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RESEARCH ARTICLE

Black carbon trends in southwestern Iberia in the context of the financial and economic crisis. The role of bioenergy

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Abstract Since black carbon concentrations are useful to reveal changes in anthropogenic activities, measurements taken from 2007 to 2015 in a Portuguese city are used to assess to which extent the ambient air was impacted by the economic crisis. The average black carbon concentrations are representative of an urban area of small size ($1.3 \pm 1.3\ \mu g\ m^{-3}$). The highest concentrations are observed in the heating season, being biomass combustion one of the causes for the high values. The daily cycle of black carbon concentrations presents both morning and evening peaks, mainly due to road traffic and, in the heating season, to domestic heating as well. The yearly averaged black carbon mass concentrations decreased 33 % from 2007 to 2015, possibly due to a combination of the economic recession and environmental legislation. The reduction in road traffic led to a decrease in the daily morning peak from 2007 to 2015. This reduction was not followed by a decrease in the evening peak, explained by an increase in biomass burning. Biomass is the cheapest heating fuel in Portugal, and its consumption increased in the aftermath of the economic crisis. The use of bioenergy is an alternative to fossil fuels and presents many advantages. However, energy policies should discourage inefficient biomass burning and promote better ways of exploiting the available energy resources and emission air pollution mitigation strategies.

Keywords Air quality · Black carbon · Bioenergy · Biomass burning · Economic crisis · Urban environment

Introduction

Recent studies have reported several, both positive and negative, impacts of economic crises on air quality. For example, Castellanos and Boersma (2012) analyzed satellite observations and reported a reduction of at least 20 % in NO_2 throughout Europe for the period 2004–2010, attributed to both the global economic recession and environmental legislation. They showed that in many large European cities, the reduction in NO_x emissions during 2009 (recession year) outweighed approximately 4 years of policy improvements. Possibly due to the economic recovery in Europe, the NO_2 reductions slowed down in 2010. Vrekoussis et al. (2013) also used satellite observations of tropospheric NO_2 columns and reported significant reductions in large parts of Greece between 2008 and 2011 (the country is still facing the crisis that started in 2008). The most significant differences occurred in Thessaloniki and Athens, the two largest cities in Greece. In the capital, the overall NO_2 decrease was in the range of 30 to 40 %. The authors also analyzed the temporal variability of surface concentrations of CO, NO_x, SO_x, and O_3 from 2004 to 2011 and identified two

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distinct periods: From 2004 to 2007, small decreases were detected in the main gaseous pollutants, while from 2007 to 2011, a sharp decrease in all primary pollutant levels occurred (around 35 % reduction of surface NO_2 levels, 25 % in carbon dioxide, and 48 % in SO_2). On the contrary, the surface ozone levels increased 25 %, possibly due to a decreased titration by NO. The significant differences reported by Vrekoussis et al. (2013) since 2008 are well correlated to various economic indicators of the anthropogenic activity and are attributed to the economic crisis. Karagiannidis et al. (2015) analyzed air quality data in a Greek city, Patras, between 2008 and 2011. They also argued that the economic crisis contributed to a significant reduction in particulate matter and several trace gases, namely CO, NO, and NO_2, due to a decrease in the anthropogenic activities. Again, an increase in the ozone concentrations was reported. According to Russell et al. (2012), in the USA, large reductions in the observations of tropospheric NO_2 vertical column densities were detected, due to regulatory efforts and to the economic recession of 2008–2009. These authors showed that emission reductions from light-duty vehicles dominated the NO_2 decreases prior to the recession and a reduction in diesel truck activity has had a larger impact on emissions reduction since the start of the recession. The abovementioned articles mainly report a positive impact of economic recession on air quality. However, other studies have shown that the economic crises can also result in serious air pollution episodes. Saffari et al. (2013) conducted a wintertime sampling campaign for fine particles (PM2.5) in Thessaloniki during the winters of 2012 and 2013. They concluded that during 2013, the PM2.5 mass concentration increased 30 % when compared to 2012, while several wood smoke tracers (including potassium, levoglucosan, mannosan, and galactosan) had a twofold to fivefold increase. Additionally, the concentration of fuel oil tracers (e.g., nickel and vanadium) declined 20 to 30 % in the same period. These results indicate that the increase in airborne fine particles was mostly due to the replacement of fuel oil by the cheaper wood for domestic heating, as the price of fuel oil has nearly tripled.

Portugal is also among the European countries facing a serious financial/economic crisis in the aftermath of the 2008–2009 global recession. According to the Eurostat, the country's real gross domestic product (GDP) growth rate was negative from 2011 to 2013 and no real growth was verified in the last 12 years (Eurostat 2016). The country seems to be recovering, with growth rates of 0.9 in 2014 and 1.5 in 2015, as estimated by the Eurostat. The prices of conventional fuels have increased between 2010 and 2015 (DGEG 2016), and families have lost income (Eurostat 2016). This might have led either to a reduction of anthropogenic activities and therefore of emissions or, on the contrary, to an increase in uncontrolled biomass burning and consequent air quality deterioration.

Combustion processes, as is the case of biomass burning, emit particles (as well as trace gases) into the atmosphere, and part of these suspended particles (atmospheric aerosols) is characterized by strong light absorption. According to Bond and Bergstrom (2006), "The strongly light absorbing component also known as black carbon has had a variety of different names, of which soot is probably the most common. The remaining less-absorbing carbonaceous aerosol is loosely called organic carbon (...)." Black carbon (BC) aerosols emerge in the atmosphere as primary particles produced in combustion processes, and more than 90 % of the BC mass suspended in the atmosphere is in the submicrometer diameter range (Seinfeld and Pandis 2012). In the case of urban sites, BC is typically the principal particulate species that absorbs radiation in the visible spectrum (Bond and Bergstrom 2006; Moosmüller et al. 2009). Thus, its measurements (or alternatively the aerosol absorption coefficient) in urban sites provide useful information on human behavior and should be able to reveal changes in anthropogenic activities.

This work presents and analyzes BC data derived from aerosol absorption coefficient measurements performed within a small-sized Portuguese city in southwestern Europe. The city, Évora, has no relevant BC sources other than vehicles and biomass burning (the latter during the coldest period of the year); its vast rural surroundings are well ventilated, and no polluting industries exist in the area. Therefore, BC levels measured at Évora tend to be moderate to low, while being modulated at both daily and seasonal timescales (Pereira et al. 2012).

The available dataset on BC, comprising 8 years of continuous data (April 2007 to February 2015), is the basis of this work, whose main objectives are to present a comprehensive analysis of this long-term dataset and to assess to which extent the ambient air was impacted by the economic crisis and increased biomass smoke emissions during the cold days. In this work, biomass mainly refers to firewood. The paper also analyzes the consequences of the economic crisis in Portugal on air quality and discusses the related social and political implications.

