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Black carbon aerosols over an urban area in south-eastern Spain: Changes detected after the 2008 economic crisis

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ABSTRACT

Continuous measurements of black carbon (BC) concentrations performed at Granada, an urban location in southeast Spain, using a Multi-Angle Absorption Photometer from December 2005 to November 2008, are analysed and discussed here. The daily mean BC concentrations showed considerable day-to-day variations and were found to vary from low values of 0.5 μg m^{-3} to high values of 8.6 μg m^{-3}, with overall mean and standard deviation of 3.0 ± 1.5 μg m^{-3}. The annual mean BC concentrations were similar during 2006 and 2007 (3.2 ± 1.4 μg m^{-3} and 3.1 ± 1.6 μg m^{-3}, respectively), but decreased by about 16–18% to 2.6 ± 1.4 μg m^{-3} in 2008. This reduction is not only observed in the mean value, but also in the median, third and first quartiles. A Mann–Whitney test at 0.05 significance level confirms that the BC concentration difference between 2006 and 2007 is statistically no significant while the BC concentration in 2008 tends to be less than that in 2006–2007. Analysis of meteorological conditions suggested that although the day-to-day variations in BC concentrations were driven mostly by meteorology, the reduction in the use of fossil fuels due to economic slowdown contributed significantly to the observed decrease in BC concentrations in 2008. Under conditions dominated by local source emissions, the effect of the economic crisis on BC concentration was more pronounced. For the three analysed years, BC concentrations obtained during winter were higher than those measured during summer, probably due to increased emissions from domestic heating and less intense vertical mixing in winter season, which lead to the confinement of the BC particles near the surface. The monthly mean BC concentrations were lower in 2008 than in 2006–2007 for almost every month of the year. In all years BC concentrations exhibited a clear diurnal pattern, with two maxima and two minima within a day. There were no differences among the daily patterns for 2006, 2007 and 2008 except for a general reduction in BC concentrations on 2008, especially during morning and evening traffic hours. For every day of the week, BC concentrations were lower on 2008 than in 2006 and 2007 and this reduction was more pronounced on working days, when BC concentrations were high.

1. Introduction

Black carbon is produced as primary particles from incomplete combustion processes, in particular from diesel engines in the transportation sector, which are the major source of black carbon in many European urban areas (Hamilton and Mansfield, 1991; Berner et al., 1996; Pakkanen et al., 2000; Bond et al., 2004). Its emissions depend on the efficiency of combustion as well as the amount and type of the consumed fuels, and any exhaust treatment (e.g. Wehner et al., 1999). For these reasons it's difficult to predict and must be measured (Chung and Seinfeld, 2002; Schaap et al., 2004).

In urban sites, black carbon (BC) is the principal particulate species that absorbs radiation in the visible spectrum (Bond and Bergstrom, 2006; Moosmüller et al., 2009). It is recognised that absorption by BC particles can greatly offset the direct radiative cooling effect of some pure scattering aerosol types (Schwartz, 1996; Haywood and Shine, 1997; Foster et al., 2007), and can even exceed greenhouse warming by gases other than carbon dioxide (Jacobson, 2001; Ramanathan and Carmichael, 2008). In 2007, the Intergovernmental Panel on Climate Change (IPCC) reported that BC contributes 0.2–0.4 Wm^{-2} to radiative forcing (Foster et al., 2007). However, in a recent study, Ramanathan and Carmichael (2008) reported that radiative forcing by BC contributes 0.9 Wm^{-2}, suggesting that BC may be the second strongest

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contributor to current global warming after carbon dioxide. Additionally, BC can have indirect radiative effects due to its action on cloud droplet number concentrations and related cloud properties (Ackerman et al., 2000; Kaufman et al., 2002). Nevertheless, the influence of BC particles on climate at regional scale is more significant than at global scale (Wang, 2004). According to Ramanathan et al. (2001), Menon et al. (2002), Ramanathan and Carmichael (2008), BC heats the air and changes regional atmospheric stability and vertical motions, resulting in alterations of the hydrologic cycle with significant regional climate impacts.

Furthermore, the absorption of sunlight by BC contributes to degraded visibility in polluted regions (Horvath, 1995). Being present mostly in the fine size range (Ruellan and Cachier, 2001), BC can have adverse effects on human health (Gurjar et al., 2010; Löndahl et al., 2010; Kim et al., 2003). Additionally, BC play an important role in atmospheric chemistry due to its porous and adsorptive nature, serving as a site for some of the chemical transformations (Fedel et al., 1995).

Depending on meteorological and geographical conditions, BC can reside in the atmosphere for days, unlike greenhouse gases such as carbon dioxide with lifetimes exceeding a hundred years. Due to the relatively short atmospheric lifetime of BC particles, controlling their emissions can be a quick way to slow global warming, in addition to improving human health (Jacobson, 2002).

Therefore, real time continuous and spatially representative measurements are required to update our understanding of the behaviour of BC particles and their impacts on climate, environment, and human health. However, real time continuous BC measurements in Spain are still limited and the sources and mechanisms that affect their concentrations in air are still ambiguous (Rodríguez and Cuevas, 2007; Rodríguez et al., 2007, 2008; Fernández-Camacho et al., 2010). The common finding from these studies is the large variability of atmospheric BC concentration and the large impact of road traffic emissions on daily and weekly evolution of atmospheric BC levels. According to them there is a need for the continuous long term monitoring of BC to determine the impact of any emissions reduction plans established or to be established in urban areas. So, additional studies based on long term BC measurements are necessary to detect trends and to estimate the impact of the changes in the source emissions.

In early 2008, the economic crisis impacted Spain and other states of the European community. This economic crisis has affected severely the productivity and employment in Spain. In addition, this economic crisis resulted in a contraction in capital investment and consumer behaviour and an associated decline in fuel consumption. Thus, due to the decline in fuels consumption it might be expected a decrease in BC emissions in the region.

At our knowledge, there are no studies in the literature about the impact of the economic crisis on the atmospheric BC levels. Recently, Arruti et al. (2011) have evaluated the economic crisis impact on some trace metal levels and particulate matter at an urban area in the Cantabria Region, Northern Spain. The main results obtained by these authors is that the impact of the crisis was higher on the PM composition, especially on the trace metal levels related to industrial activities, than on the PM levels.

In this work we study the seasonal, weekly and diurnal variations of BC concentrations at Granada, an urban site. In addition, we examine the effect of local meteorology and the patterns in anthropogenic activities to explain the atmospheric BC behaviour. In this sense, the marked reduction in fuels consumption experienced along 2008 offers a unique experiment to evaluate the direct effect of this reduction on the atmospheric BC concentrations.

2. Experimental site

The measurements presented in this study were registered at an urban site, Granada (37.16° N, 3.58° W, 680 m a.s.l), from 1 December 2005 to 30 November 2008. Granada, located in southeastern Spain, is a non-industrialised, medium-sized city with a population of 300,000, or 600,000 if the whole metropolitan area is considered. The city is situated in a natural basin surrounded by mountains with elevations between 1000 and 3500 m a.s.l. Near-continental conditions prevailing at this site are responsible for large seasonal temperature differences, with cool winters and hot summers. Most rainfall occurs during spring and winter.

The measuring station is located in the southern part of the city and is about 500 m away from the highway (A-44, E-902) that surrounds the city and about a similar distance from one of the principal traffic roads of the city "Camino de Ronda" (Fig. 1). There is no major industry in the vicinity of the measurement site. According to information from Spanish authorities (www.dgt.es), the total vehicle fleet at Granada was 576,596, 611,800 and 626,147 in 2006, 2007 and 2008, respectively. Vehicles with diesel engines accounted for about 50%, 51% and 53% of the total vehicles at Granada during 2006, 2007 and 2008, respectively. Vehicles fuelled with diesel consumed approximately 85% of transportation energy in Granada, according to data from Corporation of strategic Reserves of oil-based products (www.cores.es). Thus, according to the fact that there are no major industries in the area the main anthropogenic local source of BC particles at the measurement site is vehicle traffic, particularly those fuelled by diesel fuel. In winter, domestic heating (based on fuel oil combustion) represents an additional important source of BC aerosols.

