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

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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 x 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 to 0.07 μg m^{-3} in winter and between 0.01 to 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.

Keywords: Black Carbon, Air Quality, CAMx, Model Evaluation

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, 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 ten-minute resolution (subsequently averaged to hourly resolution). 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 to 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 10 m, air temperature at 2 m, and precipitation. Road-traffic intensity (number of vehicles per hour) was also measured on the main road (Avenida de Blas Infante) close to the Príncipes measurement site in Seville.

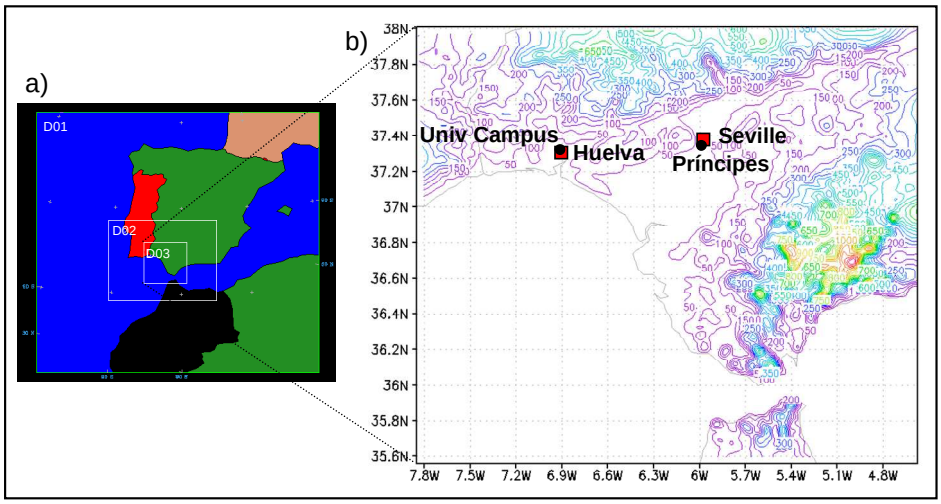

Figure 1. a) Map of the three model domains (D01, D02 and D03) and b) terrain height (m a.s.l.) of the inner domain (D03). Also shown are the locations of the measurement sites, Príncipes, Seville and University Campus, Huelva.

2.2 Model Description and Set Up

The air quality model chosen for this study was the Comprehensive Air-quality Model with eXtensions (CAMx) version 4.51 (ENVIRON, 2008). CAMx is a three-dimensional Eulerian chemistry transport model including aerosol chemistry. In this study we used the CF aerosol scheme. Three nested domains were used in the CAMx simulations, with 18 x 18 km, 6 x 6 km and 2 x 2 km horizontal resolution. The outer domain (D01) covers the Iberian Peninsula, and parts of southern France and North Africa; domain (D02) includes southern Spain and southern Portugal while the inner domain (D03) covers the study area, the south-west region of Spain (see Fig. 1).

Initial and boundary conditions for the modelling outer domain were taken from the GEOS-Chem global transport model (Bey et al., 2001; Barrett et al., 2012). The GEOS-Chem model output was for the year 2006, monthly average values were generated for January and June to

use as input for our simulation. A spin-up of 72 h was carried out prior to each CAMx simulation to minimize the sensitivity of the results to the initial conditions.

Meteorological input data for the CAMx simulations were provided by the non-hydrostatic Mesoscale Meteorological model (MM5) v3.7 (Grell et al., 1995). Three grids (using two-way nesting) with the same horizontal resolution of 18, 6 and 2 km were used. MM5 was configured with 30 vertical layers in a terrain following coordinate system, with increasing vertical resolution closer to the surface. The model top corresponds to approximately 550hPa with a surface layer of about 35 m above ground. CAMx was run with 16 of the first 18 vertical layers utilised in MM5. The European Centre for Medium-range Weather Forecasts (ECMWF) numerical weather prediction model analysis provided initial and boundary conditions for MM5 at six hourly intervals with a resolution of 0.25°.

2.3 Emissions

The emission inventory used in this study was based on Castell et al. (2010) and Milford et al. (2013). The outer domains include anthropogenic emissions from the European Monitoring and Evaluation Program (EMEP) emission inventory for the reporting year 2012 (http://www.emep.int) interpolated to 18 km and 6 km spatial resolution. The inner domain includes industrial emissions, from both area and point sources, and on-road traffic emissions.

Industrial emissions in the inner domain were taken directly from the Spanish Pollutant Release and Transfer Register (PRTR-E; http://www.prtr-es.es) for the year 2013. This register contains emissions from all large industrial installations and these emissions were then separated into area and point sources. Particulate emissions in this register are given as PM_{10}. $PM_{2.5}$ emissions were estimated for each industrial installation using source dependent $PM_{2.5}/PM_{10}$ fractions (CEIDARS, 2012). PEC (Primary Elemental Carbon) emissions were derived from $PM_{2.5}$ emissions utilising mass fractions assigned to different source profiles (EPA, 2009).

The on-road traffic emissions were obtained from the Spanish National Emission Inventory for the year 2011, as this was the most recent data available at the time of the study. These emissions were spatially disaggregated using the road network distribution in the study region and the average traffic intensity across that network. An hourly temporal profile differing for workdays and weekends was used for the temporal disaggregation of the emissions (Castell et al., 2010). Chemical speciation of primary $PM_{2.5}$ emissions for on-road traffic into PEC emissions was also conducted using source dependent mass fractions (EPA, 2009).

Residential combustion emissions are not currently included in the emission inventory for the inner domain.

