



# Analysis of PM<sub>10</sub> and PM<sub>2.5</sub> Concentrations in an Urban Atmosphere in Northern Spain

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## Abstract

This work analyses levels of particles PM<sub>10</sub> and PM<sub>2.5</sub> recorded at four air-quality monitoring stations located in the urban area of Valladolid (Spain) during 2015–2016. To achieve this, the evolution of particle concentrations at different time scales was determined. Average concentrations ranged from 15.3 to 17.6  $\mu\text{g m}^{-3}$  for PM<sub>10</sub> and between 8.9 and 14.8  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub>. The highest monthly means were recorded in autumn and winter. The difference between mean concentrations at weekends and on weekdays for PM<sub>10</sub> was around 3  $\mu\text{g m}^{-3}$  at most of the measuring stations and was 1  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub>. Two concentration peaks were found during the day, one in the morning and the other in the evening, which evidenced the influence of traffic and other anthropogenic activities on PM concentrations. Their mean values were approximately 21 and 17–21  $\mu\text{g m}^{-3}$ , respectively, for PM<sub>10</sub>. Mean maximum values for PM<sub>2.5</sub> were 12  $\mu\text{g m}^{-3}$ , except at one of the measuring sites, with 17  $\mu\text{g m}^{-3}$  for the morning maximum and 1  $\mu\text{g m}^{-3}$  more for the nocturnal peak. In addition, the impact of long-distance transport of air masses in the study area was analysed by applying a HYSPLIT trajectory model, taking into account backward trajectories of European, African, and Atlantic origins as well as local conditions. In particular, high concentration events due to Saharan dust intrusions are presented. Finally, background levels of particle concentrations estimated at most sampling areas were around 15 and 7.7  $\mu\text{g m}^{-3}$  for the PM<sub>10</sub> and PM<sub>2.5</sub> particle fractions, respectively.

Air pollution by particulate matter is related to alterations in the atmosphere's natural composition due to incoming particles from natural or anthropogenic causes. Atmospheric particulate material consists of solid and liquid particles (except water) suspended in the atmosphere. It includes both particles in suspension and sediment particles (diameter > 20  $\mu\text{m}$ ) with low residence time in the atmosphere (IPCC 2001; Puigcerver and Carrascal 2008; Salvador and Artiñano 2000). Particulate matter refers to a mixture of organic and inorganic compounds of different size and chemical composition. The main sources of natural primary particles include soil emissions, ocean surfaces, and biogenic sources (Inza 2010; Negral et al. 2008; Sánchez et al. 2007), with the long-range transport of coarse dust particles from African areas being an important contributor (Naidja et al. 2018; Kassomenos et al. 2012; Querol et al. 2009; Sánchez et al. 2007). Moreover, there are natural contributors of secondary

particle precursor gases, such as volcanic emissions, soil, and plant transpiration, as well as lightning, and there are different anthropogenic sources of particulate matter. Road traffic is the most important source of primary particles (Charron and Harrison 2005; Amato et al. 2009; Querol et al. 2012). Combustion processes in thermal power stations and other industrial sectors, as well as transport of anthropogenic aerosols from central European and Mediterranean areas together with certain agricultural activities also must be taken into account as sources of particles.

Research on atmospheric particulate matter has increased over the past two decades because of its impact on human health (Brook et al. 2010; Fernández-Camacho et al. 2016; Harrison and Yin 2000; Karagulian et al. 2015; Pope and Dockery 2006), on different ecosystems and on climate change (IPCC 2001; Paraskevopoulou et al. 2015). To minimize these impacts, it is necessary to implement measures to concentrate this complex group of pollutants, evaluate their spatial and temporal behaviour, and relate them to meteorological parameters, chemical composition, and origin so that the authorities can monitor them, establish control strategies, and reduce emissions from

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particulate material and precursor gases. In this sense, the significant technological progress made in measuring particle levels has allowed for a more precise understanding of their chemical composition and physical properties (Fuzzi et al. 2015).

Some research has shown that anthropogenic aerosol levels have increased in Asia and reveal a substantial impact of Asian pollutant flows on general circulation and global climate (Wang et al. 2014). There also are high levels of particles in North Africa, higher than those in Europe, the eastern United States, and western South America (Naidja et al. 2017; WHO 2006). However, between 1990 and 2011, total emissions of particulate matter fell considerably in the European Union (EEA 2015), particularly in the energy production and distribution sector due to factors such as the shift from coal to natural gas as the fuel for electricity generation and because of improvements in the performance of pollution elimination equipment installed in industrial facilities, as well as the reduction in road transport.

To interpret the contributions made by pollutants, such as particles, and their influence on air quality levels, the paths taken by air masses before they affect a specific location, in other words their backward trajectories, are analysed. Other complementary tools for analysing air masses can be provided by satellite data and meteorological observations, among others. Several studies have analysed Saharan dust episodes at certain locations on the Iberian Peninsula and in the Canary Islands (Alonso 2007; Cachorro et al. 2016; Querol et al. 2009; Rodríguez et al. 2001; Sánchez et al. 2007; Toledano et al. 2009; Viana et al. 2002). However, this paper seeks to further current knowledge of the distribution patterns of PM<sub>10</sub> and PM<sub>2.5</sub> particle fractions and the relationship between them. The anthropogenic effect on particle levels, as well as their background levels in a medium-sized city in the upper Spanish plateau, were considered.

The main objective of this paper is to interpret the variability of PM<sub>10</sub> and PM<sub>2.5</sub> concentrations in an urban area, Valladolid, a city located in northern Spain, and the influence of air masses by performing the following tasks: (a) analysing temporal variations of PM<sub>10</sub> and PM<sub>2.5</sub> particle fractions using data recorded at four air quality monitoring stations: Arco de Ladrillo, La Rubia Vega Sicilia, and Puente del Poniente; (b) describing the air mass movements that reach the sampling sites and their contribution to the mean levels of those particle fractions; (c) evaluating the influence of Saharan dust outbreaks on atmospheric particle concentrations and showing an important episode that occurred during the study period; (d) studying the relationship between PM<sub>2.5</sub> and PM<sub>10</sub> fractions; (e) quantifying background PM<sub>10</sub> and PM<sub>2.5</sub> concentrations at the measuring sites using data set selection so as to reduce the effect of controlling factors.