3. Instrument

The BC concentrations were recorded with a Multi-Angle Absorption Photometer (MAAP) (Thermo ESM Andersen Instruments, Erlangen, Germany), based on the light absorption properties of BC particles. The MAAP simultaneously measures radiation transmitted through and scattered back from particles deposited on the filter and uses radiative transfer calculations to determine the aerosol absorption coefficient to correct for errors that affect other conventional filter based instruments (Petzold and Schönlinner, 2004). This explicit treatment of light scattering effects caused by aerosol and filters matrix improves considerably the determination of aerosol absorption over other filter based methods, as demonstrated by comparison with a reference method (Petzold et al., 2005; Sheridan et al., 2005). A detailed description of the method is given by Petzold and Schönlinner (2004). The MAAP determines the aerosol absorption coefficient at 637 nm, where absorption by organic aerosols and dust should be very small relative to that by BC (Sokolik and Toon, 1999; Kirchstetter et al., 2004; Alfaro et al., 2004; Linke et al., 2006; Barnard et al., 2008; Moosmüller et al., 2009). A mass absorption efficiency (MAE) of 6.6 $m^2\ g^{-1}$ – recommended by the manufacturer – was used in the internal computations to convert the measured absorption coefficient to BC concentration. Assuming a λ^{-1} dependence for absorption as suggested by Bergstrom et al. (2002) and Kirchstetter et al. (2004), this MAE increases to 7.64 $m^2\ g^{-1}$ at 550 nm. However, the MAE was found to vary with aerosol sources and aerosol ageing (Liousse et al., 1993). The MAE used in this study is slightly higher than the value of 7.5 ± 1.2 $m^2\ g^{-1}$ at 550 nm suggested for fresh, uncoated BC particles by Bond and Bergstrom (2006) and lower than 10 $m^2\ g^{-1}$ at 550 nm, a maximum value suggested for aged (i.e. coated) BC particles (Fuller et al., 1999; Bond et al., 2006a). Also, the MAE used here is closer to the range of 7.4–9.0 $m^2\ g^{-1}$ at 550 nm derived by different authors at urban sites in Mexico City (Doran, 2007;

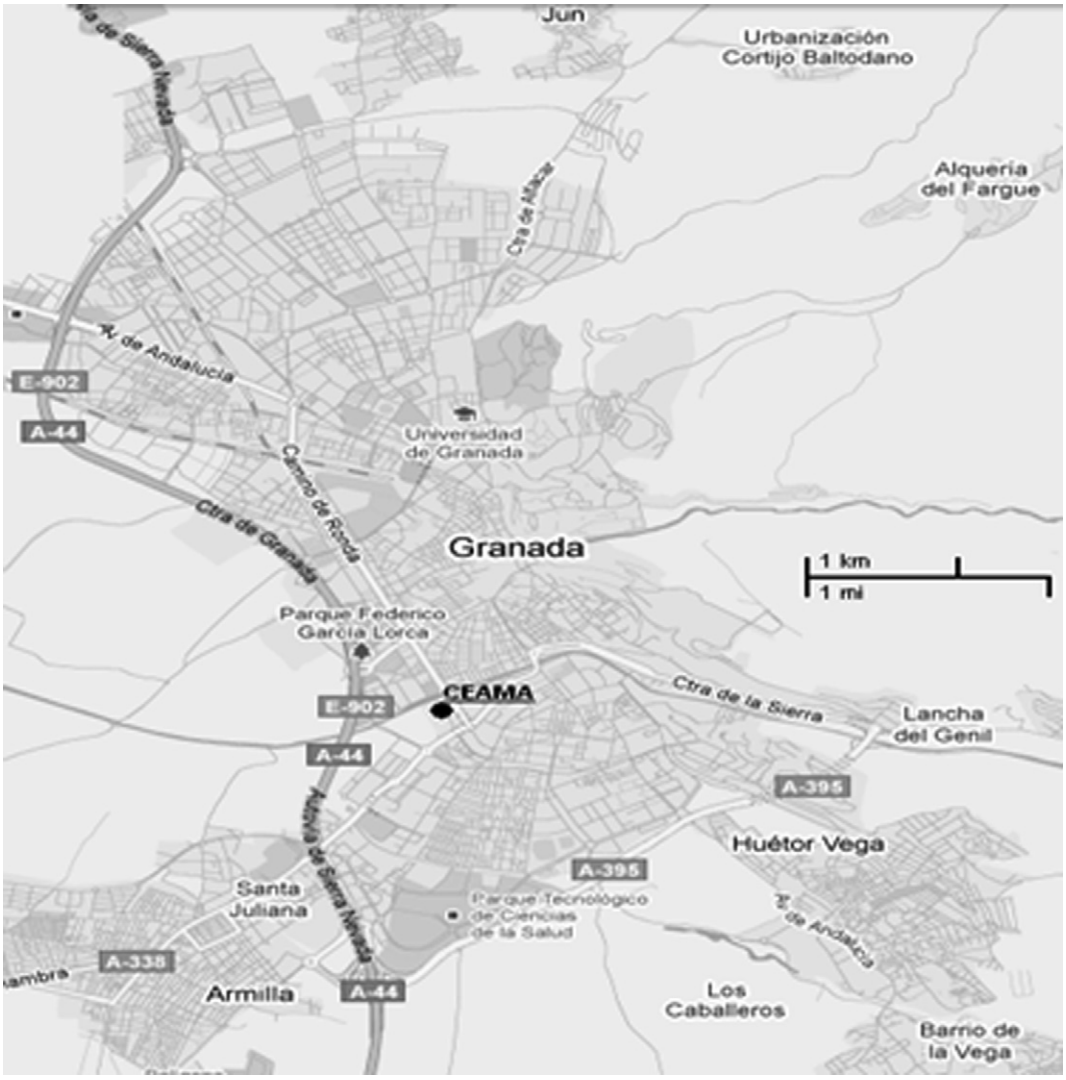

Fig. 1. Map of Granada showing the location of CEAMA monitoring site and the principal traffic roads of the city.

Barnard et al., 2007; Doran et al., 2008). Furthermore, this MAE compares reasonably well with the value ranging from 6.8 to 8.7 $m^2\ g^{-1}$ at 550 nm derived by Hitzenberger et al. (2006) at Vienna, Austria. The absolute measurement uncertainty of the MAAP has been estimated to be 12% (Petzold and Schönlinner, 2004) while laboratory inter-comparisons of multiple MAAP instruments suggested a unit-to-unit variability below 3% (Müller et al., 2011). The uncertainty in the BC mass concentration, including the uncertainty in the mass absorption coefficient, is estimated to be larger. The manufacturer's precision for the determination of BC is $<0.1\ \mu g\ m^{-3}$ with an integration time of 120 s.

Air sampling for the MAAP and other instrumentation operated side by side in our laboratory has been described in Lyamani et al. (2008, 2010). The air sampling was drawn from 15 m above the street level through a straight, vertical, stainless-steel tube of 20-cm diameter and 5-m length at a constant flow rate of 1000 $l\ h^{-1}$. The inlet is fitted with a funnel and covered by an insect screen to prevent rain and insects from entering the sample line. Measurements were performed with no aerosol size cut-off and no heating was applied to the sampled air (Lyamani et al., 2010). The diameters of stainless pipe were adjusted to maintain the laminar flow in the tube and minimise particle losses (Baron and Willeke, 2001).

Meteorological variables, including wind speed and temperature, were measured by an automatic weather station at the sampling site using a Campbell CR10X dataloger. The wind velocity was measured, at the same height of the sampling inlet, by a wind monitor (model 05103 R.M. Young) and the temperature was recorded by temperature sensor model MTH-A1. The error in temperature measurements is 0.4 °C over the range −40 °C to +110 °C and the error in wind velocity is 0.3 $m\ s^{-1}$ over the range 1–60 $m\ s^{-1}$. Meteorological data were recorded as 1 min averages, and subsequently processed to hourly means.

Source regions of air masses affecting a given area can be determined by performing a backward trajectory analysis. To identify the origin of the air masses arriving at our study area, 5-day backward trajectories ending at 12 UTC at 500 m over Granada ground level were calculated using the HYSPLIT-4 model (Draxler and Rolph, 2003). The model version employed uses GDAS Meteorological data (Global Data Assimilation System, ftp://www.arl.noaa.gov/pub/archives/gdas1/). In previous studies we showed that long range transport has a significant impact on the aerosol properties at Granada (Lyamani et al., 2008, 2010). Air masses originated in the Atlantic have been reported to be associated with low aerosol load in the study area while those originated in North Africa and/or Europe were found to relate with high aerosol load. To

determine the influence of air mass origin on BC concentration, BC measurements days have been classified according to their 120 h back-trajectories following the classification scheme developed by Toledano et al. (2009). This classification method is based on the residence time of particular trajectory within geographic sectors. We have defined five major air mass sectors: Atlantic Ocean (Western), European Continent (Northern), Mediterranean Sea (Eastern), North Africa (Southern) and the Local one (see Fig. 2).