3 Results and Discussion

3.1 Meteorology

The evaluation of the meteorological simulations against surface-based observational data showed that the model captured most of the seasonal, diurnal and spatial variability. The meteorological stations located closest to the measurement sites were Seville Tablada (located about 1 km south from Príncipes, Seville) and Ronda Este (located about 2 km north-east from University Campus, Huelva). Observed and simulated wind speed, wind direction, and temperature data are shown in Fig 2 and Fig. 3 for the Seville Tablada and Ronda Este sites for the winter (January) and summer (June) simulations, respectively.

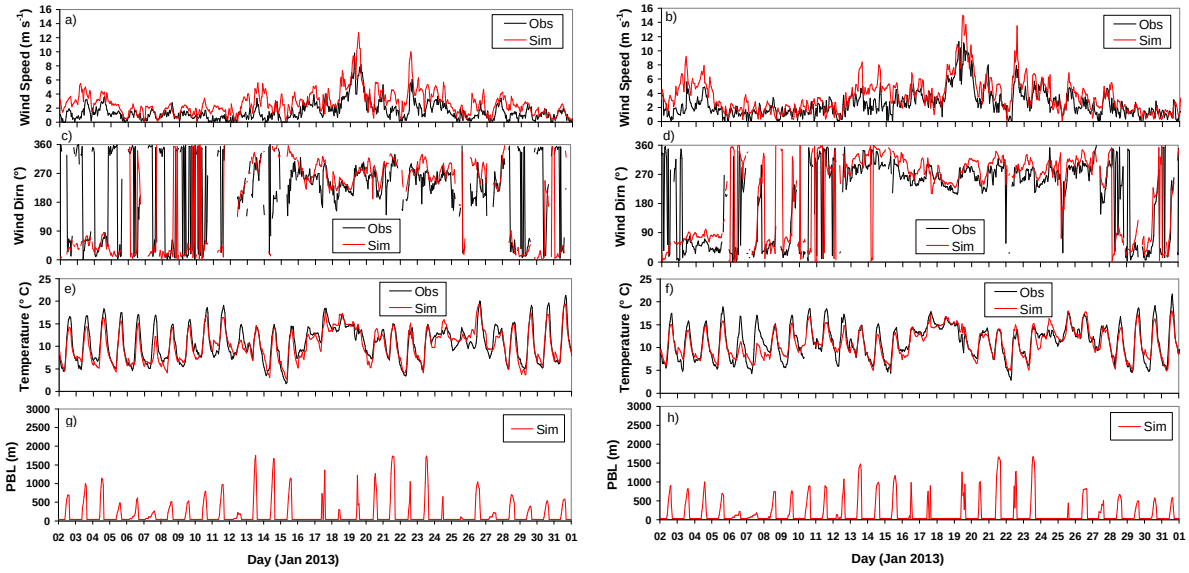

Figure 2. Observed and simulated wind speed (a, b) and wind direction (c, d) at 10 m, temperature at 2 m (e, f) and simulated PBL height (g, h) during January 2013 at Seville Tablada (left panel) and Ronda Este (right panel).

The meteorological model had a tendency to overpredict the wind speed at both the Seville and Huelva sites; mean wind speed biases were similar during the winter and summer periods for both sites and ranged from 0.9 to 1.1 m s^{-1} (Table 1). The mean biases in simulated 2 m temperature were low for both sites, with values ranging from -0.4 °C in winter to 0.4 °C for the summer period (Table 1). The simulated Planetary Boundary Layer (PBL) height for Seville and Huelva shows that the majority of days in January 2013 had PBL values < 1000 m (Fig. 2g and h), while in June 2013 in Seville, the simulated daily peak PBL values were

between 1500 to 2750 m (Fig. 3g) with the majority of days having PBL values > 2000 m. In Huelva in the summer period, the simulated PBL values were generally slightly lower than in Seville, with daily peak values between 1060 to 2450 m (Fig. 3h) with the majority of days having PBL values > 1500 m. It was not possible to compare the simulated PBL heights with observational data, as there were no radio sounding data publicly available for any sites close to or representative of Seville or Huelva during these periods.

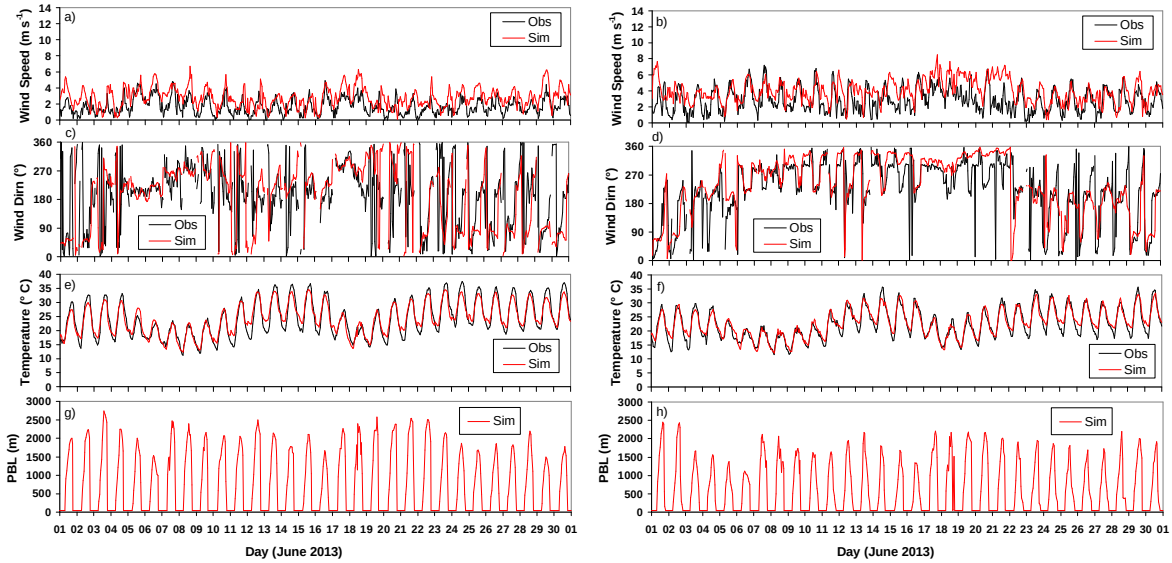

Figure 3. Observed and simulated wind speed (a, b) and wind direction (c, d) at 10 m, temperature at 2 m (e, f) and simulated PBL height (g, h) during June 2013 at Seville Tablada (left panel) and Ronda Este (right panel).