The Portuguese financial and economic crisis

In the aftermath of the global financial crisis of 2007–2008, Portugal entered a financial and economic crisis. Because the country was unable to repay or refinance its government debt, Portugal applied for a bailout program from the International Monetary Fund (IMF), the European Financial Stabilization Mechanism (EFSM), and the European Financial Stability Facility (EFSF). Figure 1a shows the GDP growth rate for Portugal between 2003 and 2015. In 2009, the country experienced a negative growth of the GDP, with the situation improving during 2010. From 2010 to 2013, the GDP showed again negative growths, but a return to modest growth was estimated for 2014 and 2015 (Eurostat 2016).

Figure 1b shows that the mean equalized net income by household has decreased 6.5 % between 2010 and 2014. Although the country officially exited the bailout program in

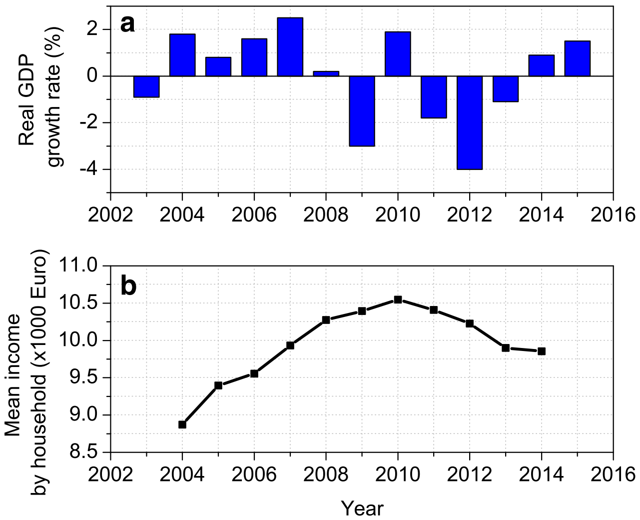

Fig. 1 **a** Gross domestic product growth rate for Portugal between 2003 and 2015 (Eurostat 2016) and **b** mean income by household for Portugal between 2004 and 2014 (Eurostat 2016)

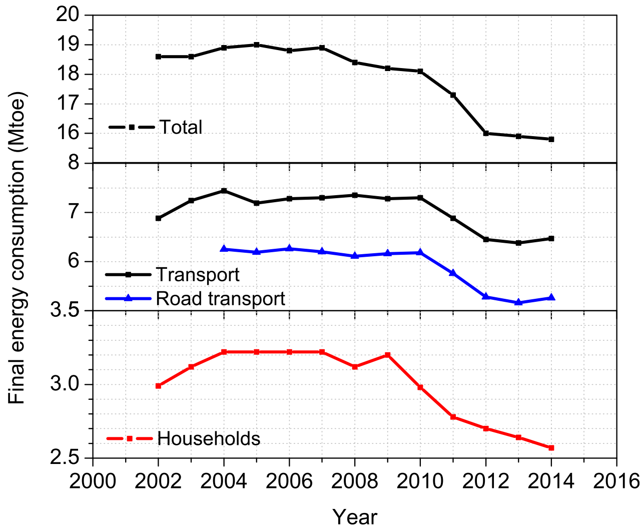

Fig. 2 Energy consumption in Portugal between 2002 and 2014 (DGEG 2016; Eurostat 2016)

2014, the families are still facing the consequences of the economic crisis.

From Fig. 2, we can see that since 2007, the total final energy consumption in the country decreased 16 % (Eurostat 2016), confirming the known positive correlation between economic activity and GDP (Siddiqi 2000). The reduction on the energy consumption has a direct impact on the environment. For example, it is probably accompanied by the reduction in the emissions of air pollutants that result from the combustion of coal, oil, or natural gas (still the most important energy sources in the country (DGEG 2016)). This reduction is mostly in areas where large amounts of fossil fuels are consumed (Castellanos and Boersma 2012; Russell et al. 2012).

Regarding the average prices of diesel, gasoline, and heating oil in Portugal (Fig. 3a), an overall increase from 2009 to 2012 can be seen. Between 2009 and 2012, the heating oil price, diesel, and gasoline increased, respectively, 90, 45, and 33 %. After that year, although the prices have been gradually decreasing, they are still higher than in 2009. Also, the electricity prices for a medium-sized household have increased in the country (Fig. 3b). From 2007 to 2015, an increase of almost 50 % was observed (DGEG 2016).

The generalized increase in fuel prices and decrease in income may have led to some change in the behavior of the Portuguese; more specifically, to a reduction of the road traffic and to a substitution of electricity and heating oil by cheaper fuels for household heating. Figure 2 shows that the final energy consumption in all types of transportation (transport sector) decreased 13 % from 2010 to 2013, having a 1 % increase in 2014 (Eurostat 2016), while in road traffic it reduced 15 % (DGEG 2016). Also, the residential energy consumption decreased 20 % from 2009 until 2014 (Fig. 2). The decreases of 18 and 21 %, respectively, in the electricity and LPG consumption of households from 2010 to 2014, along with an increase of 10 % in the solid biomass consumption, indicate a shift to solid fuels for heating (DGEG 2016). These changes can be observed in Fig. 4, where the consumption of the most important fuels used in the Portuguese households is presented. Note that the decrease in the solid biomass consumption between 2009 and 2010 is due to methodological reasons. Until 2009, the biomass energy consumption was estimated from surveys of 1988 and 1995, while from 2010 on, this parameter was estimated from a survey of 2010, updated with information from the sales of pellets and briquettes. The Portuguese have been consuming less solid biomass, when compared to 1995, but this decrease was not concentrated in 2009, as presented in Fig. 4. Solid biomass consumed by the Portuguese households is difficult to estimate. According to a survey, only 40 % of it is bought, 37 % is collected in the vicinities of the households, and 23 %

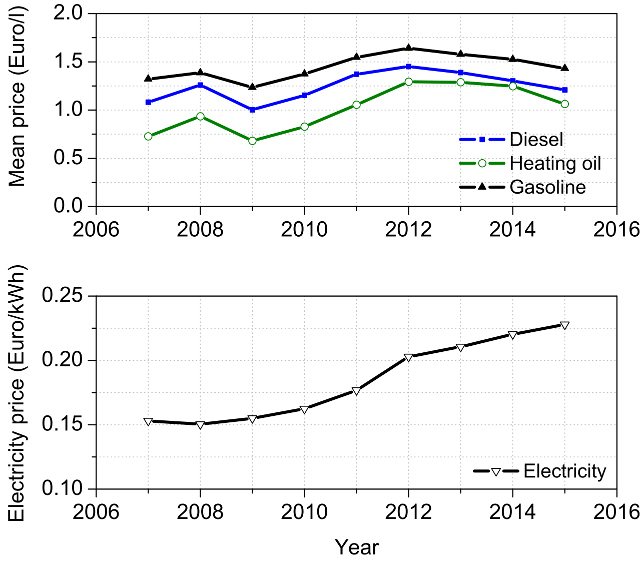

Fig. 3 **a** Liquid fuel and **b** electricity prices in Portugal between 2007 and 2015 (DGEG 2016)