4. Results

4.1. BC concentration annual variations

BC data were recorded as 1 min averages, and subsequently processed to hourly means. Fig. 3 shows the temporal evolution of daily average BC concentrations measured during the period from December 2005 to November 2008 at the study site. The daily average BC data are calculated from hourly average BC data. About 2% of the data are missing due to the participation of the MAAP in two measurement campaigns (Silva et al., 2007; Martínez-Lozano et al., 2007; Córdoba-Jabonero et al., 2011), in addition to instrument maintenance.

Daily average BC concentrations for the three years period varied by more than 16 fold from 0.5 μg m^{-3} to 8.6 μg m^{-3}, with overall mean value and standard deviation of 3.0 ± 1.5 μg m^{-3}. The large standard deviation reflects the strong day-to-day variation in the BC concentrations. This large variability in BC concentrations is related to variations in emissions sources and meteorological conditions. The reported average BC concentrations in Spain include the mean value of 1.9 μg m^{-3} obtained in a coastal city (Santa Cruz de Tenerife, Canary Islands) during 17 March-10 April 2006 (Rodríguez et al. (2008)), hourly mean values ranging from 0 to 8 μg m^{-3} measured in Huelva city during the period April 2008–September 2009 (Fernández-Camacho et al., 2010) and a mean BC concentration of 3.6 μg m^{-3} found in Barcelona during July–November 2007 (Peréz et al., 2010). It's interesting to note that the differences in the BC concentrations between these sites may result from different meteorological conditions, different period of measurements and differences in sampling site characteristics.

On the other hand, BC concentrations showed a slight decreasing tendency. Annual average BC concentrations were quite similar during 2006 and 2007 (3.2 ± 1.4 μg m^{-3} and 3.1 ± 1.6 μg m^{-3}, respectively), but experienced a decrease of approximately 16–18% to 2.6 + 1.4 μg m^{-3} on 2008. This reduction is not only observed in the annual mean value, but also in the median, third and first quartiles (Figure not shown). A Mann–Whitney test at 0.05 significance level reveals that the BC concentration difference between 2006 and 2007 is statistically no significant, whereas that for 2008 was significantly different from both 2006 and 2007. Furthermore, this test confirms that BC concentration in 2008 tends to be less than in 2006–2007.

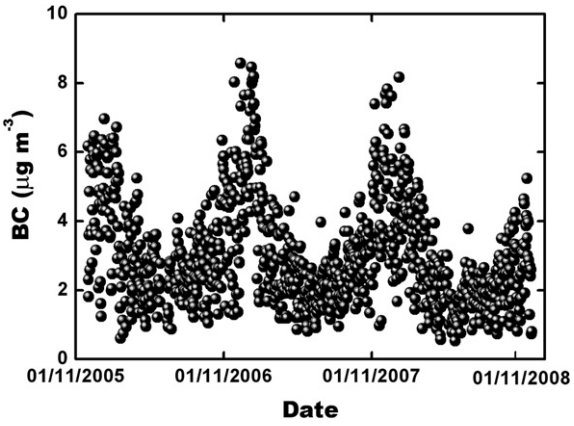

Fig. 3. Time series for daily average BC concentration measured at Granada from December 2005 to November 2008.

Meteorological conditions and emissions are competitive processes determining the magnitudes of pollutant concentrations in the atmosphere (e.g. Ruellan and Cachier, 2001). Thus, the reduction in the atmospheric BC concentrations in Granada during 2008 could be related to a decrease in BC emissions or to a drastic change in the meteorological conditions, or to both. In the next section we investigate the causes of this reduction in atmospheric BC.

4.2. Factors influencing black carbon concentrations

In this section we investigate if the change in meteorological and synoptic conditions is the cause of the reduction of BC concentration in 2008. Fig. 4 shows the temporal evolutions of daily average wind speed and rainfall measured during the period from December 2005 to November 2008 at the study site. Table 1 presents statistical summary of daily average of wind speed, temperature and rainfall and number of rainy days in 2006, 2007 and 2008. As we can see from Fig. 4 and Table 1, there were no drastic changes in the meteorological conditions during the analysed period.

In a previous study we showed that rainfall events can have a significant impact on the aerosol measured at surface at Granada, leading to the reduction of aerosol concentrations at surface (Lyamani et al., 2008). For that reason, we check if the precipitation was one of the factors responsible of atmospheric BC concentrations reduction in 2008.

Annual rainfall and the number of rainy days were relatively larger in 2008 than in 2007, but were lower compared to 2006 (Table 1). So, one may suspect that the increase in precipitation in 2008 has induced a decrease in the atmospheric BC concentration compared to 2007. To check this possibility we calculated the BC concentrations in the rainy days and in the days without precipitation during 2006, 2007 and 2008. Excluding rainy days, mean BC concentrations in 2006 and 2007 were 3.3 ± 1.4 and 3.2 ± 1.7 μg m^{-3}, respectively and decreased by 15–18% to 2.7 ± 1.4 μg m^{-3} in 2008. Considering only rainy days, mean BC concentrations were 2.7 ± 1.5 and 2.7 ± 1.2 μg m^{-3} in 2006 and 2007, respectively, and also decreased by 15% to 2.3 ± 1.2 μg m^{-3} in 2008. So it is evident that there was reduction in the annual average value of BC that affects both no rainy and rainy days. In this sense, the decrease of

Fig. 2. Map of the Iberian Peninsula illustrating the geographical location and the classification of the different origins of the air masses influencing the area of study.

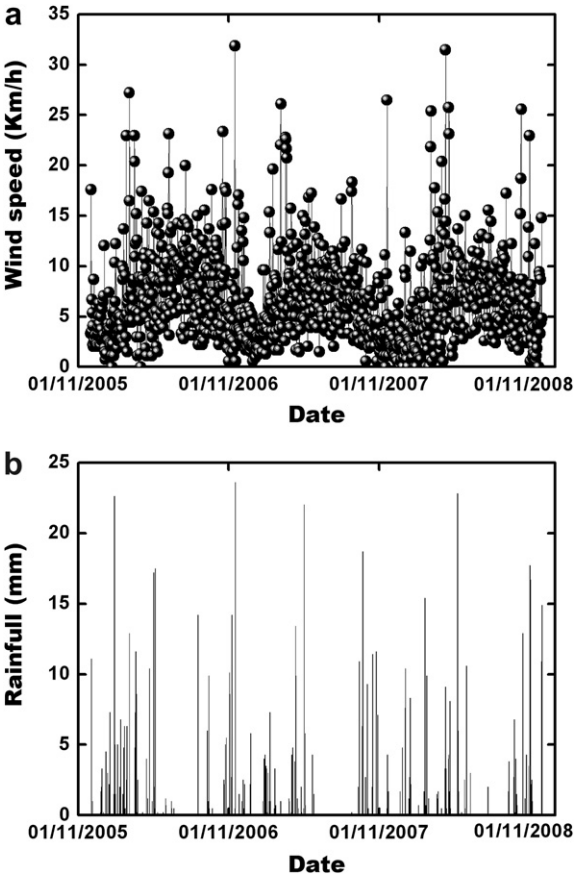

Fig. 4. Evolution of daily mean values of (a) wind speed and (b) rainfall, measured at Granada from December 2005 to November 2008.

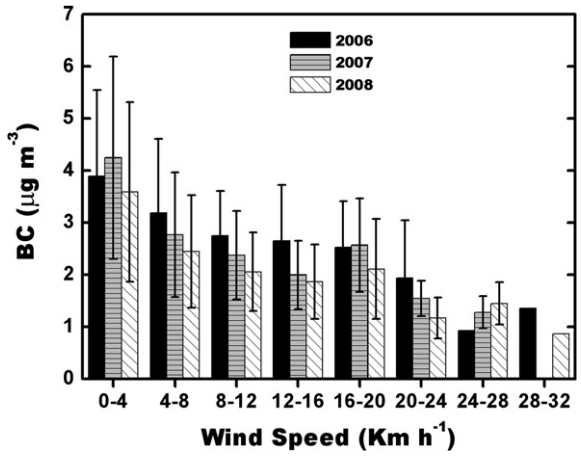

Fig. 5. BC concentrations as function of wind speeds for 2006, 2007. The error bars are standard deviations.

0.4 μg m^{-3} we determine in the last case cannot be explained as a result of precipitation scavenging. So, the inter-annual variation in precipitation is not responsible for the reduction of BC in 2008.

High wind speeds have been reported to relate with low aerosol concentrations in the study area (Lyamani et al., 2008). Therefore, we investigate if the reduction of BC concentrations in 2008 was caused by change in wind speed.