Table 1. Statistical metrics for observed and simulated meteorological variables at Seville Tablada and Huelva Ronda Este sites.

	SM	OM	MB	NMB (%)	*R*
January 2013					
WS, Seville Tablada (m s^{-1})	2.7	1.5	1.1	72	0.75
T 2m, Seville Tablada (° C)	10.4	10.8	-0.4	-3	0.91
WS, Huelva RE (m s^{-1})	3.5	2.5	0.9	36	0.80
T 2m, Huelva RE (° C)	11.1	11.4	-0.3	-3	0.90
June 2013					
WS, Seville Tablada (m s^{-1})	2.8	1.7	1.1	64	0.57
T 2m, Seville Tablada (° C)	24.3	24.2	0.2	1	0.95
WS, Huelva RE (m s^{-1})	4.0	2.8	1.1	39	0.48
T 2m, Huelva RE (° C)	22.9	22.5	0.4	2	0.95

WS (Wind speed at 10 m), T (Air temperature at 2 m), SM (Simulated mean), OM (Observed mean), MB (Mean Bias), NMB (Normalized Mean Bias), *R* (Correlation Coefficient).

3.2 BC Seasonal and Day to Day Variation

A large seasonal variation in EBC concentrations was observed at both sites, with maximum concentrations occurring in the winter period. The maximum observed hourly $PM_{2.5}$ EBC concentration at Príncipes, Seville for the winter and summer period were 10.9 μg m^{-3} and 4.0 μg m^{-3}, respectively, with mean concentrations of 1.6 μg m^{-3} and 0.6 μg m^{-3} (Fig. 4a and 5a). Observed $PM_{2.5}$ EBC concentrations were lower at University Campus, Huelva, with a mean and maximum of 0.7 μg m^{-3} and 5.7 μg m^{-3} in the winter period and 0.4 μg m^{-3} and 2.4 μg m^{-3} in the summer period, respectively (Fig. 4b and 5b).

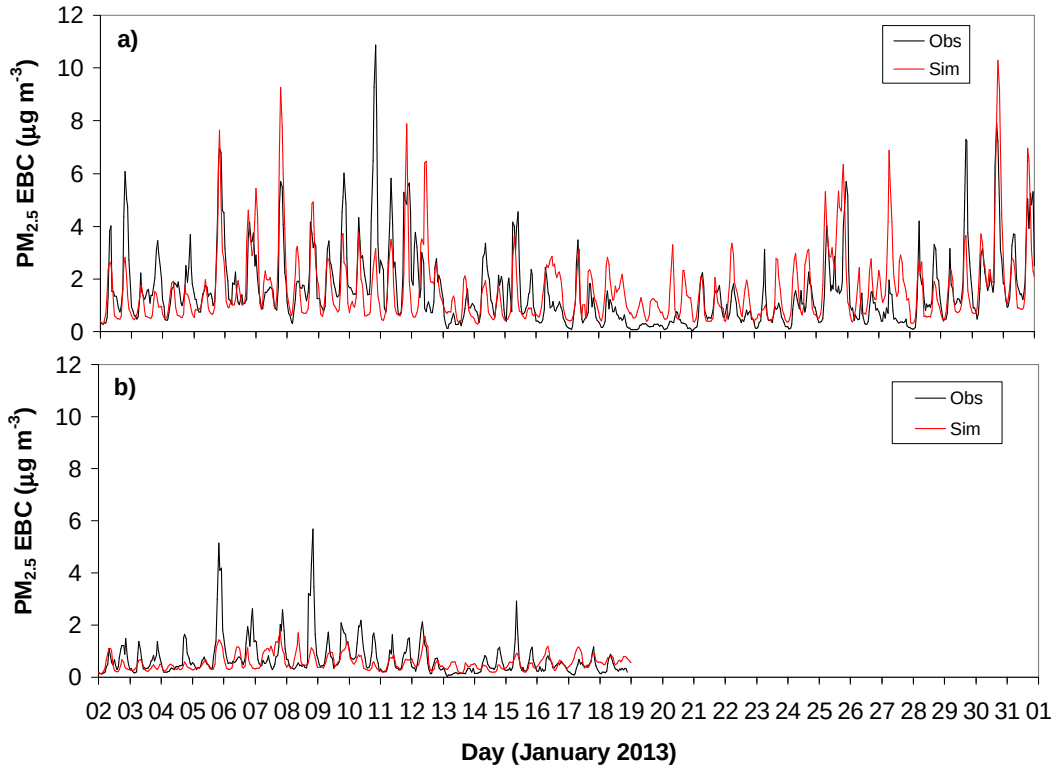

Figure 4. Observed $PM_{2.5}$ EBC and simulated $PM_{2.5}$ PEC concentrations at a) Seville and b) Huelva, during January 2013. Observation data not available in Huelva for 19-31 Jan.