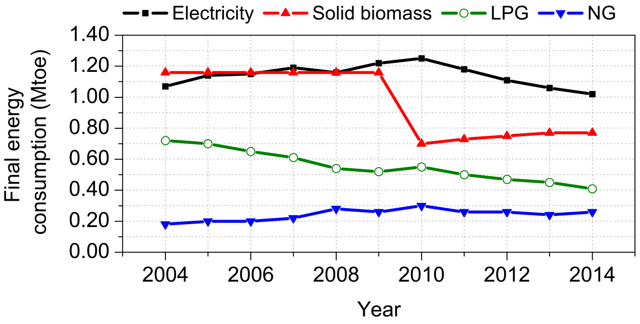

Fig. 4 Residential energy consumption between 2004 and 2014 by fuel type (DGEG 2016)

has other origins (INE and DGEG 2011). With the economic crisis, the informal ways of getting the biomass could have increased and the consumption of biomass may have been larger than that reported by DGEG.

According to the 2010 survey on energy consumption in households, the most common energy source for space heating in Portugal is firewood, with a share of 68 %, followed by heating oil and electricity, both with a 14 % share (INE and DGEG 2011). Solid biomass is used for household heating in 40 % of the Portuguese households, but not commonly used for domestic hot water. The most common equipment used for firewood burning is the open fireplace, followed by the fireplace with back boiler. The same survey refers that they are mainly operated at night (18–08 h). According to Canha et al. (2012), wood contributes to 18 % of the total Portuguese PM10 emissions.

Measurements and methods

Évora is a Portuguese city located in a rural region in southwestern Iberia. The municipality of Évora encompasses an area of 1307 km^2 and hosts circa 57,000 inhabitants.

The results of the 1981–2010 climatological normals, the latest available from the National Authorities (*Instituto Português do Mar e da Atmosfera* (IPMA)), indicate that climate in mainland Portugal is mostly temperate, type C, with the subtype Cs (temperate climate with dry summer). The region of Évora falls in the variety Csa, characterized by temperate climate with warm dry summer. Simultaneous measurements of several atmospheric quantities are carried out at the Atmospheric Physics Observatory of the University of Évora (Institute of Earth Sciences (ICT)) (38° 34′ N, 7° 54′ W, 293 m a.s.l.), within the urban center of the city. In particular, the aerosol absorption coefficient at the wavelength of 670 nm is measured at this site with a Multiangle Absorption Photometer (MAAP 5012, Thermo ESM Andersen Instruments, Erlangen, Germany). Its measurements are based on the combination of radiation transmitted and reflected by a particle loaded quartz fiber filter at different detection angles (Petzold and Schönlinner 2004). The aerosol is sampled at approximately 10 m above the ground through a PM10 inlet; the filter collects the particles within the ambient air which is sampled at a flow rate of 1000 l h^{-1}, and the instrument is set to operate at a sampling time of 1 min. Data was further averaged up to monthly data. The aerosol absorption coefficient effectively measured is converted to black carbon mass concentration by using the mass absorption coefficient of 6.6 m^2 g^{-1} recommended by the manufacturer. Standard meteorological variables such as temperature, humidity, and wind speed and direction are also continuously measured at the same site.

Total black carbon mass concentrations measured from April 2007 to February 2015 were aggregated, for example, into seasons, weekdays and Sundays, as well as colder and milder days in order to understand the influence of biomass heating on BC concentrations. Three different criteria were independently used for considering a particular day as part of a colder day dataset. This way, when analyzing the effect of biomass heating, the year was broken down in three distinct ways. One of the criteria used is based on the definition of a cold wave according to the World Meteorological Organization (Klein Tank et al. 2009), while the other two are based on the definition of the heating season of the Portuguese System for Energy Certification of Buildings (Decree-Law no. 118/2013) (Aguiar 2013).

A cold wave is defined as a period of at least six consecutive days when the maximum daily temperature is at least 5 °C below the average daily minimum temperatures for the reference period (minimum 30 years). In Évora, the number of individual days meeting the abovementioned temperature criterion between April 2007 and February 2015 is small (in total 28), causing a lack of statistical significance if the pure definition of cold wave is used; therefore, all the days that individually met the criterion were considered in the analysis despite being consecutive or not. Doing that, the “cold wave” series presents 159 days.

The definition of heating season used in this work is the one used by the Portuguese System for Energy Certification of Buildings. Conventional heating season is defined as the period of the year initiating in the first 10 days after October 1 when at each location, the average daily temperature is below 15 °C and ending on the last 10 days of May when the referred temperature is still below 15 °C. From 2006 to 2015, and for each year, the heating seasons at Évora were calculated (see Fig. 5), containing the total “heating season” series 1253 days (157 days year^{-1} on average).

A third, distinct, and simpler criterion for considering that a day was part of a colder day dataset was used in this work. The day was considered as being cold, when its average daily temperature was below 15 °C. A total of 1152 days fall into this criterion, and this is the number of days that the third series of colder days presents.

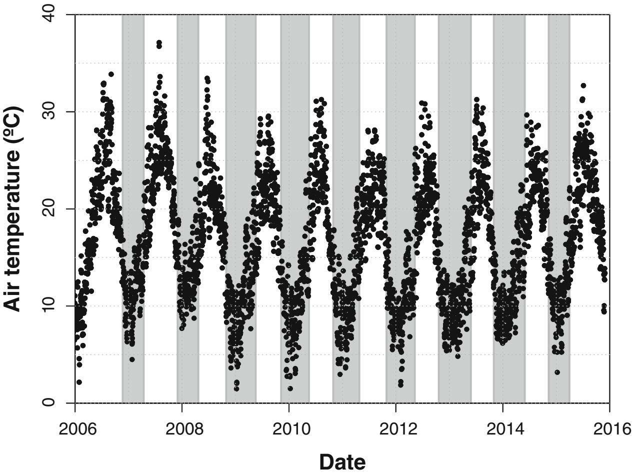

Fig. 5 Daily mean air temperature from 2006 to 2015 in Évora. The *gray bars* represent the conventional heating season

Throughout most of the paper, only the second criterion (heating season series—1253 days) is considered to differentiate colder and milder days. The days that do not fall inside this season are named "outside heating season" days. It is just at the end of BC daily cycle and the link to bioenergy section that the three different cold day criteria datasets are compared. This comparison is a test to whether the way the data was grouped affects the conclusions.