In general, mean wind speeds in the study area were very similar in 2006 and 2007, but slightly lower in 2008 (Table 1). In addition, low wind speeds (<8 km h^{-1}) were slightly more frequent in 2008 than in 2006–2007 while relatively higher wind speeds were less frequent in 2008 than in 2006–2007. As a result, one may expect higher BC concentrations in 2008 than in 2006 and 2007. However, as can be seen from Fig. 5, it is very clear that for most wind speed classes mean BC concentrations in 2008 were lower compared with 2006–2007. The low wind speed class 0–4 km h^{-1} is the most representative of Granada local emissions, being less influenced by regional and long range transport. The frequencies in this wind speed class were 29.6, 31.7 and 30.5% in 2006, 2007 and 2008, respectively. Wind speeds in this low wind speed class were more frequent in 2007 than 2006 and as expected the BC concentrations in this low wind speed class increased by 9% in 2007 versus 2006. Nevertheless, although wind speeds in this class were somewhat more frequent in 2008 than in 2006, the BC concentrations in 2008 decreased by 8% with respect to 2006. Thus, the results indicate that the inter-annual variation in wind speed is not responsible for the reduction of BC in 2008.

Now we investigate if the BC concentrations reduction in 2008 was caused by change in air mass patterns between years 2006, 2007 and 2008.

In Fig. 6a the annual frequencies of air mass types that affected our study area during 2006, 2007 and 2008 are shown. The air mass types were classified according to the sector classification method proposed by Toledano et al. (2009). As we can see from this figure there were a significant change in air masses patterns between years 2006, 2007 and 2008. During 2006, most of the air masses correspond to Northern sector (29%), while those originated from local and Mediterranean Sea were less frequent (12%). A rather significant fraction (23%) corresponds to air masses from southern and western directions. During 2007, the situation changed and the air masses originated in the southern sector were the most frequent (40%) followed by those originated in the Northern sector (28%) and Western sector (16%). The air masses coming from the Eastern and Local sectors were less frequent in this year (6% and 8%, respectively). During 2008, the situation changed again and most of the air masses originated from southern sector (30%). Air masses from Northern and Western sectors were also quite frequent (26%) on 2008. As during 2006 and 2007, air masses from Eastern and Local sectors were less frequent on 2008 (8% and 9%, respectively). In order to examine if the BC concentrations reduction in 2008 was related to the change in air mass patterns, in Fig. 6b we present BC concentrations observed at Granada as function of the origin of air masses for 2006, 2007 and 2008. Obviously, BC concentrations are strongly affected by the air mass origin. In each year of the analysed period, the highest BC concentrations were observed for local air masses and the lowest were obtained during Atlantic air masses arrivals. Intermediate BC concentrations were associated to Southern, Eastern and Northern advections. This suggests that local sources, dominated by traffic, contributed to a large fraction of BC concentrations in Granada during the study period. Another interesting feature evident in Fig. 6b is that, for all air masses types, BC concentrations in 2008 were lower compared with 2006 and 2007, suggesting a regional and local decrease in BC concentrations

Table 1
Annual mean values and standard deviations of temperature, wind speed and rainfall and number of rainy days in 2006, 2007 and 2008. The ranges in parentheses represent the daily average minimum and maximum values.

	Mean ± standard deviation		
	2006	2007	2008
Temperature (°C)	17 ± 8 (1–35)	17 ± 7 (1–32)	17 ± 7 (3–32)
Wind speed (km h^{-1})	7 ± 5 (0–31)	7 ± 4 (0–27)	6 ± 5 (0–31)
Rainfall (mm)	330 (0.2–23.0)	217 (0.2–22.0)	289 (0.2–23.0)
Number of rainy days	69	57	62

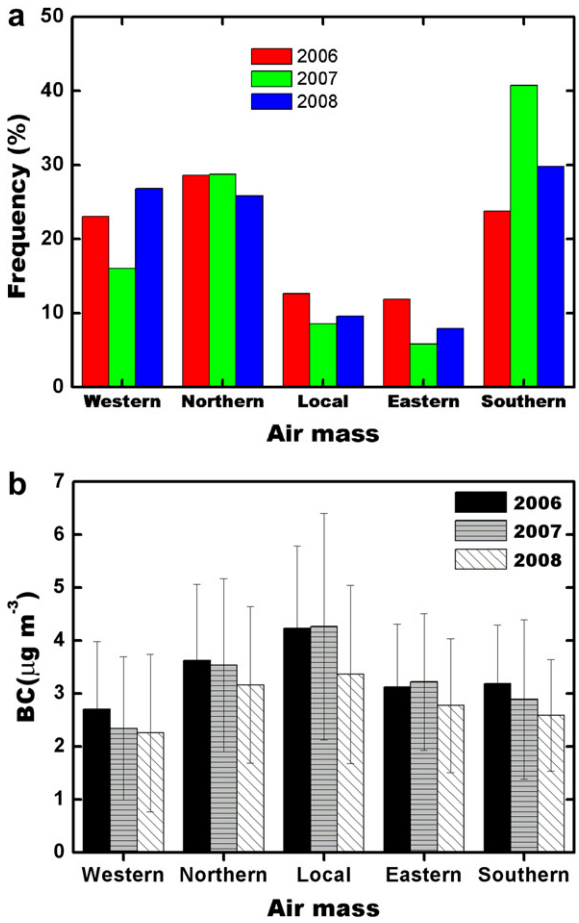

Fig. 6. BC (a) Annual frequencies of air mass types that affected Granada during 2006, 2007 and 2008 and (b) concentrations as function of air mass origin for 2006, 2007 and 2008. The error bars are standard deviations.

in 2008. For local air masses class, where local sources are dominant, the reduction in BC concentration from 2006 and 2007 to 2008 was the largest (21%). For air mass arriving from Atlantic the relative reduction in BC concentration from 2007 to 2008 was the lowest (3.7%). These results suggest that variations in inter-annual long range transport cannot be solely responsible for the BC concentrations reduction in 2008.

Thus, the reduction in BC concentrations in 2008 is probably associated with a decrease in local and regional emissions of BC particles, rather than to changes in meteorological conditions.

It should be noted that the quantity of anthropogenic BC particles emitted by fossil fuel combustions depends on how efficiently the fuel is burned in addition to the amount and type of consumed fuel (Bond et al., 2006b). According to data provided by Corporation of strategic Reserves of oil-based products (www.cores.es), diesel fuel accounted for 85.5% (85.3% in 2007) and gasoline fuel for 14.5% (14.7% in 2007) of total consumption of fuels at Granada during 2008, indicating no significant change in the type of fuels consumed. Nevertheless, as a result of the economic crisis that started in early 2008, the fuels consumption during this year at Granada, and at Spain in general, decreased with respect to 2007 (www.cores.es). In fact, according to data provided by the Corporation of strategic Reserves of oil-based products, the economic decline dragged Spain's oil products consumption down by 3.3% in 2008 (www.cores.es). Also, due to this economic crisis diesel and gasoline fuels consumption in Spain in 2008 decreased by 3.3% and 6%, respectively, with respect to 2007. At Granada, diesel fuel consumption was 662,494 tonnes in 2007 and decreased by about 6% to 618,643 tonnes in 2008, while gasoline fuel consumption was 124,466 tonnes in 2007 and decreased by 8%–114,467 tonnes in 2008. Furthermore, diesel and gasoline fuel consumption was lower in 2008 than in 2007 for almost every month of the year (www.cores.es). In addition, the consumption of fuel oil used in domestic heating was 58,998 tonnes in 2007 and also decreased by 6% to 55,413 tonnes in 2008. Thus, assuming that no appreciable change in emissions controls has occurred at Granada and at Spain in general during the analysed period, the decline in the fuel consumption during 2008 would results in a decrease in BC emissions. Therefore, the reduction in atmospheric BC concentrations during 2008 can be partly explained by the decrease in BC emissions, due to the decrease in fuel consumption in Granada and in Spain in general as a result of the economic crisis. Furthermore, due to this economic crisis the emissions of other pollutants in Spain in 2008 have also decreased markedly, according to the pollutants emissions inventory report of the Ministry of Environment (MAMR, 2010). According to this report, the emissions of carbon monoxide, CO, nitrogen oxides, NO_x, sulphur dioxide, SO_2, particle matter, PM_{10} and $PM_{2.5}$, have been decreased by 4.2%, 12.2%, 54.6%, 8.7% and 6.8%, respectively, in 2008 compared to 2007, which would leads to a decrease in the levels of these pollutants in the atmosphere. Thus, these results suggest that the economic crisis has a positive impact on reducing potential human exposures to BC and other pollutants. In addition, the results also indicate that the reduction in BC emissions results in a rapid reduction in its level in the atmosphere.