The modelling system performed generally well in capturing both these seasonal differences in concentration and the spatial variation between these two city sites. The EBC concentrations were slightly overestimated at the Seville site during winter (1.63 μg m^{-3} simulated EBC versus 1.56 μg m^{-3} measurements, MB = 0.07 μg m^{-3}, NMB = 5%), while the EBC concentrations were underestimated during winter at the Huelva site (0.58 μg m^{-3} simulated EBC versus 0.71 μg m^{-3} measurements, MB = -0.14 μg m^{-3}, NMB = -20%) (Table 2). During the summer period, the EBC concentrations showed a greater overestimation at the

Seville site (MB = 0.29 μg m^{-3}, NMB = 45%) while the mean bias was now slightly positive for the Huelva site (MB = 0.01 μg m^{-3}, NMB = 4%) (Table 2). Some of the discrepancy between model and measurements in Huelva during the winter period could be attributed to residential combustion emissions, which are not currently included in the emission inventory in the inner domain. However, we would expect the contribution from this source sector to be less in these urban areas than in rural areas and less in this southern Europe region in comparison to northern Europe due to the warmer winter climate.

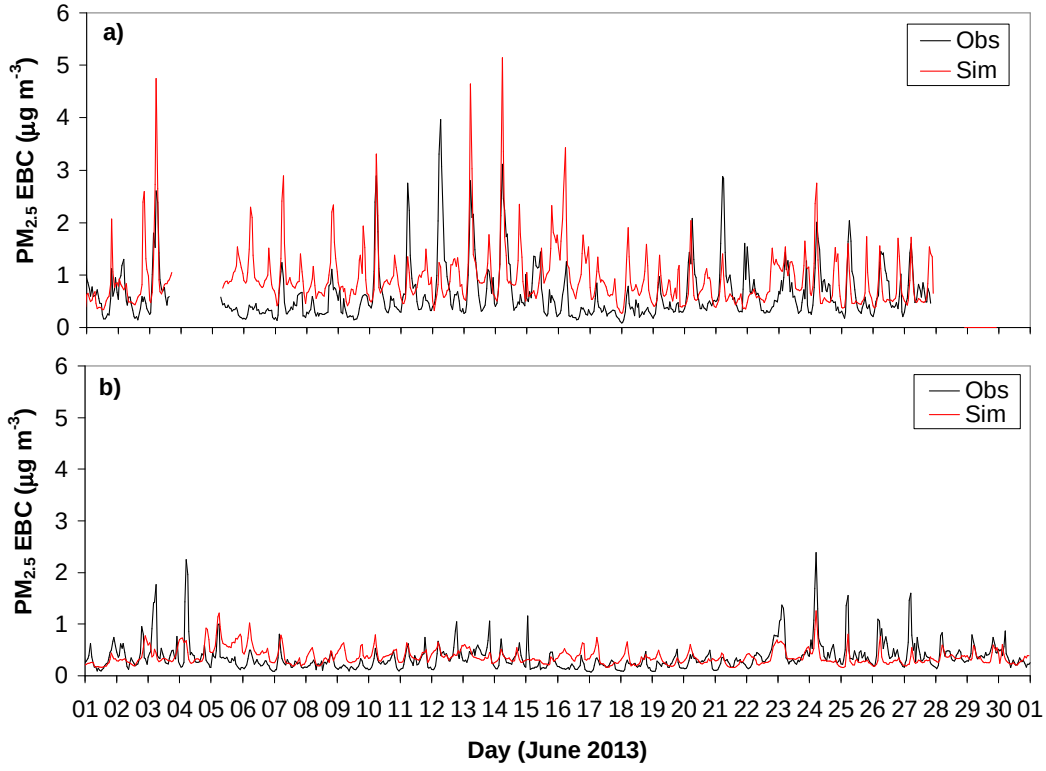

Figure 5. Observed $PM_{2.5}$ EBC and simulated $PM_{2.5}$ PEC concentrations at a) Seville and b) Huelva, during June 2013.

Table 2. Statistical metrics for observed EBC ($PM_{2.5}$) and simulated $PM_{2.5}$ PEC at Príncipes (Seville) and University Campus (Huelva).

	SM (μg m^{-3})	OM (μg m^{-3})	MB (μg m^{-3})	NMB (%)	*R*
January 2013					
Príncipes	1.63	1.56	0.07	5	0.60
University Campus	0.58	0.71	-0.14	-20	0.50
June 2013					
Príncipes	0.93	0.63	0.29	45	0.44
University Campus	0.37	0.35	0.01	4	0.37

SM (Simulated mean), OM (Observed mean), MB (Mean Bias), NMB (Normalized Mean Bias), *R* (Correlation Coefficient).

As well as capturing the seasonal variation, the model also captured the day to day variation within the months, for example capturing the peak concentrations observed at the Príncipes site on the 5, 7, 11 and 30 January 2013 as well as the lower concentrations observed on days such as the 13 and 19 January (Fig. 4a). However, as can be expected, there are some specific days where the modelling system does not capture peaks (e.g. 10 and 29 Jan, Príncipes or 5 and 8 Jan, University Campus) and this is likely due to actual emission pattern changes from day to day that are not possible to capture in the model or deficiencies in the simulated meteorological fields for that day.

In general, the modelling system captured the large range observed in the hourly EBC concentrations (0.03 μg m^{-3} to 10.9 μg m^{-3}). This is demonstrated by the Q-Q plot for observed and simulated $PM_{2.5}$ PEC concentrations (Fig. 6) which shows good agreement throughout the entire quantile range for the Príncipes site during the winter period, while demonstrating the overestimation of the quantiles in the summer period. $PM_{2.5}$ PEC simulations at University Campus show the underestimation at the highest quantiles during the winter period and the reasonable agreement in the rest of the quantile range for both summer and winter periods (Fig. 6).