Statistical data analysis of the measured BC values was performed (Gilbert 1987). For detecting and estimating the trends of BC concentrations, the nonparametric Mann-Kendall test (Gilbert 1987) for testing the presence of the monotonic increasing or decreasing trend was used. Additionally, whenever the daily average was determined through numerical integration, the trapezoidal rule was used (Gautschi 2011).

Results

Statistics and long-term trends in black carbon concentrations

The black carbon mass concentrations measured at Évora between April 2007 and February 2015 were analyzed and grouped in several categories. For example, the data was aggregated by heating season and outside heating season, by month or by the four seasons. The basic statistical results are presented in Table 1. The average value (±standard deviation) for the BC mass concentration in the whole period is 1.3 ± 1.3 μg m^{-3}; the median value is lower, 0.9 μg m^{-3}, showing that this quantity is positively skewed. These values are in accordance with the ones obtained by Pereira et al. (2012) for the same location, but for a shorter period (2007–2009), and can be considered representative of an urban area of small size.

Table 1 Basic statistical parameters of BC_{PM10} in Évora from 2007 to 2015 based on hourly values

	Mean ± SD	Median	P5	P25	P75	P95
	μg m^{-3}					
Whole year	1.3 ± 1.3	0.9	0.3	0.6	1.4	3.3
Heating season	1.4 ± 1.6	1.0	0.2	0.6	1.7	4.1
Outside heating season	1.1 ± 0.9	0.9	0.3	0.6	1.3	2.5
Winter	1.7 ± 1.8	1.1	0.2	0.6	2.0	4.9
December	1.7 ± 1.8	1.2	0.3	0.7	2.1	4.9
January	1.8 ± 1.9	1.2	0.2	0.6	2.1	5.4
February	1.5 ± 1.6	1.0	0.2	0.6	1.8	4.3
Spring	1.1 ± 0.9	0.8	0.2	0.5	1.3	2.5
March	1.3 ± 1.2	0.9	0.3	0.6	1.5	3.1
April	1.0 ± 0.7	0.8	0.2	0.5	1.2	2.3
May	0.8 ± 0.7	0.8	0.2	0.5	1.2	2.2
Summer	0.9 ± 0.7	0.8	0.2	0.5	1.1	2.1
June	1.0 ± 0.7	0.8	0.2	0.5	1.2	2.2
July	0.9 ± 0.7	0.7	0.2	0.5	1.0	2.0
August	0.9 ± 0.7	0.7	0.2	0.5	1.1	2.1
Fall	1.4 ± 1.4	1.0	0.3	0.6	1.7	3.9
September	1.2 ± 0.9	1.0	0.3	0.6	1.5	2.8
October	1.4 ± 1.3	1.0	0.3	0.6	1.7	3.7
November	1.6 ± 1.7	1.1	0.3	0.6	1.8	4.5

SD standard deviation, *Px x* percentiles

The highest BC mass concentrations are observed in fall/winter and the lowest in spring/summer, revealing a clear annual cycle. The monthly average BC mass concentrations are relatively constant between April and August (0.8–1.0 μg m^{-3}), starting to increase in September, reaching a maximum of 1.8 μg m^{-3} in January, and then decreasing. A seasonal variation in BC concentrations in Évora was observed by Pereira et al. (2012). The additional input of soot during the colder months, due to domestic heating, and a boundary layer with reduced dispersion capacity were claimed to be the reasons for that modulation. A significant contribution of combustion heating systems to ambient air pollution was reported in several other studies (e.g., Puxbaum et al. 2007; Vuković et al. 2015).

Table 1 also shows the statistics for the black carbon mass concentration inside and outside the heating period. The heating season presents higher values (1.4 ± 1.6 μg m^{-3}) than the rest of the year (1.1 ± 0.9 μg m^{-3}), corroborating the previous explanation related with the supplementary loads of absorbing aerosols in the atmosphere.

With the aim of detecting the trend of the measured BC mass concentrations during the 8-year period analyzed, the average BC mass concentrations for each year were calculated from the monthly average BC data. These results are shown in Fig. 6. It is considered that a year starts in the beginning of spring and finishes at the end of the winter of the following

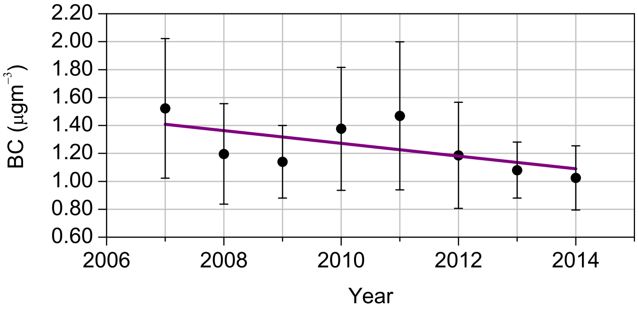

Fig. 6 Annual average of BC_{PM10} mass concentration in Évora from April 2007 to February 2015. The linear regression line applied to the data is also depicted

year (for example, when we speak of 2014, we mean the period from January to March 2014 to 28 February 2015). This way, we assure that a specific winter is treated as one season and not split.

A nonparametric Mann-Kendall test (Gilbert 1987) shows that there is a significant monotonic decreasing trend in the yearly averaged BC mass concentration in Évora (the test statistic $S = -16$). The level of significance is 0.1, i.e., there is statistical evidence that the black carbon mass concentrations decrease (corresponding to 10 % chance, there is no trend). Although the meteorological factors may have contributed to modulate the different mean annual BC values, the local anthropogenic emissions play the key role in the BC levels at the surface prevailing over meteorology when weekdays are considered as is the case here (Badarinath and Latha 2006; Järvi et al. 2008). Karagiannidis et al. (2015) showed that the most important meteorological factor that affects pollutant dispersion in winter is the surface wind. PM10 are reduced during the days of strong surface winds, which was also observed by Pereira et al. (2014) in the case of BC measurements, as a strong negative correlation with the wind speed was verified (but not with the wind direction).

The analysis of the data presented in Fig. 6 shows that the yearly averaged black carbon mass concentrations in Évora decreased 33 % from 2007 to 2014. The declining trend is in line with the results obtained in the European Union. For example, Guerreiro et al. (2014) report a reduction of 14 % in the emissions of primary PM10 from 2002 to 2011. This trend is associated with the efforts taken to reduce road transport emissions. However, the decrease in black carbon concentrations in Évora is considerably higher than in the European Union. We believe that the larger decrease is justified both by a combination of the legislation and of a reduction in the road traffic, due to the economic crisis (see Fig. 2). Also for Lyamani et al. (2011), the observed decrease (16–18 % in 2008 compared to 2006–2007) in the BC levels measured at Granada, Spain, seems to be associated to emission reductions due to economic crisis (in our case, a decrease of about 20 % is observed between 2007 and 2008).

