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Spatial and temporal variability of carbonaceous aerosols: Assessing the impact of biomass burning in the urban environment

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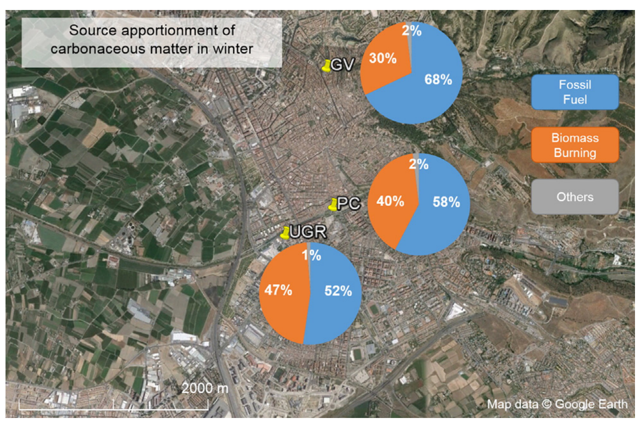
HIGHLIGHTS

- Biomass burning impact in southern Europe urban air quality sparsely investigated.
- Levoglucosan tracer and online real-time Aethalometer methods have been applied.
- High contribution of biomass burning during winter in the suburban area.
- Lower contribution of biomass burning in the city center.

GRAPHICAL ABSTRACT



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ABSTRACT

Biomass burning (BB) is a significant source of atmospheric particles in many parts of the world. Whereas many studies have demonstrated the importance of BB emissions in central and northern Europe, especially in rural areas, its impact in urban air quality of southern European countries has been sparsely investigated. In this study, highly time resolved multi-wavelength absorption coefficients together with levoglucosan (BB tracer) mass concentrations were combined to apportion carbonaceous aerosol sources. The Aethalometer model takes advantage of the different spectral behavior of BB and fossil fuel (FF) combustion aerosols. The model was found to be more sensitive to the assumed value of the aerosol Ångström exponent (AAE) for FF (AAE_{ff}) than to the AAE for BB (AAE_{bb}). As result of various sensitivity tests the model was optimized with $AAE_{ff} = 1.1$ and $AAE_{bb} = 2$. The Aethalometer model and levoglucosan tracer estimates were in good agreement. The Aethalometer model was further applied to data from three sites in Granada urban area to evaluate the spatial variation of CM_{ff} and CM_{bb} (carbonaceous matter from FF or BB origin, respectively) concentrations within the city. The results showed that CM_{bb} was lower in the city centre while it has an unexpected profound impact

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Aethalometer
Ångström exponent

on the CM levels measured in the suburbs (about 40%). Analysis of BB tracers with respect to wind speed suggested that BB was dominated by sources outside the city, to the west in a rural area. Distinguishing whether it corresponds to agricultural waste burning or with biomass burning for domestic heating was not possible. This study also shows that although traffic restrictions measures contribute to reduce carbonaceous concentrations, the extent of the reduction is very local. Other sources such as BB, which can contribute to CM as much as traffic emissions, should be targeted to reduce air pollution.

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1. Introduction

The study of ambient particulate matter (PM) concentrations in urban environments is of interest due to its adverse effects on human health (e.g. Pope and Dockery, 2006). Traffic emissions are of particular concern in urban areas and their surroundings, since traffic-related pollutants have been associated with overall mortality increase (e.g. Hoek et al., 2000), lung cancer risk (e.g. Beelen et al., 2008), and worsening of respiratory health (e.g. Brauer et al., 2002). Among atmospheric pollutants, particulate black carbon (BC) has become a matter of concern during the past years. Black Carbon (BC) is a primary product of incomplete combustion of carbonaceous fuels, normally originated from diesel engines in urban areas (e.g. Hamilton and Mansfield, 1991; Pakkanen et al., 2000). In addition to diesel engines from the traffic sector, biomass burning and domestic heating (based on fossil or biomass fuels) constitute the main sources of BC in the atmosphere. Janssen et al. (2012) reviewed epidemiological, clinical, and toxicological studies and reported sufficient evidence of both short-term and long-term health effects of BC. However, according to these authors, BC itself may not be a major toxic component of PM, but it rather acts as an indicator of other combustion-originating toxic constituents. As stated by the World Health Organization "BC may carry a wide variety of chemicals to the lungs, the body's major defense cells and possibly the circulatory system" (Janssen et al., 2012).