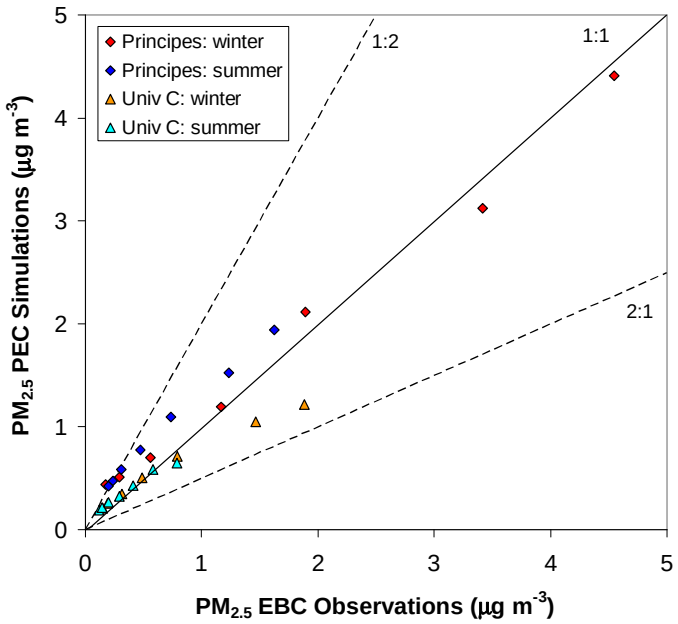

Figure 6. Q-Q plot for simulated $PM_{2.5}$ PEC concentrations versus $PM_{2.5}$ EBC observations, 5th, 10th, 25th, 50th, 75th, 90th and 95th percentiles are plotted for Príncipes and University Campus sites during winter (January 2013) and summer (June 2013).

The emissions implemented in the modelling system do not vary seasonally; there is an hourly temporal profile used to disaggregate the on-road traffic emissions which differs for workdays and weekends (see section 2.3), but no seasonal variation. Therefore, the relatively close agreement of the model simulations with measurements supports the conclusion that, both the day to day variation and the seasonal variation in BC concentration in these urban areas in southern Spain are largely driven by the seasonal and day to day variation in meteorological conditions. This has been suggested by previous measurement studies (e.g. Pereira et al., 2012; Querol et al., 2013), but the modelling analysis confirms the driving role of the meteorology. Although the meteorology largely explains the relative distribution between winter and summer and the peaks, it is important to note that the total magnitude of concentrations is still driven by the emission strength.

3.3 BC Diurnal Variation

The high temporal resolution of the measurements allows the temporal variation during the day to be explored. The mean diurnal observed and simulated $PM_{2.5}$ PEC concentrations for each day of the week are shown in Fig. 7 for both sites for the winter and summer periods, along with the mean diurnal concentrations calculated for all data for each period. At the Príncipes site, during winter, the observed $PM_{2.5}$ EBC concentrations show a maximum in the morning between 06:00-08:00 UTC and then the concentrations decrease to reach a minimum at 14:00 (Fig. 7a). In the late afternoon/evening a secondary maximum is observed between 17:00-19:00 UTC and the observed $PM_{2.5}$ EBC concentrations reach their highest values at this point. The simulated $PM_{2.5}$ PEC concentrations generally capture this winter diurnal variation well, with similar timing for the two peaks. However, the model overestimates the concentrations at the weekend, particularly on Sunday; this is likely due to an overestimation of the traffic emissions at the weekend.

In the summer period at the Príncipes site, it is interesting to note that the secondary maximum in the observed concentrations at 19:00 UTC is now very much smaller than the first maximum observed between 04:00-06:00 UTC and nearly non-existent (Fig. 7b). This marked change in the diurnal behaviour of EBC concentrations in the summer season, compared to the winter, has been observed in other studies (e.g. Saha and Despiau, 2009). They measured BC over a 14-month period at an urban coastal location in south-east France, and found a similar disappearance of the secondary BC peak in the summer. Although the model does simulate a reduced secondary peak compared to the winter period, it doesn't

capture the near disappearance of the secondary peak (Fig 7b), which helps explain the poorer correlation and greater positive bias seen in the performance of the model in the summer (Table 2). Similarly to the winter period, there is a greater discrepancy between the model and the measurements during the weekend, which suggests that the current traffic emission profile overestimates weekend emissions.