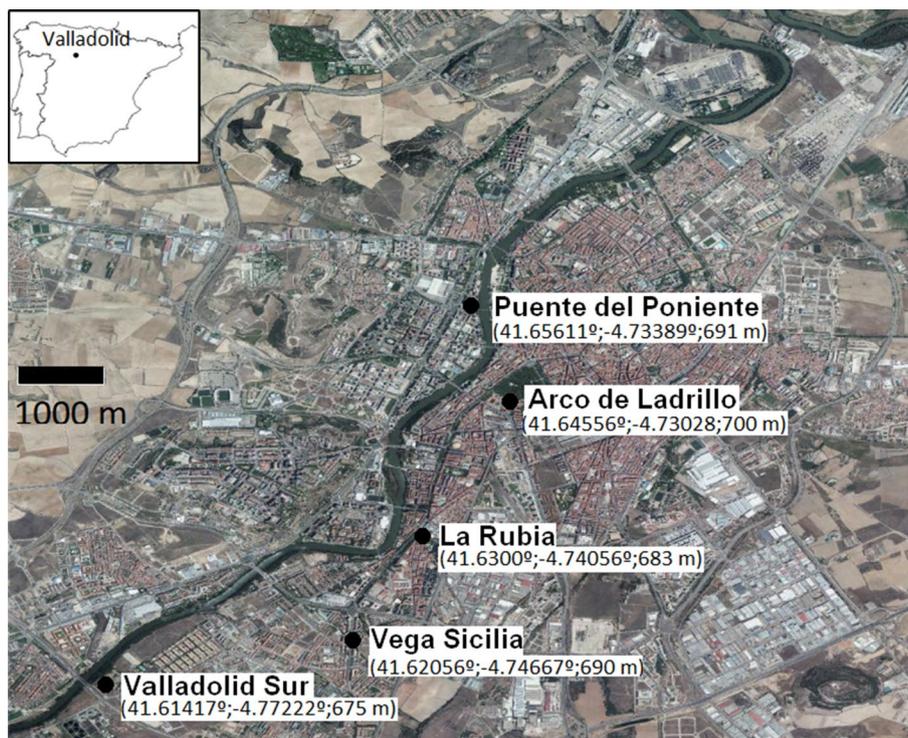
## Materials and Methods

The study is based on 2-year data, 2015–2016, provided by an automatic network of monitoring stations belonging to the Valladolid Air Quality Monitoring Network, supervised by the Valladolid City Council. The city is located in the centre of the northern upper plateau of the Iberian Peninsula at an altitude of approximately 690 m a.m.s.l. (Fig. 1). It is considered a medium-sized city and had a population of 303,905 inhabitants in 2015, which decreased to 301,876 in 2016. It also is considered the 13th Spanish city in terms of population size. The main economic activities are services (83.4%), the building sector (11.4%), industry (4.6%), and agriculture (0.5%). The number of vehicles was 386 per 1000 inhabitants; 78% of these were cars. The monitoring site consists of five stations classified as traffic type in an urban area, because they are under the influence of road traffic emissions and provide hourly data on the main atmospheric pollutants. Four of them, Arco de Ladrillo (AL), La Rubia (LR), Vega Sicilia (VS), and Puente del Poniente (PP), have PM<sub>10</sub> and PM<sub>2.5</sub> monitoring equipment. Particulate matter concentrations are measured using continuous beta attenuation monitors. The devices are connected to a data acquisition system, which allows measurements to be recorded on a database. Measurements are then stored and sent to the city council data processing centre for validation. Communication between the stations and the data processing centre is through a fibre optic network. Percentages of valid PM<sub>10</sub> and PM<sub>2.5</sub> data were > 95%, except for PM<sub>2.5</sub> data at LR and VS stations with 86% and 89%, respectively. The location and features of the monitoring stations are shown in Fig. 1.

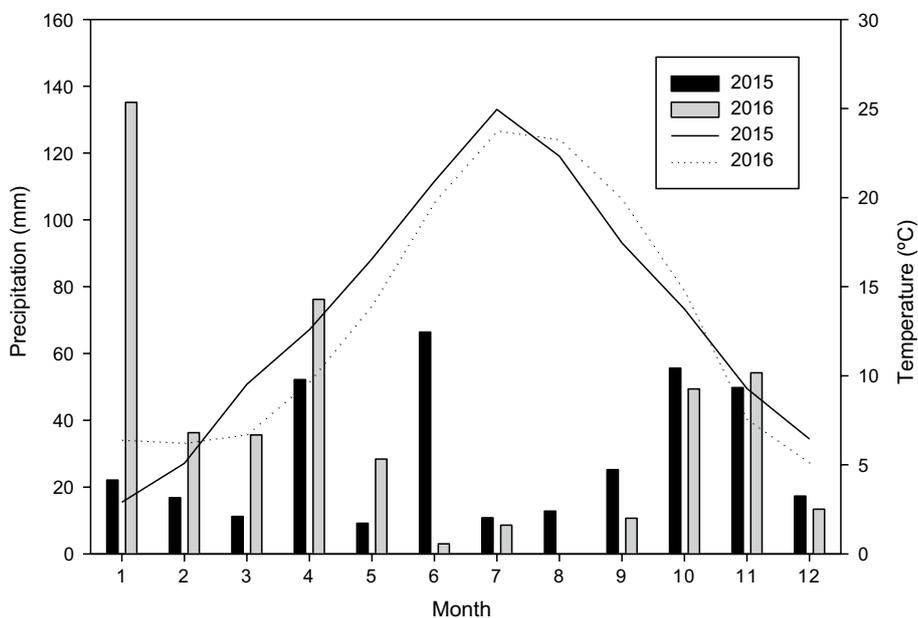
The typical meteorological features at the measuring site corresponded to a continental Mediterranean climate. Meteorological data were obtained from Meteomanz.com, the main source of data being the server of the National Oceanic and Atmospheric Administration (NOAA). Mean wind speed was 1.8 m s<sup>-1</sup>, and the prevailing wind directions based on an 8-sector wind rose were north and southwest. Figure 2 shows the temperature and precipitation of Valladolid for the sampling period. Precipitation was fairly evenly distributed throughout the year, except for a slightly drier season during the summer months of 2016 and much more abundant rainfall in January 2016, which exceeded 130 mm. The accumulated rainfall for each year of study was 349.2 and 451.0 mm for 2015 and 2016, respectively, with the latter being very close to the average of the city's 30-year historical series, which is around 430 mm. The year 2015 was less rainy than usual.