BC daily cycle and the link to bioenergy

Figure 7 shows the 8-year average daily cycle of the BC mass concentration seasonal averages for winter, spring, summer, and fall calculated from BC concentration hourly values. It is clear that there are two daily maxima of BC mass concentration: one in the morning and another in the evening. The lower levels of BC mass concentrations are observed at dawn, mainly due to the practically absent particle emission and partly also due to some deposition of previously emitted particles. As the city awakes, the average BC mass concentrations increase due to an increase in human activities, reaching a maximum around 8–10 h UTC, which is intensified by the low thickness of the planetary boundary layer (PBL) due to the nocturnal surface cooling and insufficient surface solar heating in the early morning hours, presenting stable stratification in the near surface layer. After this maximum, the BC concentrations gradually decrease, due to a reduction in traffic and intensification of atmospheric instability. The latter is associated with an increase of the PBL height due to the development of the convective boundary layer, where turbulence is predominant. This phenomenon favors the transport of air pollutants from the surface aloft and thus causes the reduction of the BC loads near the surface (Lyamani et al. 2010). In the evening, traffic increases and a maximum in the average BC mass concentration is obtained again around 19–20 h UTC, decreasing thereafter with the reduction of traffic emissions.

All seasons are characterized by these daily cycles; however, as already mentioned above in the analysis of Table 1, the BC mass concentration values are lowest in spring and summer (Fig. 7b, c). The differences are particularly evident when we consider winter and summer (Fig. 7a, c), which present the highest and lowest values for the average BC mass concentration, respectively. The winter/summer differences are undoubtedly partly connected with the increase in the PBL height in warmer seasons, driven by the enhanced convective activity due to surface heating, consequently supporting vertical movement of particles toward higher levels, reducing the BC loads near the surface layer (Lyamani et al. 2010). Wind is another factor, among the meteorological factors, that may have a role in atmospheric aerosol dispersion and deposition; nevertheless, the mean wind speed at Évora site during the period considered is very similar for all seasons (winter 2.0 ± 1.1 ms^{-1}, spring 2.2 ± 1.5 ms^{-1}, summer 2.3 ± 1.0 ms^{-1}, fall 1.8 ± 1.0 ms^{-1}); therefore, it is reasonable to conclude that its role was minor in the seasonal differences found. This may be connected with the fact that the site is located in the center of the city, surrounded by buildings and trees on the horizon. Also, the wind direction does not seem to play an important role at this site since the emission sources are located all around the measurement site (Pereira et al. 2014). Although local meteorological conditions may influence the daily cycle, the local anthropogenic activities play the key role in the particle

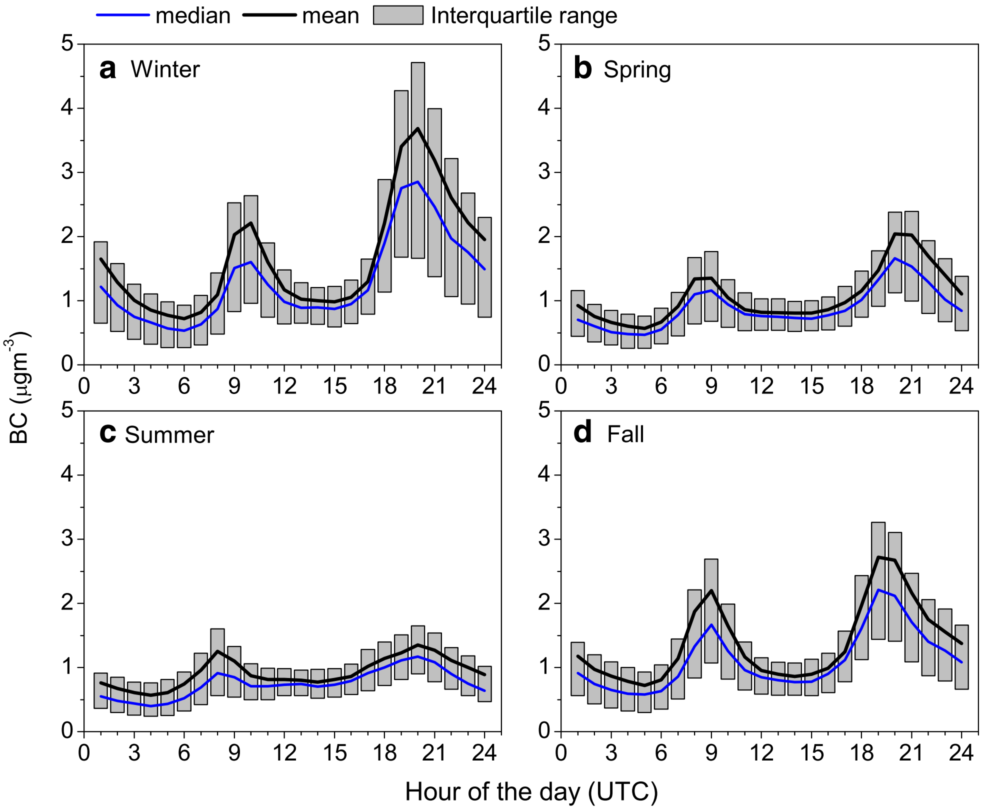

Fig. 7 Eight-year average daily cycle of the BC_{PM10} mass concentration seasonal average at Évora from April 2007 to February 2015 for **a** winter, **b** spring, **c** summer, and **d** fall

levels at the surface, which in Évora are mainly due to road traffic and seasonal activities connected with domestic heating in the heating season.

Figure 7 reveals another seasonal difference in the daily BC cycle. The morning and evening peaks in summer are quite similar, while in winter, this symmetry vanishes since the evening peak is markedly stronger than the morning one. Both daily maxima are related to the local traffic rush, and it is reasonable to consider that the quantity of vehicles in circulation is fairly similar in the morning and in the evening rush hours (Município de Évora 2007). In the heating season, biomass combustion, traditionally used for household heating, is the additional BC source. The PBL height is typically lower in the morning than in the evening (Lyamani et al. 2010). This factor alone would cause a decrease of the evening peak with respect to the morning peak, if the morning and evening emissions were roughly the same, which is the opposite of what was found here. This suggests that, in winter, biomass burning can be related to the significant increase of the evening maximum relatively to the morning peak, as well as to the maintenance of larger BC mass concentrations during nighttime until dawn. Corroborating this, INE and DGEG (2011) report that biomass household heating occurs mainly at night (18–08 h).