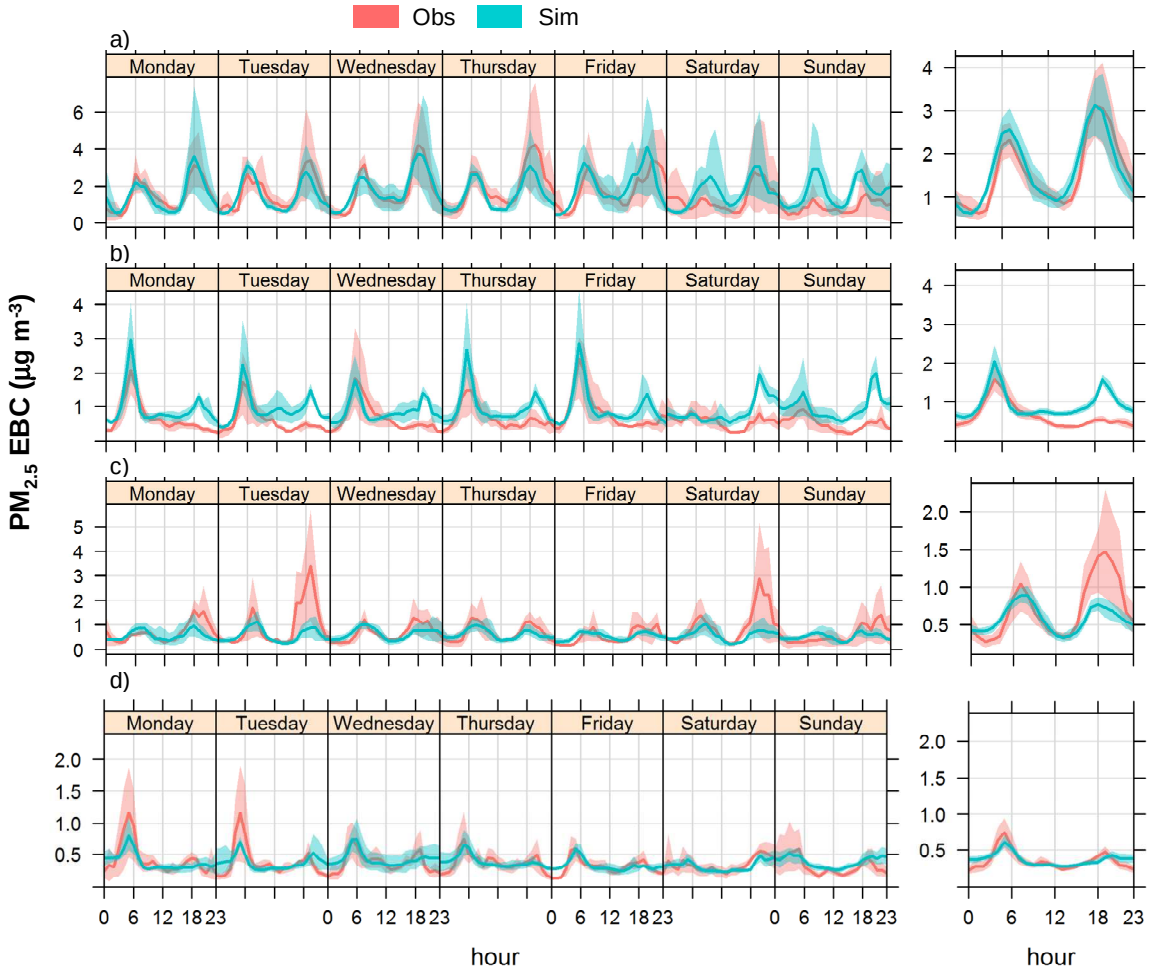

Figure 7. Mean diurnal observed $PM_{2.5}$ EBC and simulated $PM_{2.5}$ PEC concentrations for each day of the week for Príncipes, Seville during a) January 2013 and b) June 2013 and at Univ Campus, Huelva, during c) January 2013 and d) June 2013. Also shown are the mean diurnals calculated for all data for each period in the right-hand panel. Shaded areas represent 95% confidence interval in the mean. Graphics were generated with Openair software (Carslaw and Ropkins, 2012).

At the University Campus site, during winter, the observed $PM_{2.5}$ EBC concentrations also show a maximum in the morning between 06:00-08:00 UTC and a larger secondary maximum occurring in the evening between 18:00-20:00 UTC (Fig. 7c). The model simulations capture the timing of the peaks but underestimates the afternoon peak in the

observations. However, it should be noted that the afternoon peak in the observations shows a large variation in the mean, likely arising from peaks occurring on specific days (Fig. 7c).

During summer at the University Campus site the observed $PM_{2.5}$ EBC concentrations also show a maximum observed in the morning between 04:00-06:00 UTC and a much reduced secondary maximum in the evening between 18:00-20:00 UTC (Fig. 7d). The model captures this diurnal behaviour and in general, captures the magnitude of the concentrations.

Saha and Despiau, (2009) propose that the appearance of the secondary maximum in winter, and the corresponding disappearance of the secondary maximum in summer, can likely be attributed to wind speed and boundary layer dynamics. A shallower boundary layer and lower wind speeds during winter will lead to higher concentrations, while the converse, higher wind speeds and a deeper boundary layer lasting for longer in the day during summer lead to reduced concentrations. The measurements and simulations performed here support these conclusions. The mean observed diurnal wind speed in Seville, Tablada (Fig. 8a) and Huelva, Ronda Este (Fig. 8b) both show a larger maximum in wind speeds in the summer with the maximum occurring later in summer and generally lasting longer, only returning to a minimum at about 22:00 UTC.

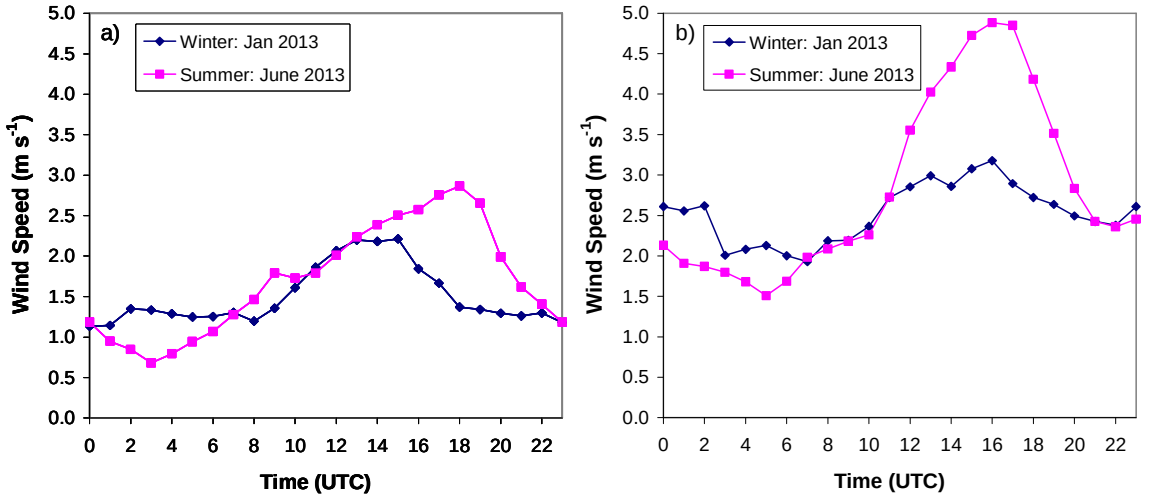

Figure 8. Mean diurnal observed wind speed for a) Seville, Tablada and b) Huelva, Ronda Este during January 2013 and June 2013.