Different techniques were applied for data processing. Temporal variations of particle matter were examined

**Fig. 1** Location of the monitoring stations in Valladolid (Castilla y León, Spain). The geographic coordinates are also included. PNOA image courtesy of © ign.es



**Fig. 2** Monthly air temperature (line plots) and cumulative monthly precipitation (vertical bars) measured in Valladolid in the study period



considering variance analysis (ANOVA method). A multiple range test is used to examine the significant differences among the group of means in a sample using Fisher's least significant difference (LSD) procedure (Wilks 2011).

The relationship between the concentration of both kinds of particulate matter was analysed calculating the ratio PM<sub>2.5</sub>/PM<sub>10</sub> and establishing linear dependence with the Pearson correlation coefficient.

Atmospheric transport pathways were used to interpret the impact of anthropogenic emissions and natural processes on PM<sub>10</sub> and PM<sub>2.5</sub> levels. Among the natural contributions, African dust outbreaks originate when air masses above the Sahara Desert move towards the Iberian Peninsula, causing an increase in suspension particle concentrations due to the high mineral dust load they contain. The origin of air masses reaching the study area was analysed

by tracking air mass back-trajectories for each day of the period 2015–2016. Trajectories were obtained using the HYSPLIT-4 Model (hybrid single-particle Lagrangian integrated trajectory) (Draxler and Rolph 2003; Rolph et al. 2017). This model has successfully been applied to interpret the influence of long-range transport on particle levels (Querol et al. 2013) as well as other air pollutants (García et al. 2016). Heights used in the model were selected following a similar method as applied for other studies, taking into account that most aerosols are within the boundary layer, around 1.5 km, this being a representative height of the top of the transport layer and commonly used as a boundary between surface and upper level winds (Katsoulis 1999). Furthermore, aerosols from the desert can move at higher altitudes (up to 5 km) (Escudero et al. 2007; Sánchez et al. 2007; Toledano et al. 2009). Five-day (120 h) backward trajectories commencing at 12:00 GMT were calculated at 750, 1500, and 2500 m (above mean sea level) using the vertical velocity approach. Trajectories reaching the study area were classified into four origins (García et al. 2016): Atlantic (AT), air masses with an origin between the northwest and southwest sectors of the Atlantic Ocean; European (EU), air masses from Europe reaching the sampling site from the northeast to southeast sectors; African (AF), trajectories originating on the African continent and reaching the Peninsula from the south, southwest, or southeast; local conditions (LC), trajectories confined within the Peninsula or its vicinity and travelling short distances. The ANOVA method was also applied to examine whether there were significant differences in concentration means.

In order to establish background particle levels in the sampling area, data affected by specific meteorological conditions or major emission sources were separated from undisturbed conditions. Different procedures can be followed to obtain the representative baseline values (García et al. 2016; Zhou et al. 2004). First, concentration values during calms, which referred to stagnant conditions and those associated with African trajectories, were excluded. Moreover, different restrictions were considered to exclude further data: (a) trajectories of Local Conditions and European origin; (b) values outside the mean  $\pm 3\sigma$  ( $\sigma$  is the standard deviation of all data), in order to avoid the highest concentrations; (c)

values outside the mean  $\pm 1.5\sigma$ ; (d) values above the 90th and below the first percentiles; (e) values above and below the mean  $\pm 1.5$  IR (interquartile range from the upper and lower quartile); (f) additionally, the moving percentile 40 was calculated for each day of measurement, considering the day under evaluation as the central day of the 30-day monthly period (Querol et al. 2013). This method is more restrictive and encompasses the different advections, except African ones.

### Temporal Variations of PM10 and PM2.5

An overall view of PM10 and PM2.5 particle fraction concentrations in the study period, 2015–2016, was performed using data from the four measuring stations in the Valladolid City Council Air Quality Network. PM10 levels for 2015–2016 presented a high range, above  $190 \mu\text{g m}^{-3}$  (Table 1). An analysis of variance was applied, ANOVA, and the multiple range test based on the LSD method used to compare particle concentration means identified three homogenous groups within which there are no statistically significant differences in their means at the 95.0% confidence level. Two of them comprised AL, LR, and PP sampling sites with a mean value around  $17.5 \mu\text{g m}^{-3}$ . However, approximately  $2 \mu\text{g m}^{-3}$  less was recorded in the remaining group, with only one station, VS. The 95th percentile is  $41 \mu\text{g m}^{-3}$ , except for LR, which was  $35 \mu\text{g m}^{-3}$ . Certain variability may be observed in mean PM2.5 levels when the ANOVA analysis is applied. There are statistically significant differences at the 95% confidence level, with means ranging from 8.9 to  $14.8 \mu\text{g m}^{-3}$ , obtained at the PP and LR sampling sites, respectively. Furthermore, the 95th percentile at LR station is higher,  $32 \mu\text{g m}^{-3}$ , compared with that of the remaining stations. The results observed at LR sampling site might be attributable to the greater influence of anthropogenic activity.

The year-on-year comparison, 2015 and 2016, shows that the annual limit values recommended by air quality standards were not exceeded at any of the sampling sites,  $40 \mu\text{g m}^{-3}$  for PM10 and  $25 \mu\text{g m}^{-3}$  for PM2.5 (BOE 2011). There were higher levels of PM10 at all stations in 2015, with concentrations ranging between 16.7 and  $18.9 \mu\text{g m}^{-3}$

