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Black Carbon aerosol measurements and simulation in two cities in south-west Spain

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HIGHLIGHTS

- Black Carbon is measured with high temporal resolution at two city sites.
- We evaluate CAMx Black Carbon simulations during a winter and summer period.
- Results show meteorology drives Black Carbon seasonal variability in these urban areas.
- Targeted mitigation strategies during wintertime are recommended.

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ABSTRACT

Black carbon (BC) has been simulated for south-west Spain with the air quality model CAMx driven by the MM5 meteorological model, with a spatial resolution of 2 km × 2 km and a temporal resolution of 1 h. The simulation results were evaluated against hourly equivalent black carbon (EBC) concentrations obtained in the cities of Seville and Huelva for a winter period (January 2013) and a summer period (June 2013). A large seasonal variability was observed in $PM_{2.5}$ EBC concentration in the two cities, with higher concentrations in wintertime; summertime EBC concentrations were typically less than half those of the wintertime. The model captured the large diurnal, seasonal and day to day variability in these urban areas, mean biases ranged between −0.14 and 0.07 μg m^{-3} in winter and between 0.01 and 0.29 μg m^{-3} in summer while hourly $PM_{2.5}$ EBC observations ranged between 0.03 μg m^{-3} to 10.9 μg m^{-3}. The diurnal variation in EBC concentrations was bimodal, with a morning and evening peak. However, the EBC evening peak was much smaller in summer than in winter. The modelling analysis demonstrates that the seasonal and day to day variability in EBC concentration in these urban areas is primarily driven by the variation in meteorological conditions. An evaluation of the role of regional versus local contributions to EBC concentrations indicates that in the medium size city of Seville, local on-road sources are dominant, whereas in the small size city of Huelva, local as well as regional sources produce a similar contribution. Considering the large diesel share of the vehicle fleet in Spain (currently ~ 56%), we conclude that continued reduction of BC from diesel on-road sources in these urban areas is indeed a priority, and we suggest that targeted mitigation strategies, for example reducing the heaviest emitters in wintertime, would yield the greatest benefits.

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1. Introduction

Black carbon (BC) has been identified as one of three key short-lived climate pollutants (SLCPs) for which emission reduction measures could contribute to slowing near-term climate change while having the co-benefit of improving air quality and thereby reducing the adverse health effects of air pollution (UNEP, 2011). Bond et al. (2013) concluded that the total black carbon climate forcing (including direct, indirect and snow and ice effects) is positive and is second only to carbon dioxide in terms of its climate forcing in our present day atmosphere. The potential for climate change mitigation through BC emission reductions depends on geographical region and is source dependent as co-emissions of organic carbon, sulphate and gaseous species affect the net climate forcing. Bond et al. (2013) suggest that diesel sources provide the most promising black carbon mitigation options due both to their positive net climate forcing and to the availability of abatement technologies and their implementation potential.

The adverse health effects of particulate matter (PM) are well known and documented (e.g. Brook et al., 2010; Lepeule et al., 2012; Pope et al., 2009; WHO, 2013). A recent extensive review of the health effects of black carbon (WHO, 2012) concluded that there is sufficient evidence of adverse effects of BC exposure and suggested that BC, although in itself may not be a major toxic component of $PM_{2.5}$, may act as a universal carrier of toxic components to the body. Furthermore, the International Agency for Research on Cancer (IARC) recently classified diesel engine exhaust as carcinogenic to humans (IARC, 2012). In addition, BC has been identified as a more sensitive indicator to vehicle exhaust related air pollution compared with measurements of PM mass such as PM_{10} and $PM_{2.5}$ (e.g. Janssen et al., 2011; Keuken et al., 2012). Reche et al. (2011) recommended the measurement of BC at air quality monitoring sites alongside measurements of PM mass and particle number concentration, to more fully reflect the impact of vehicle exhaust emissions on ambient air quality.