In addition, road-traffic intensity data measurements from the main road closest to the Príncipes measurement site in Seville (Avenida de Blas Infante) demonstrate that there is very little seasonal variation in the weekday road-traffic intensity (Fig. 9a). The only significant seasonal difference is that the road-traffic intensity decreases later during the summer (~21.00

local time) compared to the winter (~18.00 local time) as can be expected due to extra daylight hours in the summer. These measurements support the conclusions that the seasonal variation in BC concentration in these urban areas in southern Spain is largely driven by the seasonal variation in meteorology rather than a seasonal variation in emission strength.

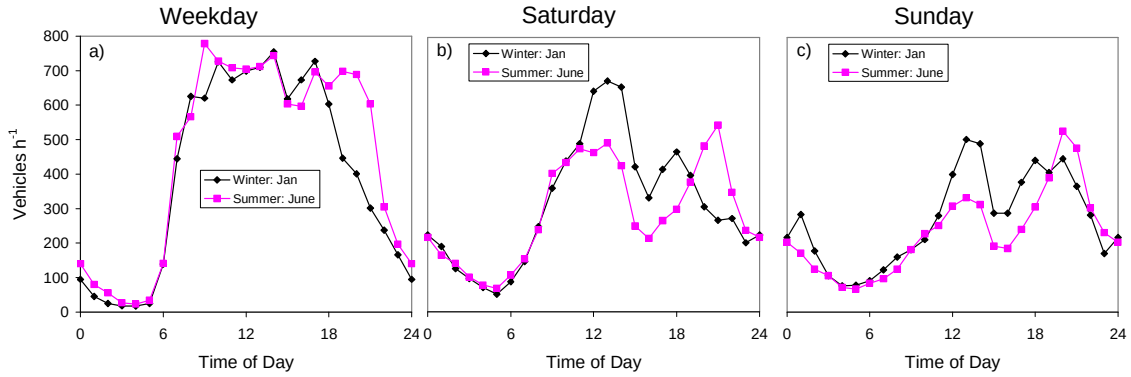

Figure 9. Mean diurnal road-traffic intensity (number of vehicles hour^{-1}) for Seville (Blas Infante) for a) Weekday, b) Saturday and c) Sunday during January 2013 and June 2013. Time is local time, UTC+1h in winter, UTC+2 h in summer.

There are some small seasonal differences in the weekend road-traffic intensity (Fig. 9b and c). The total vehicle counts are similar for January and June but the diurnal behaviour is different. In winter, the first peak in road-traffic intensity is larger than the second peak whereas the converse is true in the summer. This increase in road-traffic intensity in the evening peak with respect to the morning peak in summer is in contrast to the near disappearance of the secondary peak in EBC concentrations in the summer, therefore also providing evidence that the reduction of the secondary peak in EBC concentrations in the summer is dominated by meteorological processes not emission changes.

3.4 Evaluation of regional and local contribution to BC concentrations

In order to evaluate the role of regional transport of BC in this study area, the modelling system was used to compare the base case emission scenario, with all emissions included in all three modelling domains (denoted “all emissions”) with a scenario in which the emissions in the outer two domains were set to zero (denoted “zero D01 and D02 emissions”). As described in Section 2.2, domain D01 covers the Iberian Peninsula, and parts of southern France and North Africa while domain D02 includes parts of southern Spain and southern Portugal. For the sake of brevity, the scenario results are only shown for the Príncipes site in January 2013 (Fig. 10).

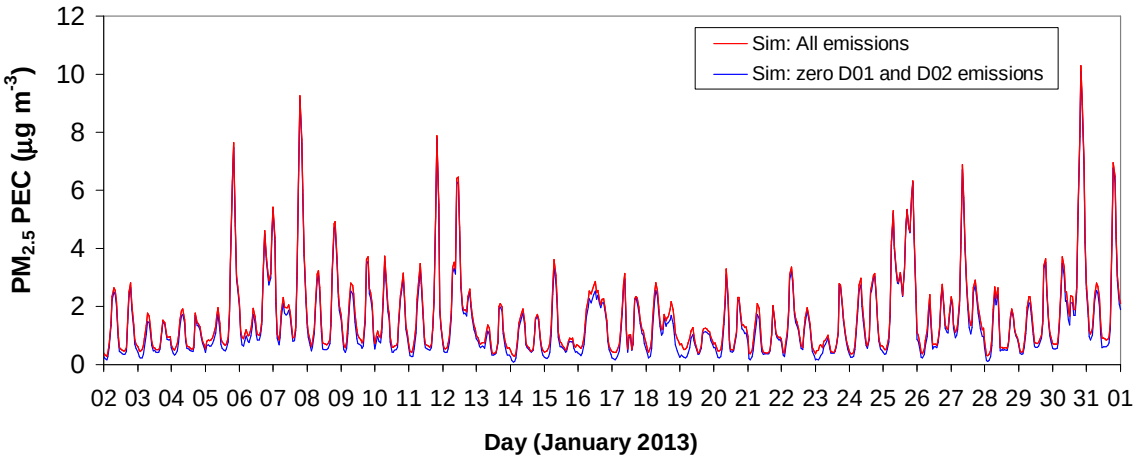

Figure 10. Simulated $PM_{2.5}$ PEC concentration (µg m^{-3}) for the base case emission scenario (All emissions) and the zero D01 and D02 emissions scenario, for Príncipes measurement site, Seville during January 2013.

