Environment 164 (2017) 1-7

ELSEVIER

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv



Technical note: Aerosol light absorption measurements with a carbon analyser – Calibration and precision estimates



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HIGHLIGHTS

• On the application of a carbon analyser as absorption photometer.

• Measuring the mass absorption efficiency with a single instrument.

• Determination of the precision of mass absorption efficiency.

ARTICLE INFO

Article history: Received 16 September 2016 Received in revised form 10 May 2017 Accepted 16 May 2017 Available online 25 May 2017

Keywords: Black carbon Elemental carbon Aerosol light absorption Mass absorption efficiency

ABSTRACT

Equivalent Black Carbon (EBC) and Elemental Carbon (EC) are different mass metrics to quantify the amount of combustion aerosol. Both metrics have their own measurement technique. In state-of-the-art carbon analysers, optical measurements are used to correct for organic carbon that is not evolving because of pyrolysis. These optical measurements are sometimes used to apply the technique of absorption photometers. Here, we use the transmission measurements of our carbon analyser for simultaneous determination of the elemental carbon concentration and the absorption coefficient. We use MAAP data from the CESAR observatory, the Netherlands, to correct for aerosol-filter interactions by linking the attenuation coefficient from the carbon analyser to the absorption coefficient measured by the MAAP. Application of the calibration to an independent data set of MAAP and OC/EC observations for the same location shows that the calibration is applicable to other observation periods. Because of simultaneous measurements of light absorption properties of the aerosol and elemental carbon, variation in the mass absorption efficiency (MAE) can be studied. We further show that the absorption coefficients and MAE in this set-up are determined within a precision of 10% and 12%, respectively. The precisions could be improved to 4% and 8% when the light transmission signal in the carbon analyser is very stable.

1. Introduction

Black carbon is a qualitative description when referring to light absorbing carbonaceous substances in atmospheric aerosol. Black carbon can be related to some measurable properties that together with the suitable method define the terminology (Bond et al., 2013; Petzold et al., 2013). The most commonly measured properties are elemental carbon (EC), a mass metric, and the absorption coefficient, determined by light absorption. The absorption coefficient is commonly converted into a mass metric by application of a massspecific absorption cross section (MAC; referred to as mass

* Corresponding author. E-mail address: bas.henzing@tno.nl (J.S. Henzing). absorption efficiency, MAE hereafter). The mass metric based on light absorption measurement is to be referred to as equivalent black carbon (EBC).

Elemental carbon (EC) and organic carbon (OC) are quantified by thermal-optical analysis. In the analyser, e.g. the Sunset Laboratory Inc. Thermal Optical Carbon Aerosol Analyser (Sunset Laboratory Inc, Tigard, USA) and the DRI Thermal Optical Carbon Analyser (Atmoslytic, Inc., Calabasas, USA), a filter punch is heated in an oven by a thermal programme, e.g. EUSAAR2 (Cavalli et al., 2010), NIOSH890 (Peterson and Richards, 2002) or IMPROVE_A (Chow et al., 2007). The separation between OC and EC is based on the thermal and chemical stability of the aerosol. During heating some particulate OC and absorbed organic vapours are converted to EC through pyrolysis in a pure helium atmosphere. Reflected or transmitted laser light is used to monitor and correct for this so called pyrolytic carbon (PC) (Turpin et al., 1990).

In monitoring networks, the absorption coefficient is generally measured with absorption photometers, e.g. the Aethalometer (AE; Magee Scientific, Berkeley, USA; Hansen et al., 1984), the Particle Soot Absorption Photometer (PSAP; Radiance Research, Seattle, USA) and the Multi-Angle Absorption Photometer (MAAP; Thermo Fisher Scientific, Waltham, USA: Petzold and Schönlinner, 2004). Absorption photometers continuously measure the attenuation of light by the aerosol and filter. To finally obtain the absorption coefficient of the aerosol in ambient air, corrections need to be performed to the attenuation coefficient, to account for aerosol-filter interactions. Different correction algorithms are used for Aethalometer (e.g. Weingartner et al., 2003; Arnott et al., 2005; Collaud Coen et al., 2010) and PSAP (Bond et al., 1999; Virkkula et al., 2005; Müller et al., 2014). The MAAP measures, besides the transmission, also the reflection at two different angles. The measured reflection and transmission signals are used in a radiative transfer model to calculate the absorption caused by particles (Petzold and Schönlinner, 2004).