4.3. Seasonal variation of black carbon concentrations

Besides the inter-annual variation, BC concentrations also exhibited clear seasonal variation, evident in Fig. 7, which presents monthly BC concentrations observed at Granada during the years 2006, 2007 and 2008. The Monthly mean BC concentrations were relatively high in winter (December–February) and autumn (September–November) and low in summer (June–August) and spring (March–May), with the highest concentrations in winter and the lowest in summer. This BC concentration seasonal cycle persisted through the three-year observation period (Fig. 7). The seasonally-averaged BC concentrations for the entire analysed period were 4.4 ± 1.7, 2.4 ± 0.9, 2.0 ± 0.7 and 3.1 ± 1.3 μg m^{-3} for winter, spring, summer and autumn, respectively. The average BC concentration in winter was more than twice that computed in summer. The winter average BC concentration (4.4 ± 1.7 μg m^{-3}) was 48% higher than the three-year average (3.0 ± 1.5 μg m^{-3}) whereas the summer average BC concentration (2.0 ± 0.7 μg m^{-3}) was 33% lower than the overall mean, suggesting that the health risk due to BC exposure in winter is higher than in summer. This seasonal variation in BC concentrations is similar to the seasonal variations in other aerosol properties observed in the study area (Lyamani et al., 2010). These seasonal variations are likely due to a combination of changes in emissions rates and seasonal meteorology (Lyamani et al., 2010). Similar seasonal variations in BC concentrations have been observed in other urban areas (e.g. Ramachandran and Rajesh, 2007; Yttri et al., 2007; Kirchstetter et al., 2008; Cao et al., 2009; Saha and Despiau, 2009), with higher BC concentrations in winter. Theses authors attributed the high BC concentrations observed in winter to the increase in BC emissions due to the increase in anthropogenic activities associated with domestic heating and to unfavourable meteorological conditions like shallow atmospheric boundary layer and low wind speed. Furthermore, the monthly mean BC concentrations were lower in 2008 than in 2006 and 2007 for almost every month of the year as can be seen clearly in Fig. 7. The decrease in BC concentrations in

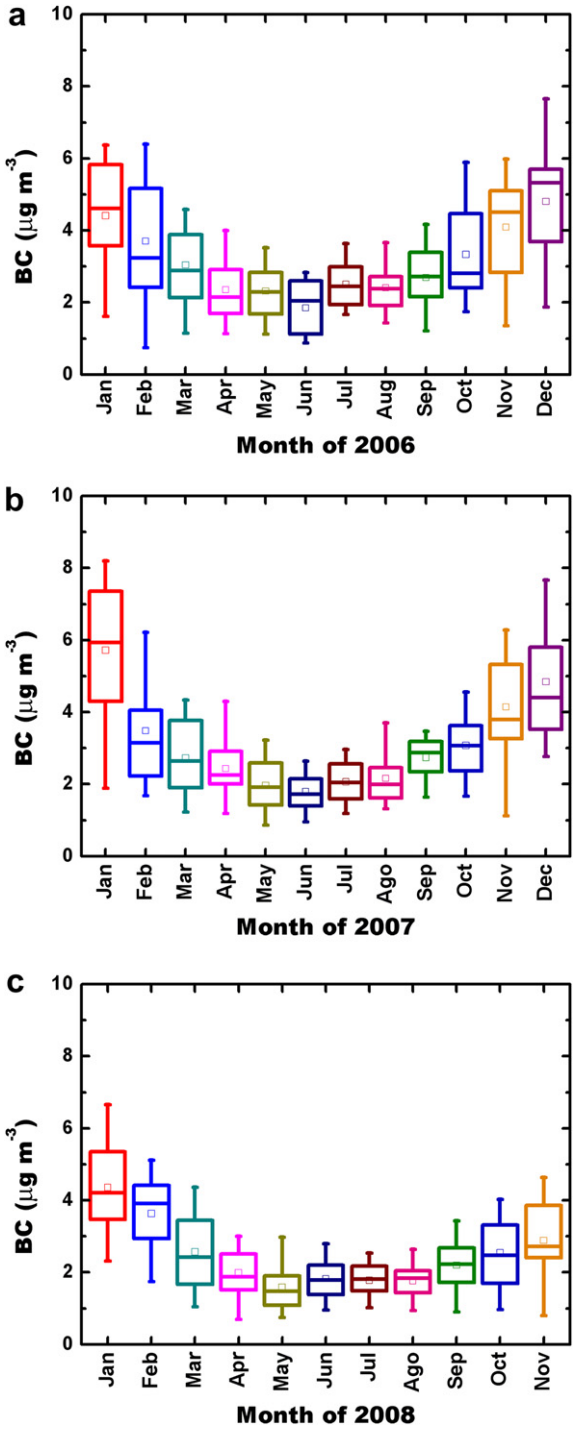

Fig. 7. Monthly mean black carbon concentrations measured at Granada for: (a) 2006, (b) 2007 and (c) 2008. Line within box is the monthly median value; square symbol is the monthly mean value; Top and Bottom of box are the 25th and 75th percentiles, respectively; ends of the whiskers are the 5th and 95th percentiles, respectively.

2008 was reflected not only in the monthly mean BC but also in the high BC levels, as indicated by 75th and 95th percentiles.

4.4. Diurnal and weekly variations in BC concentrations

Fig. 8 shows the mean diurnal variation of BC concentrations measured in Granada in 2006, 2007 and 2008. As a common feature, all years exhibited a clear diurnal pattern of BC concentrations, with

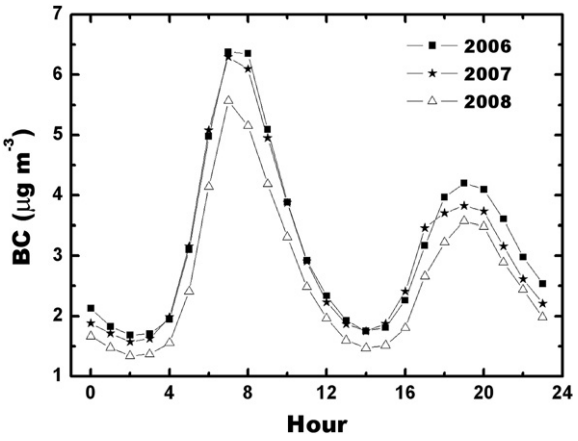

Fig. 8. Annually mean diurnal variations of BC concentrations for 2006, 2007 and 2008. Each point represents the average BC concentration for that hour for the respective years.

two maxima and two minima within a day. The BC concentration started to increase gradually from early morning around 4 GMT, reaching a sharp peak around 7–8 GMT. Then decreased and attained the diurnal minimum in the afternoon around 14 GMT. During late afternoon BC concentration increased again and reached maximum around 19–20 GMT. The morning and evening peaks in BC concentrations are primarily related to the elevated road traffic emissions during the morning and evening traffic rush hours. On other hand, the steady decrease in BC concentrations after the morning peak can be attributed to the enhanced vertical and horizontal diffusion of aerosols, due to gradual increase in boundary layer height and wind speed, in addition to the relative decrease in traffic. A drastic decrease in traffic, the main source of BC in the study area, after around 23 h may be responsible for low BC concentrations observed from midnight to early morning. Similar diurnal variations in BC concentrations have been also observed in other Spain urban areas (Rodríguez and Cuevas, 2007; Rodríguez et al., 2007, 2008; Fernández-Camacho et al., 2010; Peréz et al., 2010) and in worldwide urban sites (Ruellan and Cachier, 2001; Latha and Badarinath, 2005; Baumgardner et al., 2007; Safai et al., 2007; Saha and Despiau, 2009; Dutkiewicz et al., 2009).

An interesting feature of Fig. 8 is that diurnal variations in BC were similar in 2006, 2007 and 2008, with similar timing of the peaks and minima, indicating similar features in source emissions

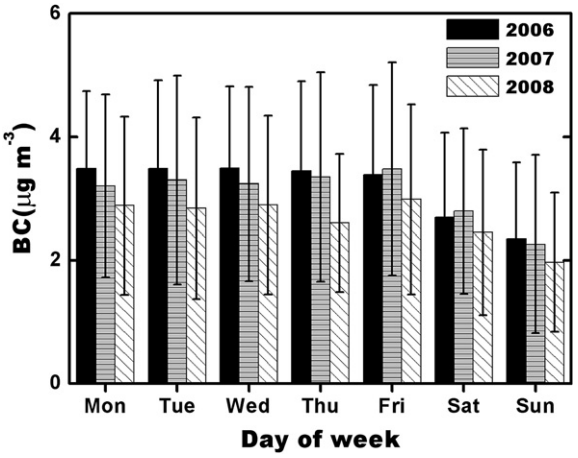

Fig. 9. Mean weekly variations of BC concentrations measured in Granada for 2006, 2007 and 2008. The error bars are standard deviations.