We observe that the simulated EBC concentrations only decrease slightly, with respect to the base case scenario, when we remove the outer domain emissions (Fig. 10). The mean relative changes in the simulated $PM_{2.5}$ EBC concentration for the Príncipes site during January and June 2013 were -16% and -17%, respectively, while the mean absolute changes in the simulated $PM_{2.5}$ EBC concentration were -0.17 µg m^{-3} and -0.14 µg m^{-3}, respectively. This indicates that regional transport of BC is not a large contributor to EBC concentrations in the medium size urban area of Seville and local sources are dominant.

The mean absolute changes in simulated $PM_{2.5}$ EBC concentration for the Huelva site during January and June 2013 were -0.19 µg m^{-3} and -0.18 µg m^{-3}, respectively. These absolute values in Huelva were similar to those in Seville. However, as the overall magnitude of EBC concentrations was less in Huelva, the relative percentage contribution of the regional transport of BC was larger. The mean relative changes in the simulated $PM_{2.5}$ EBC concentration, with respect to the base case scenario, when we remove the outer domain emissions for the Huelva site during January and June 2013 were -37% and -46%, respectively. Therefore, in the small size city of Huelva, regional sources potentially provide a contribution to EBC concentrations similar to that of local sources.

4 Conclusions

A measurement and modelling program has been initiated in south-west Spain to characterise the concentrations and behaviour of black carbon in two different cities. A large seasonal variability was observed in $PM_{2.5}$ EBC concentration in the two cities, with higher

concentrations in wintertime. The summertime EBC concentrations were typically less than half those of the wintertime.

The modelling system reproduces the diurnal, seasonal and day to day variation in EBC concentrations, with mean biases at the two sites in the winter and summer period ranging from -0.14 to 0.29 $\mu g\ m^{-3}$. Although there are some specific days where the modelling system does not capture peaks, in general the large range observed in the hourly EBC concentrations is reproduced. This suggests that the system is capturing the dominant processes that affect the EBC concentrations in these urban areas, namely the meteorology and the spatial and temporal distribution of emissions.

The modelling analysis demonstrates that the seasonal variation in EBC concentration in these urban areas in southern Spain is largely driven by the seasonal variation in meteorology, with reduced dispersion conditions in wintertime leading to higher concentrations. There is little seasonal variation in emission strength as also demonstrated by road traffic intensity measurements near the Seville site. There is a large day to day variation in EBC concentration observed within both the winter and summer months, with more than an order of magnitude difference between the highest (3.4 $\mu g\ m^{-3}$) and lowest (0.2 $\mu g\ m^{-3}$) mean daily concentrations at the Príncipes Seville site, during January 2013, for example. This day to day variability also seems to be primarily driven by the variation in meteorological conditions, along with some contribution from the differences in emission intensities between weekday and weekend days. It is important to note that although the meteorological conditions largely explain the relative distribution between winter and summer and the peaks on different days, the total magnitude of concentrations is still driven by emission strength.

The high temporal resolution in the measurements aids the understanding of the diurnal variability of EBC concentrations and demonstrates that the diurnal variation of EBC in these urban areas is bimodal, with a morning and evening peak. However, there was a marked seasonality in the diurnal pattern, with the EBC evening peak being larger than the morning peak in the wintertime, while nearly disappearing in the summertime. This is consistent with a deeper boundary layer lasting for longer in the day during summer reducing EBC concentrations. Although the model does simulate a reduced secondary peak compared to the winter period, it doesn't fully simulate the near disappearance of the secondary peak which helps explain the larger discrepancy between the model and measurements observed during the summer period at the Seville site. The comparison between model and measurements also

indicates that the profile utilised for the temporal disaggregation of the road traffic emissions during the weekend is likely overestimating the weekend emissions and future work will enable an improvement in these temporal profiles.

An evaluation of the role of regional transport of BC in this study area, comparing scenarios with and without outer domain emissions, demonstrated that regional transport of BC is not a large contributor to EBC concentrations in a medium sized urban area, such as Seville, and local sources, such as on-road traffic emissions, are dominant. However, in a small sized city, such as Huelva, local sources play a less dominant role and the regional contribution is potentially similar to that of local sources. The contribution from residential combustion, which is not present in the emission inventory, is considered to be minor. However, it could contribute to the underestimation of EBC concentrations observed in Huelva in the winter period and future improvements to our modelling system would include the implementation of this emission source.

It has been suggested that diesel sources provide the most promising black carbon mitigation options (Bond et al., 2013) and we conclude that reduction of BC from diesel on-road sources in these urban areas would indeed be a way forward to gain maximum benefits in both climate mitigation and air quality and resulting health benefit. Based on the results here, we propose that mitigation strategies aimed at a targeted control, for example reducing the heaviest on-road emitters in wintertime, would yield the greatest benefits.

The issue of reduction of on-road diesel BC emissions is particularly relevant for Spain as it is one of the countries in Europe which has experienced the largest dieselisation of its vehicle fleet in recent years. Its percentage share of diesel cars in its total passenger car fleet rose from 15% in 1995 to 50% in 2009, placing it 5th out of the 30 EEA member countries (EEA, 2011) in terms of its diesel share and substantially greater then the EEA average of 28%. The diesel percentage share of the total passenger car fleet and indeed of the total vehicle fleet has continued to rise in Spain, to 55% and 56% in 2013 (DGT, 2013). Taking into account that BC has been identified as a more sensitive indicator to vehicle exhaust related air pollution, the parallel measurement and modelling of BC in urban areas constitutes a useful tool to both evaluate the potential of any proposed traffic emission reduction measures while also monitoring the effectiveness of any traffic emission reduction measures already implemented.

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