**Table 1** Main statistics of PM10 and PM2.5 concentrations at the different monitoring stations

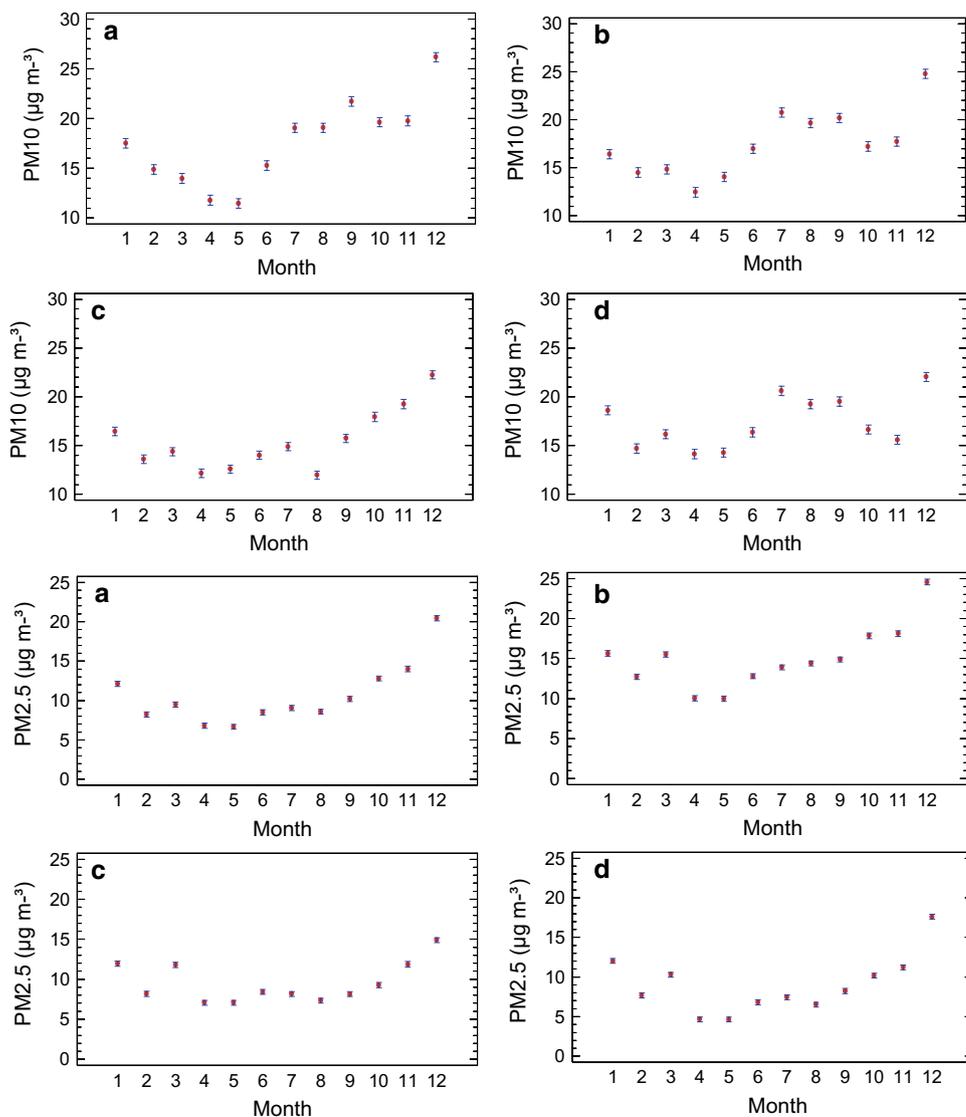
Statistics ( $\mu\text{g m}^{-3}$ )	Arco de Ladrillo		La Rubia		Vega Sicilia		Puente del Poniente	
	PM10	PM2.5	PM10	PM2.5	PM10	PM2.5	PM10	PM2.5
Mean	17.63	10.49	17.49	14.81	15.32	9.44	17.33	8.93
Median	15.00	8.00	15.00	12.00	13.00	7.00	14.00	6.00
Standard deviation	13.61	9.43	13.88	9.34	11.94	8.63	13.25	9.02
Maximum	225.00	97.00	227.00	131.00	191.00	108.00	206.00	104.00
Minimum	1.00	1.00	1.00	1.00	2.00	1.00	1.00	1.00

and reaching a difference of  $4.1 \mu\text{g m}^{-3}$  compared with the mean values obtained in 2016, such as those recorded at the PP station.  $\text{PM}_{2.5}$  concentrations were practically the same for both years at the AL and LR stations ( $10.8$  and  $15.2 \mu\text{g m}^{-3}$ , respectively, and exceeded  $1.4 \mu\text{g m}^{-3}$  in 2015 at the VS and PP sampling sites ( $10.2$  and  $9.8 \mu\text{g m}^{-3}$ , respectively). It should be noted that the results obtained may be related to changes in the controlling factors such as meteorological conditions, intrusions of air masses and anthropogenic activities, which may induce variations in the annual average of the thickest atmospheric particle matter.

The seasonal evolution showed defined patterns. Evolutions of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  concentrations are depicted in Fig. 3 in a means plot in which LSD intervals at the 95% confidence level also are depicted. Maximum mean  $\text{PM}_{10}$  concentrations were observed in autumn and winter, with December values of  $26.2$  and  $24.8 \mu\text{g m}^{-3}$  recorded at the

AL and LR sites, respectively, and approximately  $22 \mu\text{g m}^{-3}$  at the other sampling locations. Changes in particle fraction sources, dust outbreaks, and atmospheric conditions at this time of the year would contribute to the cycle shown. Typical synoptic winter conditions were characterised by anti-cyclonic situations in the area, strong thermal inversions on the surface, and a significant reduction of the mixing layer, leading to atmospheric stagnation. Consequently, there was a lower dispersive capacity in the atmosphere (Artiñano et al. 2001; Viana et al. 2003). A greater accumulation and concentration of pollutants at urban or suburban stations (traffic, heating), as well as at industrial stations was noted, yielding a winter maximum and giving higher concentrations than in the remaining seasons. Many outlier values appeared in February as a result of the natural contribution of African dust as will be shown in the section addressing trajectory analysis. Minimum average values were obtained in spring, April

**Fig. 3** Monthly evolution of mean  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  concentrations in 2015–2016: **a** Arco de Ladrillo, **b** La Rubia, **c** Vega Sicilia, **d** Puente del Poniente. Least significant difference (LSD) intervals at the 95% confidence level are shown