In order to explore this seasonal diurnal difference, a relation between the evening and morning black carbon peaks and the respective black carbon baselines is proposed here. This relation, hereinafter referred to as evening morning ratio (EMR), is defined as follows:

$$\mathrm{EMR} = \frac{\max\left(\mathrm{BC}_{\mathrm{evening}}\right)\text{-}\min\left(\mathrm{BC}_{\mathrm{afternoon}}\right)}{\max\left(\mathrm{BC}_{\mathrm{morning}}\right)\text{-}\min\left(\mathrm{BC}_{\mathrm{night}}\right)} \qquad (1)$$

In Eq. 1, max($BC_{evening}$) and max($BC_{morning}$) are computed as the maximum black carbon concentrations observed in the evening and morning, at each day and at a certain hour, respectively, while min($BC_{afternoon}$) and min(BC_{night}) are the minimum black carbon concentrations observed in the afternoon and night, respectively. The numerator accounts for the BC mass concentration due to the afternoon traffic and additional emissions of biomass burning caused by heating (Município de Évora 2007; INE and DGEG 2011), whereas the denominator results from the BC mass concentration due to the morning traffic (Município de Évora 2007). These amounts are estimated considering that the mid-afternoon and dawn baselines (typically 13:00–15:00 UTC and 04:00–06:00 UTC) represent the background BC levels prior to each of the peaks, allowing for estimating the BC amounts corresponding to each peak and subsequently the ratio between afternoon and morning peak mass concentrations. These peaks tend to happen in the same periods of the day: 08:00–10:00 UTC for the morning peak and 19:00–21:00 UTC for the afternoon peak.

EMR was only calculated for the weekdays (Monday to Friday), since the anthropogenic activity in those days is very different from that on the weekends. On weekends, particularly Sundays, traffic is quite reduced in the morning period, but not in the afternoon, which reflects on the absence of a morning peak. As a consequence, using the equation for these days should bring no information regarding the balance between biomass burning BC and traffic BC. This can be confirmed from the comparison presented in Fig. 8 that presents the 8-year average daily cycle of the BC mass concentration for weekdays and Sundays calculated from the BC concentration hourly values. Note that for these calculations, public holidays were also considered and treated as Sundays.

Table 2 shows the EMR values calculated from the 8-year averaged hourly BC mass concentrations for each season, as well as for the heating season and outside the heating seasons. The EMR values are significantly higher in the heating season than in the rest of the year (also in winter than in summer).

The results presented in Table 2 seem to be in agreement with the argumentation that the significant increase of the evening maximum relatively to the morning peak in the heating season is linked to biomass burning. In the colder months, people heat their homes mainly in the evening resulting in a higher evening peak of BC mass concentration. The diurnal variation reported here was already observed, for example, in Thessaloniki (Saffari et al. 2013) and attributed to increased emissions from residential heating sources in the evening.

The global decrease in the BC concentrations from 2007 to 2014 reported in Fig. 6 does not mean that all economic sectors have reduced BC emissions during this period. To try to understand if the household fuel combustion sector has decreased, maintained, or increased its BC emissions, the yearly trend of EMR, presented in Fig. 9, was analyzed (remember that a higher value of EMR was associated to biomass combustion).

Figure 9 clearly shows that EMR increased between 2007 and 2014 in Évora. A nonparametric Mann-Kendall test shows that there is a significant monotonic increasing trend in EMR ($S = 16$). The level of significance is 0.1, meaning that there is a 10 % chance that there is no trend. The morning BC peak presents a significant monotonic decreasing trend ($S = -20$, the level of significance is 0.05), while the evening peak does not show a significant trend. The morning peak decrease (51 % in 8 years) is explained with the efforts taken to reduce road transport emissions and with a decrease in traffic verified in Évora due to the economic crisis. This would also result in a decrease in the evening peak, which, however, could not be confirmed by the data. It may be concluded that another factor led to the increase in the evening peak and the most likely explanation is an increase in biomass combustion during the analyzed period. The indicator of biomass combustion, EMR, increased 48 %, supporting the idea that biomass combustion increased in this period. To further explore this idea, the data was separated into BC mass concentrations in the heating season and outside the heating season (defined in Measurements and methods section).

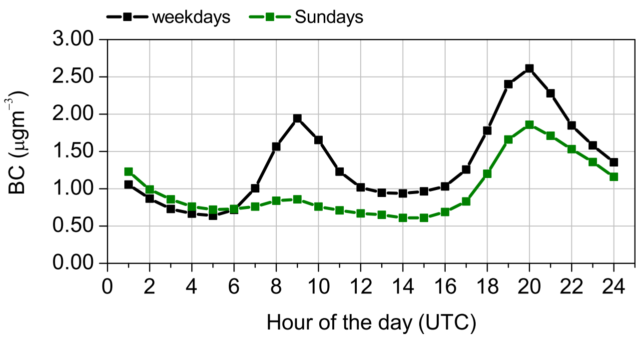

Fig. 8 Eight-year average daily cycle of the BC_{PM10} mass concentration for weekdays and Sundays at Évora from April 2007 to February 2015

Figure 10 shows the comparison between the 8-year average daily variations of BC mass concentration obtained from hourly values in Évora, from April 2007 to February 2015, for days in the heating season and outside the heating season.

Figure 10 shows that the morning peak does not differ significantly for the heating season and outside it. This was expected, since the morning peak is mainly affected by the emissions due to traffic, which is not correlated to temperature. However, it can be seen that the colder days present a considerably higher evening peak, confirming that biomass combustion has an important contribution. Table 2 shows that the ratio between the morning and evening peaks is significantly higher for the heating season than outside this period, confirming this hypothesis.

As an attempt to quantify the increase in BC emissions due to biomass combustion, the daily average BC mass concentration was calculated for each of the seasons by numerically integrating the average hourly BC mass concentration in

Table 2 Ratio between the evening and morning peaks of BC_{PM10} in Évora from 2007 to 2015

Winter		1.4	Dec.
	1.5	1.4	Jan.
		2.0	Feb.
Spring		1.9	Mar.
	1.3	0.7	Apr.
		0.7	May.
Summer		0.6	Jun.
	0.7	0.6	Jul.
		0.8	Aug.
Fall		0.9	Sep.
	1.1	0.9	Oct.
		1.4	Nov.
Heating season	1.6		
Outside heating season	0.8		

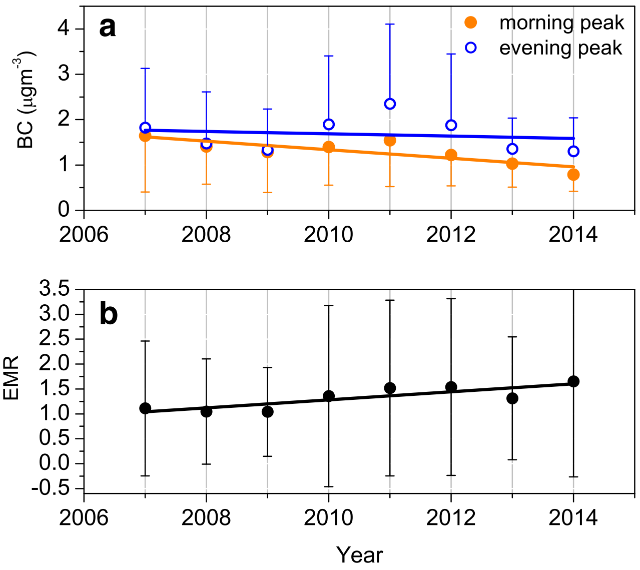

Fig. 9 **a** Morning and evening peaks and **b** EMR in Évora from April 2007 to February 2015 based on monthly values. The linear regression lines are also shown

1 day. The difference between both integrals corresponds to the BC emission due to biomass combustion, and it is estimated to be 8.2 μg m^{-3} day.