Residential wood burning is an increasingly common popular alternative energy source to fossil fuels since the last years (Fuller et al., 2013). The use of biomass as a heating source has recently increased due to the promotion of CO_2 neutral policies, higher taxes in heating diesel and the financial crisis with significant repercussions on the average household income. In this sense, wood smoke can be an important source of particulate and gaseous pollution significantly affecting rural and urban locations (Viana et al., 2013). This is particularly important during winter when the meteorological conditions in combination with the increase use of biomass burning might significantly and detrimentally impact air quality in urban areas, exposing large populations to pollutant concentrations exceeding regulatory limits. In Central and Northern Europe biomass burning emissions have been identified as a major source of air pollutants (e.g. Yttri et al., 2005; Alfarra et al., 2007; Sandradewi et al., 2008), accounting for up to 80% of fine aerosols (Larssen et al., 2006) and being one of the major sources of organic aerosols (Caseiro et al., 2009) during winter time. In Southern Europe, domestic biomass burning is a much less frequent practice. However, recent studies conducted in Mediterranean urban areas have identified residential biomass burning as an increasing practice contributing to air quality degradation (Viana et al., 2013; Paraskevopoulou et al., 2015; Sarigiannis et al., 2015; Nava et al., 2015). In addition to biomass burning for residential heating, burning of agricultural waste is a common practice that releases significant amount of gaseous and particulate pollutants into the atmosphere (Andreae and Merlet, 2001; Dambruoso et al., 2014; Kostenidou et al., 2013). For example, Dambruoso et al. (2014) and Kostenidou et al. (2013) showed the large impact of olive tree branches burning on Mediterranean air quality. Farms can be close to densely populated areas, and so, burning of agricultural waste may cause higher exposure to air pollutants increasing potential risks for human health.

Several approaches have been used in the literature to identify biomass burning contribution to aerosolized particulate matter in ambient air. Most of the approaches focus on the determination of chemical biomass burning tracers in PM samples such as water soluble potassium, anhydrosugars like levoglucosan or mannosan, and/or organic carbon (Khalil and Rasmussen, 2003; Alves et al., 2011; Zotter et al., 2016). Other approaches take advantage of the spectral dependence of the absorption coefficient of carbonaceous particles from different sources (Sandradewi et al., 2008; Cazorla et al., 2013; Ealo et al., 2016). Carbonaceous particles from biomass burning sources feature enhanced absorption at shorter wavelengths (Sandradewi et al., 2008). The main advantage of this technique is the availability of real-time online measurements. As a major drawback is important to highlight that other aerosol species like dust may also have an absorption enhancement at shorter wavelengths (Valenzuela et al., 2015).

Atmospheric aerosol optical properties have been widely investigated in the city of Granada at surface level (e.g., Lyamani et al., 2008; Lyamani et al., 2010; Titos et al., 2012; Segura et al., 2014), in the vertical column with height resolution (e.g., Guerrero-Rascado et al., 2008; Alados-Arboledas et al., 2011; Bravo-Aranda et al., 2015 and references therein) and column-integrated (e.g., Lyamani et al., 2005; Valenzuela et al., 2012). The meteorological conditions together with the topography of the city sited in a valley surrounded by mountains of high elevation (about 3500 m a.s.l.) contribute to high concentrations of pollutants close to the surface and their slow dispersion, especially in winter season (Lyamani et al., 2012). Previous studies pointed to traffic as one of the major sources contributing to air quality degradation in Granada; BC mass concentrations exhibited two distinct maxima during the day in coincidence with traffic rush hours (Lyamani et al., 2011; Titos et al., 2012; Segura et al., 2014). The chemical composition of fine particulate matter (PM_1) during the period 2006–2010 showed that carbonaceous matter (EC + OM) was one of the major contributor to PM_1 at Granada accounting for around 40% of PM_1 and with higher concentrations in winter than in summer (Titos et al., 2014a). OC/EC ratio in Granada is similar to that found in other urban areas in Spain like Madrid or Zaragoza (Querol et al., 2013). Recent research by Titos et al. (2014a) identified and apportioned the main aerosol sources of fine and coarse PM in Granada applying the PMF technique (Paatero, 1997) to speciated PM_{10} and PM_1 samples. Traffic exhaust was found to be the major aerosol source contributing around 50% to PM_1 levels during winter (Titos et al., 2014a).

In order to reduce local air pollutants and improve air quality, in the last years local authorities have implemented various measures focusing on reducing traffic emissions, especially in the city centre. These emission abatement measures led to a significant reduction of BC mass concentrations of 37% in the city centre (Titos et al., 2015). However, the efficiency of these measures in the whole urban area was not clear (Titos et al., 2015). Pollutant emissions from other sources such as biomass burning were not included in the emission abatement strategies due to the lack of information on their magnitude and sources. This information is crucial for the implementation of effective mitigation measures. In Andalusia, burning of agricultural residues in the field is allowed during cold season, approximately from 15 October to 1 June (Junta de Andalucía, http://www.juntadeandalucia.es/medioambiente/web/aplicaciones/Normativa/ficheros/

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