There have been various global (e.g Koch et al., 2009; Gilardoni et al., 2011) and regional modelling studies of BC or elemental carbon (EC) (e.g. Schaap et al., 2004; Simpson et al., 2007; Tsyro et al., 2007; Genberg et al., 2013; Hienola et al., 2013). However, modelling studies focussing on the mesoscale are scarcer. Sciare et al. (2010) modelled inorganic and carbonaceous aerosols and their relative contribution to $PM_{2.5}$ mass in the Paris area while Couvidat et al. (2013) modelled elemental carbon and organic carbon (OC) also in the Paris area. In general, both studies reported a satisfactory performance for EC with Couvidat et al. (2013) observing both over and under-estimation of EC depending on the measurement site and measuring period. Ensberg et al. (2013) conducted a modelling study of black carbon and inorganic aerosols in the Los Angeles Basin and reported that BC predictions were generally in good agreement with the measurements at their ground site although the model did miss peak concentrations on specific days. These urban studies were conducted during spring or summer periods. Keuken et al. (2013) report a modelling study of EC at regional, urban and traffic sites in The Netherlands for 2011 and found that the model overestimated concentrations at regional sites and at urban background sites, likely due to too high primary $PM_{2.5}$ emissions and/or EC fractions and a too low road traffic emission height. They found good agreement between their modelled and measured traffic contribution to EC.

A limitation in the evaluation of modelling studies of BC is the availability of high quality measurements, particularly as monitoring of BC in ambient air at urban sites is, up to this date, not required by EU legislation (EEA, 2013) and so is not frequently measured. In light of these aspects, a BC measurement program with high temporal resolution was initiated in two cities in south-west Spain in 2012 to characterise the behaviour of this species. In addition to the measurement program, a three-dimensional air quality model (CAMx) was implemented to investigate the dynamics of this primary aerosol and its spatial and temporal variability and to explore the controlling factors on the BC concentrations in these urban areas. This study presents measurements and simulation of BC for a winter period (January 2013) and a summer period (June 2013) from this measurement dataset. The structure of the paper is as follows, Section 2 describes the measurements and model set up, Section 3 includes results and discussion of the evaluation of the model simulations on a seasonal and diurnal scale and an assessment of regional versus local sources, while conclusions are presented in Section 4.

2. Methodology

2.1. Measurements

Black Carbon was measured, simultaneously, at two urban sites in the cities of Seville (~700,000 inhabitants) and Huelva (~150,000 inhabitants), in the south-west of Spain. Optical measurements of black carbon in PM_{10} were conducted with a Multi-Angle Absorption Photometer (Thermo™, model CARUSSO 5012) measured with a 10-min resolution (subsequently averaged to hourly resolution). The uncertainty of the absorbance measurement of the MAPP according to Petzold and Schönlinner (2004) is ±12%. The instrument set up is described in Fernández-Camacho et al. (2010). These were converted to equivalent black carbon (EBC) mass concentrations for each site by comparing with PM_{10} filter samples (quartz-fibre, MUNKTELL™) collected in a high volume Graseby Anderson™ sampler (68 m^3 h^{-1}) analysed for elemental carbon (EC) in the laboratory using the Thermo Optical Transmittance technique with a Sunset Laboratory™ OC-EC analyser and the EUSAAR2 (European Supersites for Atmospheric Aerosol Research) protocol (Cavalli et al., 2010). The term equivalent black carbon is used hereafter for the measurements reported here, following recommendations on Black Carbon terminology by the GAW Aerosol Scientific Advisory Group (GAW/WMO, 2011) and Petzold et al. (2013). The site specific mass-absorption efficiencies (MAE) obtained were 9.79 and 10.31 m^2 g^{-1} for Seville and Huelva, respectively. The mean measured $PM_{2.5}/PM_{10}$ BC ratio (0.74 ± 0.025) was utilised to determine the $PM_{2.5}$ BC concentration (see Fernández-Camacho et al., 2010 for more details).

The measurement sites used were (see Fig. 1):

- Príncipes (37.375° N, 6.006° W, 8 m a.s.l.), an urban background site influenced by road traffic located in a park in the south-west of the city of Seville. The closest roads lie about 50 m to the east and 65 m to the north-west of the measurement site.
- University Campus (37.272° N, 6.925° W, 17 m a.s.l.), an urban background site located on the north-east side of the city of Huelva. There are some minor roads within the university campus, aside from these, the closest roads lie within about 150 m–250 m of the measurement site.

The measurements were conducted in Seville for a period of 18 months (June 2012 to November 2013) and in Huelva for a period of 12 months (December 2012 to November 2013). In this study, we present measurements and simulation of EBC for January and June 2013.

In addition, the meteorological simulations that determine the transport and dispersion of BC were compared against surface-based observational data from 12 meteorological stations, belonging to the Meteorological State Agency of Spain (AEMET). The measurements used to evaluate the meteorological simulations were hourly measurements of wind speed and wind direction at