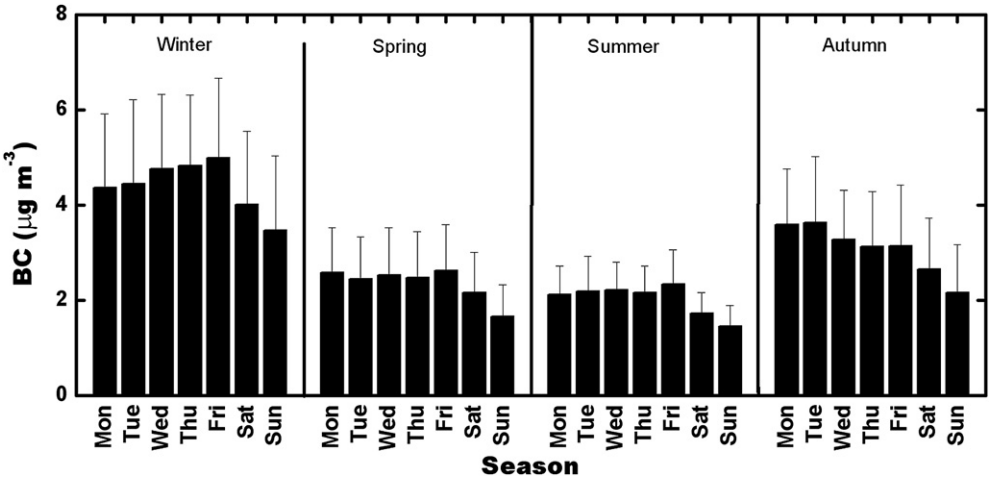

Fig. 10. Seasonal weekly variations of BC concentrations measured in Granada for the entire study period 2006–2008. The error bars are standard deviations.

and atmospheric boundary layer dynamics. However, it is evident that at every hour of the day the BC concentrations were always lower in 2008 compared with 2006 and 2007 (Fig. 8). This reduction was more pronounced during morning and evening traffic rush hours, when BC concentrations were highest. A Mann–Whitney test at 0.05 significance level shows that in the morning (6–10 GMT) and evening (18–21 GMT) rush hours the BC concentration difference between 2006 and 2007 is statistically no significant and that BC concentration in 2008 in these rush hours tends to be less than in 2006–2007. Therefore, the reduction in BC concentrations in 2008 was most likely due to a decrease in the emissions sources (e.g. traffic).

Fig. 9 shows the average BC concentrations by day of week measured in Granada for 2006, 2007 and 2008. BC concentrations in the study area showed a pronounced weekly cycle, which persisted throughout the 3-years observation period. In all years, the BC concentrations were higher on workdays (Monday–Friday) and lower on weekends (Saturday and Sunday), with the lowest BC concentrations occurring on Sunday. The weekend BC concentrations reductions remained relatively constant at about 30% in the 3-years analysed period. This significant reduction in BC concentrations during weekends is related to a decrease in human activities and commuter traffic during weekends due to holidays for offices, schools. A weekly cycle for BC concentrations with a Sunday minimum has also been found in other urban areas (Järvi et al., 2008; Kirchstetter et al., 2008).

This weekly feature was observed in all seasons as can be seen in Fig. 10. It is interesting to note that the relative weekly variation of BC concentration is higher in summer, when the average working day BC concentration was 1.5 times higher than on weekends, while in winter this ratio decrease to 1.2. However, concerning the absolute differences we can see that in winter the average working days BC concentration was 0.89 μg m^{-3} higher than on weekends, while in summer this difference decrease to 0.69 μg m^{-3}. This seasonal difference in the weekly cycle is due to the high BC baseline concentration in winter due to the increase in the anthropogenic activities associated with domestic heating. Furthermore, in winter the reduction of the urban activity during the weekends affects not only to the traffic but also to the central heating use in working spaces in the city. Kirchstetter et al. (2008) also observed significant seasonal change in weekly BC concentrations at California, USA.

Besides a weekly cycle, the BC concentrations on every day of the week in 2008 were lower compared to those in 2006 and 2007 (Fig. 9). Again we have applied a Mann–Whitney test at 5% significance level to compare BC concentrations in working days obtained in 2006, 2007 and 2008. We also applied this test to weekends BC data obtained on these three years. This test confirms that BC concentrations in working days as well as in weekends in 2008 tend to be less than in 2006–2007. On working days, the mean BC concentrations were quite similar in 2006 and 2007 (3.5 ± 1.4 μg m^{-3}, and 3.3 ± 1.6 μg m^{-3}, respectively) and decreased by 15–20% to 2.8 ± 1.4 μg m^{-3} in 2008. On weekends, the mean BC concentrations were the same (2.5 ± 1.4 μg m^{-3}) during 2006 and 2007, respectively, and decreased by 12% to 2.2 ± 1.3 μg m^{-3} in 2008. Thus, the decrease in BC concentrations in 2008 was more pronounced during working days, when traffic emissions are expected to be higher. Järvi et al. (2008) have analysed the effect of different meteorological variables (wind speed, temperature, mixing height etc.) and traffic on BC concentrations measured during working days and weekends. These authors showed that on working days the traffic had clearly the most important influence on BC concentrations, whereas on weekends the wind speed had the strongest influence. Thus, the decrease in BC concentrations in 2008 was most likely due, at least partly, to the reductions in traffic-related emissions.

5. Conclusions

In this study we have investigated BC concentrations in an urban area, Granada (Spain), from December 2005 to November 2008. The daily mean BC concentrations showed considerable day-to-day variations and were found to vary from 0.5 μg m^{-3} to 8.6 μg m^{-3}, with an overall mean and standard deviation of 3.0 ± 1.5 μg m^{-3}. This large variability in BC concentrations was related to variations in emissions sources and meteorological conditions.

Annual BC concentrations were quite similar during 2006 and 2007 (3.2 ± 1.4 μg m^{-3} and 3.1 ± 1.6 μg m^{-3}, respectively), but experienced a decrease of approximately 16–18% to $2.6 + 1.4$ μg m^{-3} in 2008 compared to the two previous years. This reduction was observed, not only in the mean value, but also in the median, third and first quartile. In addition, the reduction in BC concentrations in 2008 was observed for almost every month of the year. A Mann–Whitney test at 0.05 significance level shows that the BC concentration difference between 2006 and 2007 is statistically no significant and that BC concentration in 2008 tends to be less than in 2006–2007. After checking the role of meteorological parameters in the BC concentrations, we can state that the reduction in BC

mass concentrations in 2008 is a result of the decrease in traffic-related emissions. Thus the reduction in BC concentration is directly related to the decline in fuels use as a result of economic crisis, rather than to a drastic change in meteorological conditions. The impact of the economic crisis on BC concentrations was more pronounced under conditions of local source dominance.

Furthermore, the BC concentrations in winter were twice those registered in summer, likely due to higher emissions caused by domestic heating, less intense vertical mixing, and lower wind speeds. This evident BC seasonal cycle persisted through the three-year observation period. Besides the seasonal variation, in all studied years BC concentrations also exhibited a pronounced diurnal variation, with two local maxima occurring in early morning and late evening. This diurnal cycle is mainly attributed to the diurnal evolution of the atmospheric boundary layer and to local anthropogenic activities. A strong weekly cycle was also evident, with weekend BC concentrations significantly lower than on weekdays, due to decreased traffic on weekend. There were no differences between the daily and weekly patterns between 2006, 2007 and 2008, except for generally lower BC concentrations in 2008.

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References

Ackerman, A.S., Toon, O.B., Stevens, D.E., Heymsfield, A.J., Ramanathan, V., Welton, E.J., 2000. Reduction of tropical cloudiness by soot. Science 288, 1042–1047.

Alfaro, S.C., Lafon, S., Rajot, J.L., Formenti, P., Gaudiche, A., Maillé, M., 2004. Iron oxides and light absorption by pure desert dust: an experimental study. Journal of Geophysical Research 109, D08208. doi:1010.29/2003JD004374.

Arruti, A., Fernández-Olmo, I., Irabien, A., 2011. Impact of the global economic crisis on metal levels in particulate matter (PM) at an urban area in the Cantabria Region (Northern Spain). Environmental Pollution 159, 1129–1135.