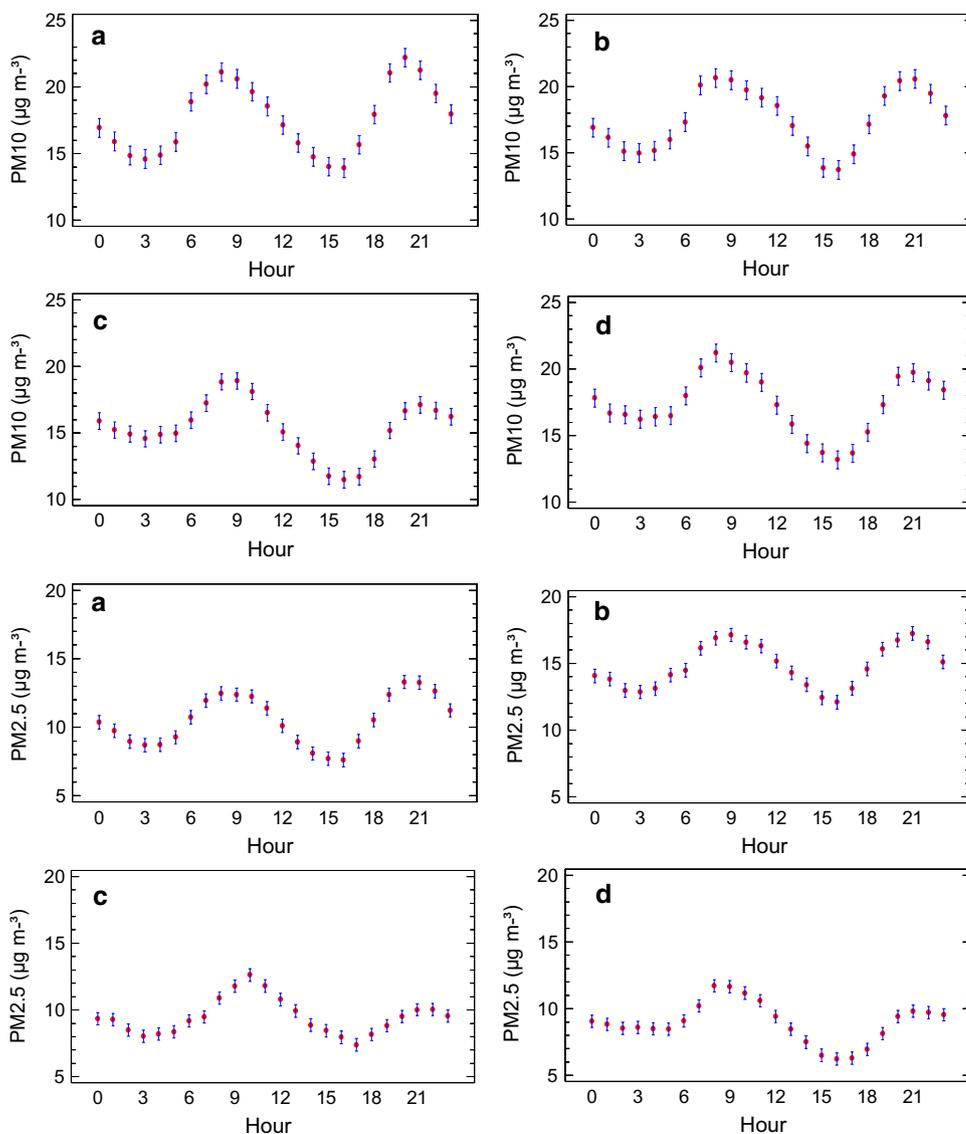


and May. Particularly worthy of note is the  $12 \mu\text{g m}^{-3}$  mean value recorded at all the sampling stations, except PP, with  $14 \mu\text{g m}^{-3}$  in April. This behaviour in spring may be related to atmospheric instability, Atlantic airmass advections, and the highest rainfall recorded this month in 2 years. There was an inverse relationship between particle concentration and precipitation as an atmospheric wash occurs, which was an efficient particle removal process (Inza et al. 2005). In addition, not only were low levels of particles recorded during rain events but also in a later period since processes, such as resuspension were inhibited. Furthermore, less precipitation, greater particle resuspension, formation of secondary particles by photochemical processes, and dust outbreaks contributed to increased PM10 concentrations in summer months (Escudero et al. 2005; Viana et al. 2002). The mean value increased significantly, up to  $4\text{--}5 \mu\text{g m}^{-3}$ , at all measuring stations except the VS station,  $2 \mu\text{g m}^{-3}$ . The PM2.5 particle

fraction showed a similar evolution than that of PM10. However, the maximum mean value obtained in December varied between stations, ranging between  $15 \mu\text{g m}^{-3}$  at VS station and  $25 \mu\text{g m}^{-3}$  at LR station. The minimum value obtained in April–May was between 5 and  $10 \mu\text{g m}^{-3}$  at PP and LR stations, respectively.

Further analysis of atmospheric PM10 and PM2.5 concentrations showed the variability of the diurnal pattern. This provided important information for identifying potential emission sources and the time of day when maximum levels are recorded. Figure 4 shows hourly means plots of PM10 and PM2.5 concentrations. Two well-defined PM10 maxima were observed in the morning between 7:00–9:00 GMT and 20:00–22:00 GMT in the evening corresponding to traffic exhaust emission peaks and human activities. The mean value associated was nearly  $21 \mu\text{g m}^{-3}$ , although nocturnal peaks at VS and PP were smaller. The highest

**Fig. 4** Daily evolution of mean PM10 and PM2.5 concentrations in 2015–2016: **a** Arco de Ladrillo, **b** La Rubia, **c** Vega Sicilia, **d** Puente del Poniente. Least significant difference (LSD) intervals at the 95% confidence level are shown



average values for PM<sub>2.5</sub> were also obtained approximately at the same time intervals as for PM<sub>10</sub>. The LR station presented the highest mean values in the morning, approximately  $17 \mu\text{g m}^{-3}$ , whereas the value for the remaining stations was  $5 \mu\text{g m}^{-3}$  lower. However, the lowest mean values were obtained at 3:00–4:00 GMT in the morning and 15:00–16:00 GMT in the afternoon. PM<sub>10</sub> values ranged from 11 to  $16 \mu\text{g m}^{-3}$  at the VS and PP stations, respectively. As regards PM<sub>2.5</sub> (Fig. 4), mean values were between 6 and  $8 \mu\text{g m}^{-3}$ , except at the LR station, which had higher values, 12– $13 \mu\text{g m}^{-3}$ . Hourly outlier values were observed up to  $200 \mu\text{g m}^{-3}$  for PM<sub>10</sub>. Moreover, extreme PM<sub>2.5</sub> values were above  $120 \mu\text{g m}^{-3}$  at LR site and reached  $100 \mu\text{g m}^{-3}$  at the remaining measuring stations. Taking into account current regulations for daily PM<sub>10</sub> concentrations,  $50 \mu\text{g m}^{-3}$ , which must not be exceeded more than 35 times per year (BOE 2011), and  $25 \mu\text{g m}^{-3}$  for PM<sub>2.5</sub> according to the WHO (World Health Organization) (2013), the number of daily exceedances in 2015 and 2016 are presented in Table 2. Compliance with limit values at some locations was conditioned by the influence of natural phenomena such as particle transport from arid regions.

