Atmospheric Environment 129 (2016) 18-26

Contents lists available at ScienceDirect

Atmospheric Environment

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

Multiyear in-situ measurements of atmospheric aerosol absorption properties at an urban coastal site in western Mediterranean



ATMOSPHERIC

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HIGHLIGHTS

- In-situ aerosol absorption properties are measured during 4 years in an urban site.
- Seasonal differences are considered for the Aethalometer calibration parameters.
- Calibration parameters are obtained for the 7 different aethalometer wavelengths.
- Aerosol absorption coefficients and absorption Ångström Exponent are analysed.
- Mixing layer height and traffic emissions have been considered in this study.

ARTICLE INFO

Article history: Received 15 October 2015 Received in revised form 18 December 2015 Accepted 5 January 2016 Available online 13 January 2016

Keywords: Atmospheric aerosols Aethalometer Absorption coefficient Absorption Ångström exponent In-situ measurements

ABSTRACT

In-situ aerosol absorption properties measured in Valencia (Spain) for four years, from February 2011 to February 2015, have been analysed. Spectral absorption properties have been obtained using a sevenwavelength Aethalometer AE-31 which covers the range from UV (370 nm) to IR (950 nm). In order to obtain the absorption coefficients, compensation parameters have been calculated for the Aethalometer considering seasonal and spectral differences. For this multiyear measurement period, seasonal sitespecific calibration parameters have been obtained. Furthermore, estimations of the absorption Ångström Exponent (α_{abs}) have been calculated using the seven Aethalometer wavelengths. The averaged absorption coefficients (plus/minus the standard deviation) obtained for the seven channels range between 9 \pm 4 Mm⁻¹ at 950 nm and 33 \pm 18 Mm⁻¹ at 370 nm. These results are typical of a moderate polluted environment. The obtained absorption Ångström Exponent (plus/minus the standard deviation) is 1.42 ± 0.08 , which suggests the presence of brown carbon or black carbon coated by non-absorbing particles in our site. Seasonal and daily variations, together with the effect of both the boundary layer height and traffic, have been also studied. Strong seasonal differences in the absorption coefficient are found, mainly due to seasonal variation of the mixing layer height. On the opposite, the study of the diurnal variations of the absorption Ångström Exponent proves that this parameter is more affected by traffic emissions than by the evolution of the mixing layer height.

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1. Introduction

Atmospheric aerosols, due to their high spatial and temporal variability, are one of the major sources of uncertainty in different processes affecting climate (IPCC, 2013). Among their effects over climate, their role in the radiative forcing in the Earth's atmosphere is highly important (Charlson et al., 1992; Alados-Arboledas et al.,

http://dx.doi.org/10.1016/j.atmosenv.2016.01.008 1352-2310/© 2016 Elsevier Ltd. All rights reserved. 2003). On one hand, they have a direct effect by scattering and/or absorbing earthbound solar radiation, which contributes to the cooling and warming of the atmosphere. On the other hand, they have an indirect effect acting as cloud condensation nuclei (CCN) (Ramanathan et al., 2001), modifying the microphysical and radiative characteristics of clouds (Kaufman et al., 2005). By absorbing light, aerosols contribute to the heating of the atmosphere, and thus they introduce a positive radiative forcing (Johnson et al., 2004). Therefore, absorption is an important component of the radiation budget and it needs to be better characterized to reduce uncertainties in predictions performed by climate models



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(Ramanathan et al., 2001).

Different methods have been used to measure light absorption based on filter (Hansen et al., 1984; Müller et al., 2011) or photoacoustic techniques (Moosmüller et al., 2009). The most widely used method for in-situ measurements is the filter-based technique, which obtains the aerosol absorption coefficient by measuring the change of light attenuation due to aerosol loading on a filter matrix (Hansen et al., 1982). Instruments based on this technique are the Aethalometer (Hansen et al., 1984), the Particle Soot Absorption Photometer (PSAP) (Bond et al., 1999), the Multi-Angle Absorption Photometer (MAAP) (Petzold and Schönlinner, 2004), and the Continuous Soot-Monitoring System (COSMOS) (Miyazaki et al., 2008).