Baron, P.A., Willeke, K., 2001. Aerosol Measurement. Principles, Techniques, and Applications. John Wiley and Sons, Inc., ISBN 0-471-35636-0.

Barnard, J.C., Kassianov, E.I., Ackerman, T.P., Johnson, K., Zuberi, B., Molina, L.T., Molina, M.J., 2007. Estimation of a "radiatively correct" black carbon specific absorption during the Mexico City Metropolitan Area (MCMA) 2003 field campaign. Atmospheric Chemistry and Physics 7, 1645–1655.

Barnard, J.C., Volkamer, R., Kassianov, E.I., 2008. Estimation of the mass absorption cross section of the organic carbon component of aerosols in the Mexico City Metropolitan Area (MCMA). Atmospheric Chemistry and Physics 8, 6665–6679.

Baumgardner, D., Kok, G.L., Raga, G.B., 2007. On the diurnal variability of particle properties related to light absorbing carbon in Mexico City. Atmospheric Chemistry and Physics 7, 2517–2526.

Bergstrom, R.W., Russell, P.B., Hignett, P., 2002. Wavelength dependence of absorption of black carbon particles: predictions and results from the TARFOX experiment and implications for the aerosol single scattering albedo. Journal of Atmospheric Sciences 59, 567–577.

Berner, A., Sidla, S., Galambos, Z., Kruisz, C., Hitzenberger, R., ten Brink, H.M., Kos, G.P.A., 1996. Modal character of atmospheric black carbon size distributions. Journal of Geophysical Research 101, 19559–19565.

Bond, T.C., Street, D.G., Yarber, K.F., Nelson, S.M., Woo, J.H., Klimont, Z., 2004. A technology-based global inventory of black and organic carbon emissions from combustion. Journal of Geophysical Research 109, D14203. doi:10/1029/2003JD003697.

Bond, T.C., Bergstrom, R.W., 2006. Light absorption by carbonaceous particles: an investigative review. Aerosol Science Technology 40, 27–67.

Bond, T.C., Habib, G., Bergstrom, R.W., 2006a. Limitations in the enhancement of visible light absorption due to mixing state. Journal of Geophysical Research 111, D20211. doi:10.1029/2006JD007315.

Bond, T.C., Wehner, B., Plewka, A., Wiedensohler, A., Heintzenberg, J., Robert, J., Charlson, R.J., 2006b. Climate-relevant properties of primary particulate emissions from oil and natural gas combustion. Atmospheric Environment 40, 3574–3587.

Cao, J., Zhu, C., Chow, J.C., Watson, J.G., Han, Y., Wang, G., Shen, Z., An, Z., 2009. Black carbon relationships with emissions and meteorology in Xi'an, China. Atmospheric Research 94, 194–202.

Chung, S.H., Seinfeld, J.H., 2002. Global distribution and climate forcing of carbonaceous aerosols. Journal of Geophysical Research 107 (D19), 4407. doi:10.1029/2001JD001397.

Córdoba-Jabonero, C., Sorribas, M., Guerrero-Rascado, J.L., Adame, J.A., Hernández, Y., Lyamani, H., Cachorro, V., Gil, M., Alados-Arboledas, L., Cuevas, E., de la Morena, B., 2011. Synergetic monitoring of Saharan dust plumes and potential impact on surface: a case study of dust transport from Canary Islands to Iberian Peninsula. Atmospheric Chemistry and Physics 11, 3067–3091.

Doran, J.C., 2007. Corrigendum to "The T1–T2 study: evolution of aerosol properties downwind of Mexico City" published in Atmospheric Chemistry and Physics 7, 1585–1598, 2007. Atmospheric Chemistry and Physics 7, 2197–2198.

Doran, J.C., Fast, J.D., Barnard, J.C., Laskin, A., Desyaterik, Y., Gilles, M.K., 2008. Applications of Lagrangian dispersion modeling to the analysis of changes in the specific absorption of elemental carbon. Atmospheric Chemistry and Physics 8, 1377–1389.

Draxler, R.R., Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model Access via NOAA ARL READY. NOAA Air Resources Lab, Silver Spring, MD. Website: http://www.arl.noaa.gov/ready/hysplit4.html.

Dutkiewicz, V.A., Alvi, S., Ghauri, B.M., Choudhary, M.I., Husain, L., 2009. Black carbon aerosols in urban air in South Asia. Atmospheric Environment 43, 1737–1744.

Fedel, W., Matter, D., Burtscher, H., Schmidt-Ott, A., 1995. Interaction between carbon or iron aerosol particulates and ozone. Atmospheric Environment 29, 967–973.

Fernández-Camacho, R., Rodríguez, S., de la Rosa, J., Sánchez de la Campa, A.M., Viana, M., Alastuey, A., Querol, X., 2010. Ultrafine particle formation in the inland sea breeze airflow in Southwest Europe. Atmospheric Chemistry and Physics 10, 9615–9630.

Foster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D.W., Haywood, J., Lean, J., Lowe, D.C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., Van Dorland, R., 2007. Changes in atmospheric constituents and in radiative forcing. In: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K.B., Tignor, M., Miller, H.L. (Eds.), Climate Change 2007. The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Fuller, K.A., Malm, W.C., Kreidenweis, S.M., 1999. Effects of mixing on extinction by carbonaceous particles. Journal of Geophysical Research 104, 15941–15954.

Gurjar, B.R., Jain, A., Sharma, A., Agarwal, A., Gupta, P., Nagpure, A.S., Lelieveld, J., 2010. Human health risks in megacities due to air pollution. Atmospheric Environment 44, 4606–4613.

Hamilton, R.S., Mansfield, T.A., 1991. Airborne particulate elemental carbon: its sources, transport and contribution to dark smoke and soiling. Atmospheric Environment 25A, 715–723.

Haywood, J.M., Shine, K.P., 1997. Multi-spectral calculations of the direct radiative forcing of tropospheric sulphate and soot aerosols using a column model. Quarterly Journal of the Royal Meteorological Society 123, 1907–1930.

Hitzenberger, R., Petzold, A., Bauer, H., Ctyroky, P., Pouresmaeil, P., Laskus, L., Puxbaum, H., 2006. Intercomparison of thermal and optical measurement methods for elemental carbon and black carbon at an urban location. Environmental Science and Technology 40, 6377–6383.

Horvath, H., 1995. Estimation of the average visibility in central Europe. Atmospheric Environment 29, 241–246.

Jacobson, M.Z., 2001. Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. Nature 409, 695–697.

Jacobson, M.Z., 2002. Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming. Journal Geophysical Research 107 (D19), 4410. doi:10.1029/2001JD001376.

Järvi, L., Junninen, H., Karppinen, A., Hillamo, R., Virkkula, A., Mäkelä, T., Pakkanen, T., Kulmala, M., 2008. Temporal variations in black carbon densities with different time scales in Helsinki during 1996–2005. Atmospheric Chemistry and Physics 8, 1017–1027.

Kaufman, Y.J., Tanré, D., Boucher, O., 2002. A satellite view of aerosols in the climate system. Nature 419, 215–223.

Kim, H.J., Liu, X.D., Kobayashi, T., Kohyama, T., Wen, F.Q., Romberger, D.J., Conner, H., Gilmour, P.S., Donaldson, K., MacNee, W., Rennard, S.I., 2003. Ultrafine carbon black particles inhibit human lung fibroblast-mediated collagen gel contraction. American Journal of Respiratory Cell and Molecular Biology 28, 111–121.

Kirchstetter, T.W., Novakov, T., Hobbs, P., 2004. Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon. Journal of Geophysical Research 109, D21208. doi:10.1029/2004JD004999.

Kirchstetter, T.W., Aguiar, J., Tonse, S., Fairley, D., Novakov, T., 2008. Black carbon densities and diesel vehicle emission factors derived from coefficient of haze measurements in California: 1967–2003. Atmospheric Environment 42, 480–491.

Latha, K.M., Badarinath, K.V.S., 2005. Seasonal variations of black carbon aerosols and total aerosol mass densities over urban environment in India. Atmospheric Environment 39, 4129–4141.

Linke, C., Möhler, O., Veres, A., Mohácsi, Á, Bozóki, Z., Szabó, G., Schnaiter, M., 2006. Optical properties and mineralogical composition of different Saharan mineral dust samples: a laboratory study. Atmospheric Chemistry and Physics 6, 3315–3323.

Liousse, C., Cachier, H., Jennings, S.G., 1993. Optical and thermal measurements of black carbon aerosol content in different environments – variation of the specific attenuation cross-section, sigma (sigma). Atmospheric Environment 27 (8), 1203–1211.