An anthropogenic effect on urban particle levels can also be observed by identifying the weekly variability of PM<sub>10</sub> and PM<sub>2.5</sub> concentrations. Daily mean values were considered in order to appreciate the changes in PM concentrations between weekdays and weekends. The mean particle concentration in Table 3 shows a decrease in PM<sub>10</sub> values at weekends compared to the average values of weekdays, associated with changes in anthropogenic activities, especially traffic, although meteorological conditions also influence the variability of particle levels (Giet and Klemm 2009). The mean value on weekdays is approximately  $18 \mu\text{g m}^{-3}$  at most stations, except VS station, which recorded  $16 \mu\text{g m}^{-3}$ . However, 16 and  $14 \mu\text{g m}^{-3}$  are obtained at weekends, respectively. These results are assessed applying analysis of variance to particle concentrations. The mean at the VS station presented significant differences from the other means at the 95% confidence level on weekdays and at weekends. PM<sub>10</sub> concentration fell by 17% at the LR station when comparing mean values obtained on weekdays and those at weekends, and the smallest difference, 14%, is found for the VS and PP stations. Greater variability on weekdays is seen for the PM<sub>2.5</sub> particle fraction, ranging from 9.3 to  $15.7 \mu\text{g m}^{-3}$

**Table 3** Mean concentrations and standard deviation of PM<sub>10</sub> and PM<sub>2.5</sub> for weekdays and weekends

Station	PM <sub>10</sub> ( $\mu\text{g m}^{-3}$ )		PM <sub>2.5</sub> ( $\mu\text{g m}^{-3}$ )	
	Weekend	Weekdays	Weekend	Weekdays
Arco de Ladrillo	$15.6 \pm 9.7$	$18.4 \pm 10.7$	$9.9 \pm 7.1$	$11.1 \pm 7.5$
La Rubia	$15.2 \pm 9.5$	$18.4 \pm 10.9$	$13.8 \pm 7.0$	$15.7 \pm 7.9$
Vega Sicilia	$13.8 \pm 8.1$	$15.9 \pm 9.6$	$8.8 \pm 5.9$	$9.9 \pm 6.7$
Puente del Poniente	$15.5 \pm 9.2$	$18.1 \pm 10.5$	$8.3 \pm 6.9$	$9.3 \pm 7.1$

at the PP and LR stations, respectively. The multiple range test identified three homogenous groups of means on weekdays within which there are no statistically significant differences at the 95% confidence level; one group with the PP and VS sampling sites having the lowest mean concentrations, another group with the AL station, and with the highest concentration being related to the LR station. Mean values at weekends were between 8.3 and  $13.8 \mu\text{g m}^{-3}$ , at the same stations as on weekdays at the PP and LR stations, respectively. The largest reduction in mean concentrations was observed at the LR station, 12%, although it was only slightly higher than that recorded at the remaining stations, 11%. The statistical analysis identified similar groups to those found on weekdays.

### Relationship Between PM<sub>2.5</sub> and PM<sub>10</sub>

The PM<sub>2.5</sub>/PM<sub>10</sub> ratio has previously been studied using data from many locations on Earth and is highly variable due to geological, climatological, and atmospheric features, as well as pollutant sources that condition the size of the particle matter measured at different locations, even within the same city. Average values are between 0.40 and 0.74 in Spain (Querol et al. 2004a; Rojas 2005).

The PM<sub>2.5</sub>/PM<sub>10</sub> ratios for measuring stations in Valladolid using daily data are shown in Table 4, together with the Pearson correlation coefficients between the two variables. Values are within the range 0.49–0.95. A ratio above 0.6 may indicate the direct influence of combustion sources on coarse particles rather than a resuspension of dust or a wind event (Querol et al. 2004b). A value of 0.6 was obtained at the AL and VS stations, which is similar to that for areas in the north, northwest and centre of Spain, with values between

**Table 2** Number of exceedances of daily PM<sub>10</sub> and PM<sub>2.5</sub> levels

Year/Station	Arco de Ladrillo		La Rubia		Vega Sicilia		Puente del Poniente	
	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>	PM <sub>10</sub>	PM <sub>2.5</sub>
2015	7	27	7	37	4	23	5	18
2016	3	17	4	39	2	4	2	9

Reference limits:  $50 \mu\text{g m}^{-3}$  for PM<sub>10</sub> (BOE 2011) and  $25 \mu\text{g m}^{-3}$  for PM<sub>2.5</sub> (WHO 2013)

**Table 4** PM<sub>2.5</sub>/PM<sub>10</sub> ratio of daily overall data and the relationship between PM<sub>10</sub> and PM<sub>2.5</sub> concentrations (confidence level 99%)

Station	PM <sub>2.5</sub> /PM <sub>10</sub> ratio	Correlation coefficient
Arco de Ladrillo	0.6 ± 0.2	0.792
La Rubia	0.9 ± 0.3	0.804
Vega Sicilia	0.6 ± 0.2	0.831
Puente del Poniente	0.5 ± 0.2	0.818

0.6 and 0.7 (Viana et al. 2003). The highest value found at the LR station, 0.9, showed that the fine particle fraction was the predominant component of particulate matter in this area, higher than in the remaining stations. In contrast, a value of 0.5 is obtained at the PP sampling site, which may indicate the greater contribution of coarse particles as was also observed from the statistics of the concentration results.

Although the Pearson correlation coefficients of the linear fits calculated from the daily concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> particle fractions showed a strong relationship, with values between 0.792 and 0.831 at the AL and VS stations, respectively, they were not good enough either to indicate whether variations in PM<sub>2.5</sub> and PM<sub>10</sub> concentrations may be due to the same phenomenon or to accurately predict PM<sub>2.5</sub> concentrations from the PM<sub>10</sub> values at these stations.

### Trajectory Analysis

The air mass trajectories calculated for the study period using the HYSPLIT model and following the procedure given in the “Materials and Methods” section were analysed. The frequency of the four origins was similar for the all sampling sites. For the 750-m level, Atlantic trajectories had the highest frequency of occurrence, 54%, nine times greater than the frequency of the African trajectories (AF).