Ram and Sarin (2009) introduced the first method exploiting the transmission measurements of a thermal-optical analyser to infer the light absorption coefficient. They used a correction used by Weingartner et al. (2003) and Bond et al. (1999) to correct for a multiple scattering enhancement and a loading effect in the Aethalometer. The multiple scattering enhancement and the loading effect are parametrised by the factors C and R(ATN), respectively. The absorption coefficient is calculated by

$$b_{\rm abs} = \frac{b_{\rm ATN}}{C \cdot R(\rm ATN)} \tag{1}$$

Ram and Sarin (2009) used $C = 2.14\pm0.21$ and the function for R(ATN) as found by Weingartner et al. (2003), although they acknowledged that the value for C may not be optimal, because this value is determined for fresh diesel aerosol. Later, Andersson et al. (2011) used $C = 3.6\pm0.6$, which is determined for internally mixed aerosol. Collaud Coen et al. (2010) showed that the value of C is site specific. Therefore, we chose not to use one of these values for C. We chose to use a MAAP as reference instrument, because these instruments are widely available at monitoring stations. Furthermore, the MAAP has a low unit-to-unit variability of 3% (Müller et al., 2011), which is sufficient for the method and means that the calibrations can be performed by different instruments with a high reproducibility. An in-depth review of light absorption measurement methods is provided by Moosmüller et al. (2009).

The advantage of using the carbon analyser as absorption photometer is that both elemental carbon concentration and absorption coefficient can be determined by a single instrument. This enables the calculation of the mass absorption efficiency of EC (MAE, in $m^2 g^{-1}$) that is crucial for aerosol climate impact studies. The mass absorption efficiency is calculated by

$$MAE = \frac{b_{abs}}{EC}$$
(2)

Section 2 describes the used method and the calibration we added to the method of Ram and Sarin (2009). In Sect. 3, we calibrate the attenuation coefficient data inferred from the carbon analyser from a field campaign at the CESAR observatory near Cabauw, the Netherlands, performed in September and October 2014 and used an independent data set from June and July 2010 from the same site to validate the calibration. Finally, the results are discussed in Sect. 4.

2. Method

2.1. Attenuation coefficient

The optical attenuation of light (ATN) is defined as

$$ATN \equiv \ln\left(\frac{I_0}{I}\right) \tag{3}$$

where I_0 is the intensity of the incident laser beam and I is the intensity of the light passing through the filter.

With a carbon analyser, the attenuation cannot be determined directly, because the initial laser intensity I_0 is unknown. However, the difference in attenuation Δ ATN, which is linked to the absorption, can be measured and is given as

$$\Delta \text{ATN} = \ln\left(\frac{I_0}{I_{T_d}}\right) - \ln\left(\frac{\eta I_0}{I_{T_w}}\right) = \ln\left(\frac{I_{T_w}}{I_{T_d}}\right) - \ln\left(\frac{\eta I_0}{I_0}\right) \simeq \ln\left(\frac{I_{T_w}}{I_{T_d}}\right)$$
(4)

where I_{T_w} is the intensity of the transmitted light passing through a white filter, I_{T_d} is the intensity of the transmitted light through a laden, and thus dark, filter and η the variation of the incident beam in time. η is assumed to be close to 1 during the analysis, so the second logarithm, $\ln(\eta I_0/I_0)$, is negligible. Because the carbon analyser measures the intensity of the transmitted light through the white and laden filter, the difference in attenuation is a measurable quantity.

The first step in obtaining Δ ATN is performing an OC/EC analysis with a carbon analyser according to an analysis protocol. After the measurement, the OC and EC mass concentrations can be calculated with the calculating software of the manufacturer of the instrument.

We obtained I_{T_d} by averaging the transmission signal over the first 60 s of the analysis, in this 60 s the filter is still dark and pyrolysis has not occurred yet. I_{T_w} is obtained by a measurement of the transmission signal for a white filter. This can either be at the end of the analysis protocol when all carbon has evolved from the filter or a blank filter from the same batch.

The measured difference in attenuation is expressed in the attenuation coefficient $b_{\text{ATN,Sunset}}$ (with the units of m⁻¹):

$$b_{\text{ATN,Sunset}} = \frac{A}{Q} \frac{\Delta ATN}{\Delta t} = \frac{A}{V} \ln \left(\frac{I_{T_w}}{I_{T_d}} \right)$$
(5)

Where *A* is the filter area (in m²), *Q* is the volumetric flow rate (in m³ s⁻¹) and Δt is the sampling time (in s). In the last step, the total sampled volume $V \equiv Q\Delta t$ (in m³) is included. The factor A/V now represents the inverted column of air that is put onto the filter.

2.2. Calibration to absorption photometer

The measured attenuation differs from the absorption of the aerosols in air. This is caused by a multiple scattering enhancement (Liousse et al., 1993; Ballach et al., 2001) and a loading effect, which is sometimes also referred to as 'shadowing effect' (Reid et al., 1998). To correct for these artefacts, a correction function $C \cdot R(\Delta ATN)$ is introduced by e.g. Weingartner et al. (2003), where *C* parametrises the multiple scattering enhancement and $R(\Delta ATN)$ parametrises the loading effect. There are more recent and more complex corrections available (Collaud Coen et al., 2010; Müller et al., 2014). However, the correction function proposed by Weingartner is simple and the more recent corrections perform only slightly better, but add more complexity.

In the calibration, the attenuation coefficient from the carbon

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