Löndahl, J., Swietlicki, E., Lindgren, E., Loft, S., 2010. Aerosol exposure versus aerosol cooling of climate: what is the optimal emission reduction strategy for human health? Atmospheric Chemistry and Physics 10, 9441–9449.

Lyamani, H., Olmo, F.J., Alados-Arboledas, L., 2008. Light scattering and absorption properties of aerosol particles in the urban environment of Granada, Spain. Atmospheric Environment 42, 2630–2642.

Lyamani, H., Olmo, F.J., Alados-Arboledas, L., 2010. Physical and optical properties of aerosols over an urban location in Spain: seasonal and diurnal variability. Atmospheric Chemistry and Physics 10, 239–254.

MARM, Ministerio de Medio Ambiente, y Medio, Rural y Marino, 2010. Inventario de Gases de Efecto Invernadero de España. Edición 2010 (Serie 1990–2008). Sumario de Resultados. Dirección General de Calidad y Evaluación Ambiental.

Martínez-Lozano, J.A., Alados-Arboledas, L., De la Morena, B.A., Estellés, V., Exposito, F.J., Pey, J., Sicard, M., Sorribas, M., 2007. Intercomparison of Instrumentation for Atmospheric Aerosol Measurements. European Aerosol Conference. DAMOCLES campaign, Salzburg, Austria. Abstract T13A230.

Menon, S., Hansen, J., Nazarenko, L., Luo, Y., 2002. Climate effects of black carbon aerosols in China and India. Science 297, 2250–2253.

Moosmüller, H., Chakrabarty, R.K., Arnott, W.P., 2009. Aerosol light absorption and its measurement: a review. Journal of Quantitative Spectroscopy and Radiative Transfer 110, 844–878.

Müller, T., Henzing, J.S., de Leeuw, G., Wiedensohler, A., Alastuey, A., Angelov, H., Bizjak, M., Collaud Coen, M., Engström, J.E., Gruening, C., Hillamo, R., Hoffer, A., Imre, K., Ivanow, P., Jennings, G., Sun, J.Y., Kalivitis, N., Karlsson, H., Komppula, M., Laj, P., Li, S.-M., Lunder, C., Marinoni, A., Martins dos Santos, S., Moerman, M., Nowak, A., Ogren, J.A., Petzold, A., Pichon, J.M., Rodriquez, S., Sharma, S., Sheridan, P.J., Teinilä, K., Tuch, T., Viana, M., Virkkula, A., Weingartner, E., Wilhelm, R., Wang, Y.Q., 2011. Characterization and intercomparison of aerosol absorption photometers: result of two intercomparison workshops. Atmospheric Measurement Techniques 4, 245–268.

Pakkanen, T.A., Kerminen, V.M., Ojanen, C.H., Hillamo, R.E., Aarnio, P., Koskentalo, T., 2000. Atmospheric black carbon in Helsinki. Atmospheric Environment 34, 1497–1506.

Peréz, N., Pey, J., Cusack, M., Cristina, R., Querol, X., Alastuey, A., Viana, M., 2010. Variability of particle number, black carbon and PM_{10}, $PM_{2.5}$ and PM_1 levels and speciation: influence of road traffic emissions on urban air quality. Aerosol Science and Technology 44, 487–499.

Petzold, A., Schönlinner, M., 2004. Multi-angle Absorption photometry – a new method for the measurement of aerosol light absorption and atmospheric black carbon. Journal of Aerosol Science 35, 421–441.

Petzold, A., Schloesser, H., Sheridan, P.J., Arnott, W.P., 2005. Evaluation of multi-angle absorption photometry for measuring aerosol light absorption. Aerosol Science and Technology 39 (1), 40–51.

Ramanathan, V., Crutzen, P.J., Kiehl, J.T., Rosenfield, D., 2001. Aerosols, climate, and the hydrological cycle. Science 294, 2119–2124.

Ramachandran, S., Rajesh, T.A., 2007. Black carbon aerosol mass densities over Ahmedabad, an urban location in western India: comparison with urban sites in Asia, Europe, Canada, and the United States. Journal of Geophysical Research 112, D06211. doi:10.1029/2006JD007488.

Ramanathan, V., Carmichael, G., 2008. Global and regional climate changes due to black carbon. Nature Geoscience 1, 221–227.

Rodríguez, S., Van Dingenen, R., Putaud, J.P., Dell'Acqua, A., Pey, J., Querol, X., Alastuey, A., Chenery, S., Ho, K.F., Harrison, R., Tardivo, R., Scarnato, B., Gemelli, V., 2007. A study on the relationship between mass densities, chemistry and number size distribution of urban fine aerosols in Milan, Barcelona and London. Atmospheric Chemistry and Physics 7, 2217–2232.

Rodríguez, S., Cuevas, E., 2007. The contributions of "minimum primary emissions" and "new particle formation enhancements" to the particle number densities in urban air. Aerosol Science 38, 1207–1219.

Rodríguez, S., Cuevas, E., González, Y., Ramos, R., Romero, P.M., Pérez, N., Querol, X., Alastuey, A., 2008. Influence of sea breeze circulation and road traffic emissions on the relationship between particle number, black carbon, PM_1, $PM_{2.5}$ and $PM_{2.5-10}$ densities in a coastal city. Atmospheric Environment 42, 6523–6534.

Ruellan, S., Cachier, H., 2001. Characterisation of fresh particulate vehicular exhausts near a Paris high flow road. Atmospheric Environment 35, 453–468.

Safai, P.D., Kewat, S., Praveen, P.S., Rao, P.S.P., Momin, G.A., Ali, K., Devara, P.C.S., 2007. Seasonal variation of black carbon aerosols over a tropical urban city of Pune, India. Atmospheric Environment 41, 2699–2709.

Saha, A., Despiau, S., 2009. Seasonal and diurnal variations of black carbon aerosols over a Mediterranean coastal zone. Atmospheric Research 92, 27–41.

Schaap, M., Van Der Gon, HA.C.D., Dentener, F.J., Visschedijk, A.J.H., Van Loon, M., ten Brink, H.M., Putaud, J.P., Guillaume, B., Liousse, C., Builtjes, P.J.H., 2004. Anthropogenic black carbon and fine aerosol distribution over Europe. Journal of Geophysical Research 109, D18207. doi:10.1029/2003JD004330.

Schwartz, S.E., 1996. The white house effect-short wave radiative forcing of climate by anthropogenic aerosols – an overview. Journal of Aerosol Science 27, 359–382.

Sheridan, P.J., Arnott, W.P., Ogren, J.A., Anderson, B.E., Atkinson, D.B., Covert, D.S., Moosmüller, H., Petzold, A., Schmid, B., Strawa, A.W., Varma, R., Virkkula, A., 2005. The reno aerosol optics study: an evaluation of aerosol absorption measurement methods. Aerosol Science and Technology 39, 1–16.

Silva, A.M., Costa, M.J., Wagner, F., Bortoli, D., Pereira, S., Belo, N., Bugalho, L., Alados-Arboledas, L., Lyamani, H., Guerrero-Rascado, J.L., Kindred, D., Smith, M., 2007. CAPEX–AEROPOR overview: objectives and first results. European Aerosol Conference 2007, Salzburg, Abstract T13A185.

Sokolik, I.N., Toon, O.B., 1999. Incorporation of mineralogical composition of aerosols into models of radiative properties of mineral aerosol from the UV to IR wavelengths. Journal of Geophysical Research 104 (D8), 9423–9444.

Toledano, C., Cachorro, V.E., De Frutos, A.M., Torres, B., Berjón, A., Sorribas, M., Ston, R.S., 2009. Air mass classification and analysis of aerosol types at El Aeronosillo (Spain). Journal of Applied Meteorology and Climatology 48, 962–981.

Wang, C., 2004. A modeling study on the climate impacts of black carbon aerosols. Journal of Geophysical Research 109, D03106. doi:10.1029/2003JD004084.

Wehner, B., Bond, T.C., Birmili, W., Heintzenberg, J., Wiedensohler, A., Charlson, R.J., 1999. Climate-relevant particulate emission characteristics of a coal fired heating plant. Environmental Science and Technology 33, 3881–3886.

Yttri, K.E., Aas, W., Bjerke, A., Cape, J.N., Cavalli, F., Ceburnis, D., Dye, C., Emblico, L., Facchini, M.C., Forster, C., Hanssen, J.E., Hansson, H.C., Jennings, S.G., Maenhaut, W., Putaud, J.P., Tørseth, K., 2007. Elemental and organic carbon in PM10: a one year measurement campaign within the European Monitoring and Evaluation Programme EMEP. Atmospheric Chemistry and Physics 7, 5711–5725.