The two other criteria defined in Measurements and methods section to differentiate the colder and the warmer days were used in order to substantiate the conclusions obtained so far and to test the aggregation of days chosen to link BC concentrations and biomass combustion. The days were separated into two additional groups: (i) days with daily average temperatures below and above 15 °C and (ii) days falling in the category of cold wave and outside this class. Figure 11 shows the 8-year average daily cycle of the BC mass concentration obtained separately for the three categories considered. It is interesting to note that the 8-year average daily variations of BC mass concentration at Évora for the three different groups of colder days present identical behavior, although the number of days falling in each category can be considerably different (see end of Measurements and methods section), supporting the previously drawn conclusions. In the light of what has been discussed, this means that, for these three distinct groups of cold days, the daily BC emissions are identical.

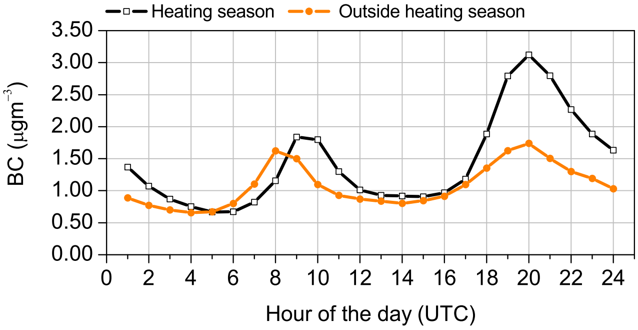

Fig. 10 Eight-year average daily cycle of the BC_{PM10} mass concentration at Évora from 2007 to 2015 for days in the heating season and outside the heating season

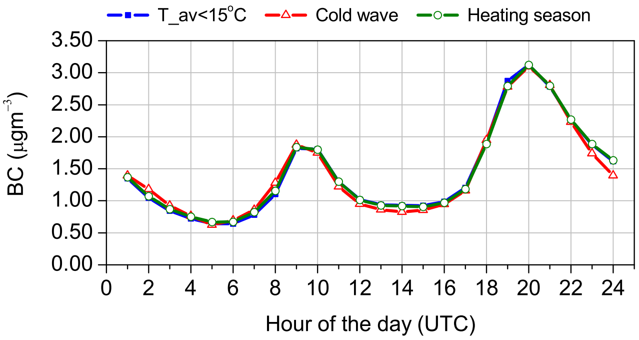

Fig. 11 Daily cycle of the BC_{PM10} mass concentration average at Évora based on hourly values from 2007 to 2015 for three different categories of cold days

Social and policy implications

The results presented in this paper point out to a consistent decrease in the particles emitted by road traffic in the last 8 years in Évora and to an increase of biomass burning particles in the same period. Both can be linked to the economic crisis that the country faces, although other reasons may contribute to these trends. It is a fact that, in Europe, pollutant emissions from road transport and industry have been reducing due to the existing legislative regulations and abatement strategies (Guerreiro et al. 2014). The rate at which they are decreasing in Évora is higher though, likely due to the traffic reduction in the aftermath of the economic crisis. The study of Lyamani et al. (2011) also reports a decrease in BC concentrations in Granada, Spain, due to a drop in the road traffic as a result of the economic crisis. Granada is also located in southwestern Iberia, and there was also an economic crisis in Spain. Our analysis also shows that the particles emitted from household biomass combustion have increased. This is consistent to what has been observed in Europe, where PM10 emissions from household combustion have slightly increased (Guerreiro et al. 2014).

The combustion of biomass leads to the emission of airborne pollutants, among others CO, NO_x, SO_2, hydrocarbons, and particulate matter. It is known that wood smoke particles contain several toxic organic compounds such as methoxyphenols (Kjällstrand and Petersson 2001) and PAHs (Bari et al. 2009). The exposure to wood smoke emissions has been linked to several adverse effects on human heath, such as pulmonary diseases (Orozco-Levi et al. 2006), effects on the cardiovascular system (McCracken et al. 2007), and increased markers of inflammation, coagulation, and lipid peroxidation (Barregard et al. 2006). It is worth mentioning that increased ambient particulate matter pollution was in 2010 the ninth most important risk factor for global disease burden, while household air pollution from solid fuels the fourth (Lim et al. 2012).

Another impact of particles is that they can influence climate through the increase of back-scattered solar radiation and absorption of solar and longwave radiation, thus directly affecting the Earth's radiation budget. On the other hand, aerosols may enter cloud microphysical processes as cloud condensation nuclei, changing the microphysical properties of clouds and their lifetime, thereby modifying the planetary albedo and precipitation efficiency, entering delicate mechanisms such as the water cycle (Cattani et al. 2006; Santos et al. 2008). The biomass burning aerosols are considered one of the main reasons for this kind of cloud property modification. In addition, this type of aerosols contains organic compounds and black carbon, the latter being a strong absorber of solar radiation that can greatly impact cloud formation and evaporation (Ackerman et al. 2000).

The conclusion of the above paragraphs should not be that biomass combustion should be stopped; only that care must be taken. The use of biomass, a renewable energy source (RES), in both industrial and nonindustrial sectors, is an alternative to the use of fossil fuels and contributes to the reduction of greenhouse gas emissions. It presents many advantages when compared to fossil fuels and also some advantages over some other RES, namely its low cost, provision of alternative sources of income for farmers, or less dependence on short-term weather changes (EC 2005). Other advantage of biomass combustion may be the harvesting of forest and agriculture residues, therefore contributing to the reduction of forest fires and the preservation of important traditional biodiversity-rich ecosystems, such as the *montado* (Malico et al. 2016).