A greater percentage of these trajectories were observed at higher levels, 1500 and 2500 m, increasing up to 12% of occurrences. Trajectories arrived from Europe (EU) with approximately the same frequency at both levels, 750 and 1500 m (21%), whereas frequency was lower at 2500 m (17%). Trajectories associated with LC were frequent at low levels (18% and 13% at 750 and 1500 m, respectively). The main particle levels of the air mass trajectories for each monitoring station are shown in Table 5 for PM<sub>10</sub> and PM<sub>2.5</sub> particle fractions. The highest PM<sub>10</sub> concentration levels were observed for the AF trajectories at all the stations, with a mean value of 27.9 µg m<sup>-3</sup> at 2500 m obtained at the AL station, although this kind of trajectory was not common. AT origins presented similar values at all heights, around 14.5 µg m<sup>-3</sup>. The occurrence of EU trajectories provided a mean value close to 19 µg m<sup>-3</sup> at 750 m at the AL and LR stations and 1.4 µg m<sup>-3</sup> less at the VS and PP stations. Lower mean concentrations were recorded at the other heights. As regards days assigned to LC, higher mean values were obtained at lower heights in most of the stations, 24 µg m<sup>-3</sup>. Similar behaviour to the PM<sub>10</sub> is found for the PM<sub>2.5</sub> particle fraction. AF trajectories provided the highest concentrations at more elevated heights, with a maximum value of 21.4 µg m<sup>-3</sup> at LR station. The contribution of AT origins was similar at all heights, with an average value between 6.8 and 12.8 µg m<sup>-3</sup> at the PP and LR stations, respectively. EU trajectories presented mean PM<sub>2.5</sub> values of approximately 12 µg m<sup>-3</sup> at AL and VS stations at all heights. The highest mean value was recorded at the LR station, 17 µg m<sup>-3</sup>. As regards LC trajectories, lower heights were associated with higher particle concentrations, reaching 20 µg m<sup>-3</sup> at the LR sampling site. Mean PM<sub>2.5</sub> levels were lower at the other stations, with concentrations between 11 and 13 µg m<sup>-3</sup> at the VS and PP stations, respectively.

Analysis of the variance of mean PM<sub>10</sub> concentrations using trajectories as a factor revealed that the means associated to AF and LC trajectories did not show statistically

**Table 5** Mean PM<sub>10</sub> and PM<sub>2.5</sub> concentrations as a function of height for each trajectory at the measuring sites

	AF			AT			EU			LC		
	750	1500	2500	750	1500	2500	750	1500	2500	750	1500	2500
PM <sub>10</sub> (µg m <sup>-3</sup> )/height (m)												
Arco de Ladrillo	21.9	25.3	27.9	14.4	14.7	14.9	18.6	17.8	17.5	24.4	24.2	22.4
La Rubia	20.5	24.4	27.6	14.5	14.7	14.9	18.8	17.8	17.4	24.0	24.1	22.1
Vega Sicilia	19.0	22.2	24.4	12.5	12.9	13.1	17.4	16.7	16.3	20.4	19.1	17.8
Puente del Poniente	20.4	23.6	26.9	14.1	14.4	14.6	19.7	18.6	18.3	23.5	23.8	21.8
PM <sub>2.5</sub> (µg m <sup>-3</sup> )/height (m)												
Arco de Ladrillo	13.6	16.1	16.9	8.1	8.5	8.8	12.5	12.3	12.3	15.4	14.4	13.2
La Rubia	16.8	20.0	21.4	12.5	12.8	13.2	17.3	16.9	16.8	20.1	19.5	17.5
Vega Sicilia	10.2	13.0	14.2	7.3	7.6	7.9	12.2	11.7	11.6	13.0	12.4	11.2
Puente del Poniente	11.1	13.8	14.8	6.5	6.8	7.1	11.0	10.7	10.7	13.4	12.7	11.3

Trajectory origins: AF African, AT Atlantic, EU European, LC Local conditions

significant differences at the 95% confidence level in most stations when considering the lowest heights, 750 and 1500 m. However, means showed significant differences at all trajectories for the 2500 m height.

### Influence of Saharan Dust Outbreaks on Particle Concentrations

The results obtained in the previous section regarding air mass trajectories were suitable for determining the strong influence of African dust outbreaks on particle concentrations, which depends on the frequency and intensity of episodes occurring in the study period. Days with trajectories of African origin at any of the heights applied were considered and PM<sub>10</sub> and PM<sub>2.5</sub> concentration dataset analysed. Results allowed us to identify the number of outbreak events reaching the sampling sites, their duration, and the mean concentration during the days of intrusion over the 2 years (Table 6). The greatest persistence of African air mass events occurred in April and December 2015, up to 8 days, and in July and December 2016, 5 days. Isolated days with little impact on particle levels also were seen. Episodes are dominated by anticyclonic meteorological situations or high pressures located in north or northeast Africa. During 2015, significant intrusion episodes because of their mineral dust load, mainly PM<sub>10</sub>, provided a mean value of between 20 and 30  $\mu\text{g m}^{-3}$  in most months; between 30 and 40  $\mu\text{g m}^{-3}$  in March, April, August, November, and December; > 40  $\mu\text{g m}^{-3}$  in December with two persistent occurrences, which did not on average exceed 45  $\mu\text{g m}^{-3}$  for PM<sub>10</sub> and 39  $\mu\text{g m}^{-3}$  for PM<sub>2.5</sub>. However, in 2016, Saharan dust episodes did not reach 20  $\mu\text{g m}^{-3}$  in April, May, November, and December; they were between 20 and 30  $\mu\text{g m}^{-3}$  in most months; between 30 and 40  $\mu\text{g m}^{-3}$  in events occurring in July, September and November; > 40  $\mu\text{g m}^{-3}$  on one isolated day in September and in February, when a spectacular episode contributed to a mean concentration value of 102.5  $\mu\text{g m}^{-3}$  at LR station (84  $\mu\text{g m}^{-3}$  were exceeded as a global mean) for the 2 days it lasted. Average PM<sub>2.5</sub> concentrations did not show a significant increase with intrusion events observed mainly in summer. However, they were strongly influenced in the February 2016 episode and in some events in autumn–winter (reaching 30  $\mu\text{g m}^{-3}$ ), although only 10% of mean concentrations exceeded 25  $\mu\text{g m}^{-3}$ .

#### Study Case: February 21–22, 2016 Episode

The analysis of a strong Saharan dust outbreak during the study period was included in this paper to show the importance that long-range transport of African dust might have in northern Spain. During intense Saharan dust outbreaks, substantial concentrations of particulate matter were recorded

and might have an impact on human health, ecosystems, climate, and materials.