Being able to obtain the aerosol absorption coefficient at different wavelengths is important as the spectral dependence of absorption is related to aerosol physical and chemical properties. The major light absorbing aerosol components are carbonaceous substances and mineral dust (Rizzo et al., 2011). In urban sites, carbonaceous particles known as black carbon (BC) are the main responsible for the absorption of solar radiation (Jacobson, 2001; Kumar et al., 2011). BC is produced from incomplete combustion processes, in particular from diesel engines, which are a major source in these areas (Jacobson, 2001). Radiative forcing due to BC is globally ranked as the third most important anthropogenic climate-warming agent after carbon dioxide and methane (IPCC, 2013). Recent studies show that components of the aerosol organic fraction (OA) known as brown carbon (BrC), also contribute to light absorption, most efficiently at short wavelengths (Park et al., 2010).

Numerous studies have identified the Mediterranean basin as one of the most prominent "Hot-Spots" in projected climate change assessments (Mallet et al., 2013). In order to quantify aerosol effects over this region, some important experiments such as the Chemistry-Aerosol Mediterranean Experiment (ChArMEx) have been developed (Sicard et al., 2014). The objective of the current study is to present a four year period of aerosol absorption properties using a seven-wavelength Aethalometer. Aethalometer measurements were compensated by calculating site-specific calibration parameters attending to their spectral and seasonal variation. The temporal evolution of the seven absorption coefficients and the absorption Ångström exponent has been analysed. Other parameters influencing these aerosol absorption properties, such as the mixing layer height and traffic, have been also analysed and related to the diurnal variations of the aerosol absorption properties.

2. Measurements and techniques

In-situ measurements presented in this study were performed in Burjassot (39.51° N, 0.42° W, 60 m a.s.l.), from February 2011 to February 2015. Burjassot is a town of 38,200 inhabitants within the metropolitan area of Valencia, in the eastern coast of Spain (Fig. 1). Valencia's metropolitan area has a population of 1,557,000 inhabitants, from which 796,000 live in the city. Burjassot is located 5 km northwest from the city's centre, which makes the measurement site to be directly affected by the urban and industrial pollution typical of the area. Its closeness to the western coast of the Mediterranean Sea (10 km west) also determines the type of aerosols in the region, influenced by particles brought from such distant regions such as the Sahara, industrial continental Europe or oceanic regions.

Sampling for all the different instruments has been performed at ambient relative humidity without aerosol cut-off at about 15 m above ground level (Esteve et al., 2012). The aerosol absorption coefficient (b_{abs}) has been measured using two different filter-

45°N 40°N 40°N 40°N 40°N 5°E 10°E Lon

Fig. 1. Geographic location of the Burjassot monitoring site.

based instruments. One of them is the Aethalometer model AE-31-ER (Magee Scientific). This instrument measures the light attenuation at seven different wavelengths (λ) covering the UV (370 nm), visible (470, 520, 590, and 660 nm), and NIR (880 and 950 nm) ranges. A complete description of the operating principles of Aethalometers can be found in Hansen (2005). In our work the Aethalometer has been operated with a flow rate of 4 L min⁻¹ and a sampling interval of 5 min.

A Multi-Angle Absorption Photometer (MAAP) model 5012 (Thermo Scientific) which measures the light transmitted through and backscattered by a particle-loaded filter at 637 nm (Müller et al., 2011), has also been used to measure b_{abs} . A detailed description of this method is provided by Petzold and Schönlinner (2004). The MAAP works at a constant flow rate of 16.7 L min⁻¹ and was set to provide measurements every minute from February 2013 to February 2015. The method's total uncertainty for the aerosol absorption coefficient inferred from the MAAP's measurements is around 12% (Petzold and Schönlinner, 2004).

3. Data compensation

3.1. MAAP data compensation

It is commonly known that all filter-based absorption photometers are affected by several artefacts which need to be compensated (Müller et al., 2011). Most of the methods proposed rely on the continuity of the data (e.g., Collaud Coen et al., 2010; Hyvärinen et al., 2013) to derive compensation parameters. In this study, the MAAP has been used as a reference instrument to obtain the Aethalometer compensation factors following the method proposed in Segura et al. (2014).

Hyvärinen et al. (2013) observed that MAAP measurements were also affected by artefacts at high BC concentrations, which were different than those observed for other filter-based instruments. The reflected signal measured by the photodetector located at 165° was proven to be free of this artefact and thus it has been chosen to obtain the absorption coefficient at 637 nm (Segura et al., 2014). Petzold et al. (2005) determined an empirical method to compensate this artefact using test aerosols. Therefore, following Hyvärinen et al. (2013), the aerosol absorption coefficient obtained with the MAAP (b_{abs,MAAP}) has been calculated using the reflected signal as:

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