However, care must be taken to assure that biomass is used sustainably and that the impacts of biomass combustion are studied. An example of such a study is the work of Rafael et al. (2015) where the impact of energy conversion of forest biomass residues in Portugal on air quality was evaluated. The authors concluded that the most important existing and planned biomass power plants contribute to an increase of the pollutant concentration in the atmosphere, however restricted to the surrounding areas of the thermal plants, and most significantly for NO_2 and O_3. Since this study concentrated in power plants, that have flue gas treatment with bag filters and mostly with electrostatic precipitators, the emission of particulate matter was not significant.

The emission factors of particles in domestic fireplaces, common in Évora, is, however, comparatively higher. Kocbach Bølling et al. (2009) analyzed the literature and reported that the PM emission factors from residential biomass combustion appliances are highly uncertain. They stated that pellet stoves and boilers emit in the range of 10 to 50 mg MJ^{-1}, modern wood stoves in the range of 34 to 330 mg MJ^{-1}, and open fireplaces in the range of 160–910 mg MJ^{-1}. The particle emission factors are highly dependent on the type of combustion facility, chimney characteristics, and operational parameters like fuel load and airflow. The burning conditions in small domestic stoves, in particular when manually fired, are highly variable, hampering a systematic study on the factors that influence the particle emissions (Schmidl et al. 2011). Independent of all these uncertainties is the fact that the use of the most efficient domestic wood burners should be promoted, in detriment of the manually fired systems.

In the study of Schmidl et al. (2011), a number of maloperation tests were performed to investigate a potential user-derived influence on emissions. Their results showed that user habits have a major influence on emissions from manually fired systems (for small-scale logwood stoves, the PM10 emissions can be four times higher than in standard operation). For the automatically fired systems, the user cannot have much influence on the operation. The study of Schmidl et al. (2011) highlighted the importance of detailed, high-quality user information and instructions about the optimum operation of the stoves.

Emissions from fireplaces are not only dependent on the characteristics of the combustion facility and operational parameters but also dependent on fuel type burned. Gonçalves et al. (2010) compared the emission factors of wood species growing in Portugal and commonly used in fireplaces (eucalyptus, *Eucalyptus globulus*, pine, *Pinus pinaster*, cork aok, *Quercus suber*, and golden wattle, *Acacia longifolia*). They concluded that the highest particulate matter emissions are produced by oak wood combustion (almost three times higher than pine emissions). Oak wood comes from a common species widely spread in the central southern regions of Portugal and used in Évora.

The BC measurements taken at Évora show that a more detailed analysis of urban biomass heating on the local air quality is recommended. If preventive and corrective actions to decrease the PM10 levels are needed, it is important to understand the sources of particles and on which of them it is possible to act (Canha et al. 2012). The results of this paper indicate that, in Évora, the contribution of residential biomass combustion to ambient air pollution in the heating season is important and that it has increased between 2007 and 2015. It is likely that this evolution is mainly driven by economic reasons and to a lesser extent by policies and the public awareness of biomass as a green fuel (residential biomass burning is not particularly promoted by the current public policies). National policies should be aware of this growth and be adjusted to mitigate the potential harmful effects of an increase of residential biomass combustion. For example, the use of inefficient, manually fired fireplaces should be discouraged. This may, for example, involve the certification of equipment with lower emissions or subsidies for the replacement of old equipment by more efficient equipment with lower particle emission. Also, adequate user habits and adequate maintenance and operation of combustion equipment should be promoted. Likewise, public policies can promote the use of standardized fuels that lead to lower emissions (for example, the VAT of pellets and briquettes could be reduced). This type of policies could also boost the Portuguese pellet market, currently facing some obstacles. In 2014, Portugal was the seventh largest pellet producer in the world, but the internal demand for pellets was small, and the country relied heavily on exports (Malico et al. 2017). Promoting the internal pellet market is positive for the sustainability of this industry.

As discussed above, understanding the levels of PM10 in general and black carbon in particular is crucial to support the national and regional policies and to know which challenges need to be tackled. The study presented in this paper intends also to be a contribution, supporting the notion that the heating season black carbon pollution in Évora can be controlled if the authorities focus on emission mitigation strategies at the local level. It also suggests that the need for effective particulate matter regulations in Évora (and Portugal) increased during the past years. Although no serious wintertime air pollution episodes occurred in the city, this study points out to raise of particles with harmful health effects. It is important that the energy policies reflect this threat.

Another important aspect to consider is that the apparently positive airborne particle reduction observed in Évora due to the economic crisis might only have a short-term effect and that in the long run, measures to improve the quality of the local and global environmental might have conducted to better results. This is discussed in the paper by Siddiqi (2000) in the context of the Asian financial crisis. Due to the economic crisis, the government and people tend to postpone the investments in environment-friendly measures, such as the replacement of less efficient equipment by more efficient ones, investments on renewable energies, or increasing the energy efficiency of existing buildings. The latter is another measure that, through the reduction of the heating loads, would contribute to better air quality in Évora and to a reduction in greenhouse gas emissions.

Conclusions

The economic crisis in Portugal (from 2011 onward) resulted in a reduction of road transport and, as a consequence, of emissions to the atmosphere. This is confirmed by measurements of black carbon mass concentrations in a small town in southwestern Iberia, Évora. The main sources of black carbon in Évora are traffic and biomass combustion. These two anthropogenic activities modulate the daily black carbon concentration cycle in the city. In the morning and evening rush hours, there are two peaks of soot concentrations. Outside the heating season, these two peaks are identical in magnitude, whereas in the heating season, the evening peak is markedly higher, due to the additional particulate matter emission from biomass combustion for heating. The measurements taken at Évora from 2007 to 2015 show that the reduction in road traffic lead to a 50 % decrease in the daily morning peak from 2007 to 2014 and that this reduction was not followed by a similar decrease in the evening black carbon peak concentration. From 2007 to 2014, the ratio between these two peaks corrected by the night and afternoon minimum concentrations increased almost 50 %. This fact can be explained by a growth in biomass burning for heating. In fact, wood biomass is the cheapest fuel for household heating in Portugal, and in the context of an economic crisis, our study points in the direction of people using more biomass, with local impacts on air quality.

Using biomass for energy production in and of itself is not seen as negative. The utilization of biomass, a renewable energy source, in both industrial and nonindustrial sectors, is an alternative to the use of fossil fuels and presents many advantages. Nevertheless, national and regional energy policies should promote the best ways of exploiting the available energy resources, and authorities should focus on emission mitigation strategies at the local level. The following recommendations are made:

- Promotion of the most efficient domestic biomass burners, in opposition to the manually fired systems, commonly used in Portugal.
- Promotion of detailed and high-quality user information and instructions about the optimum operation of the combustion equipment they use.
- Increasing and encouraging the energy efficiency of existing buildings.
- Supporting more detailed studies on urban biomass heating and its impacts on local air quality.
- Supporting measurements to quantify the atmospheric levels of PM10 in general and black carbon in particular.

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