The period February 21–22, 2016 was characterised by a strong influence of a dust outbreak on the city, which was noted in the data recorded at the measuring sites. The prevailing synoptic meteorological situation in those days confined the Iberian Peninsula to an anticyclonic situation centred on the Azores, as well as in eastern Spain and northern Africa. As a result, warm African air masses reached Spain. The situation changed on the 24 with the arrival of cold polar air, affecting the whole of the Iberian Peninsula on February 27. Backward air mass trajectories on February 21 (left) and 22 (right) performed with the HYSPLIT model are depicted in Fig. 5. The trajectory associated to 2500-m height presented an African origin and differs from those corresponding to the lower altitudes, with an advection of air masses commencing in the Atlantic and entering the southeast of the Peninsula. On February 22, the trajectory associated to 2500-m height also showed an African origin, and the trajectories linked to lower heights modified their pathway. The 750-m height trajectory is associated with LC, and the 1500-m height trajectory displayed a shorter trajectory, originating over the Atlantic Ocean. In addition, there were no air masses of African origin on February 20 and 23.

The impact of Saharan dust outbreaks on hourly PM<sub>10</sub> and PM<sub>2.5</sub> concentrations was spectacular (Fig. 6). It was not persistent, lasting only 2 days, but was very intense with PM<sub>10</sub> concentrations ranging between 84.4 and 102.5  $\mu\text{g m}^{-3}$  recorded at the VS and LR sites, respectively (Table 7). Maximum values were observed from 16:00 GMT (February 21), when they began to increase, to 17:00 GMT (February 22) when they decreased. Two peaks were recorded at 18:00–19:00 GMT and 10:00–11:00 GMT, reaching 225  $\mu\text{g m}^{-3}$  at the AL and LR stations, and approximately 200  $\mu\text{g m}^{-3}$  at the VS and PP stations. Similar behaviour was seen in this episode for PM<sub>2.5</sub> concentrations, with high mean values between 27.2 and 35.4  $\mu\text{g m}^{-3}$  being recorded at the VS and LR sites, respectively. PM<sub>2.5</sub> concentrations ranged between 29.9 and 36.3  $\mu\text{g m}^{-3}$  at AL and LR stations, respectively, on February 22. Maximum values occurred at a similar time interval to PM<sub>10</sub>. The peak value recorded on February 21 ranged between 67.0 and 91.0  $\mu\text{g m}^{-3}$  at the VS and LR stations, respectively. The second peak was slightly above 100  $\mu\text{g m}^{-3}$  at the PP sampling site, a value not reached at the VS station and which did not exceed 80  $\mu\text{g m}^{-3}$  at the AL and LR, stations.

#### PM<sub>10</sub> and PM<sub>2.5</sub> Background Concentrations

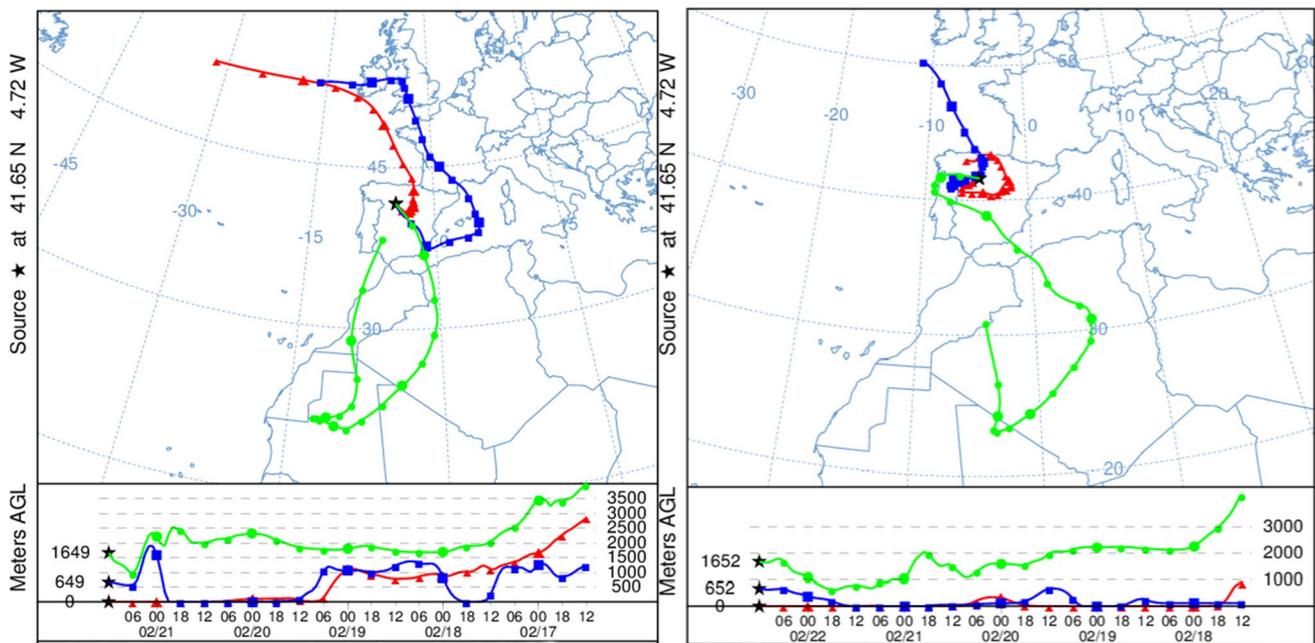
The results obtained in the previous sections were suitable for determining the representative conditions of the background particle matter level at the sampling sites. This is a

**Table 6** Monthly study of Saharan dust outbreaks calculated using the HYSPLIT model and mean PM10 and PM2.5 concentrations ( $\mu\text{g m}^{-3}$ ) obtained in 2015 and 2016 during intrusion days

	No. of events	No. of days	Arco Ladrillo		Rubia		Vega Sicilia		Puente Poniente	
			PM10	PM2.5	PM10	PM2.5	PM10	PM2.5	PM10	PM2.5
2015										
January	1	2	25.1	23.6	22.4	21.3	25.2	22.3	31.9	23.0
February	1	2	22.3	16.9	23.8	21.0	22.4	17.5	29.2	16.3
March	2	1	29.5	21.4	32.2	30.7	30.7	23.3	39.0	24.4
		3	13.3	11.8	12.9	16.5	14.1	13.1	18.7	14.0
April	1	4	31.8	9.7	32.5	14.9	33.0	12.0	40.1	10.9
May	1	3	23.9	10.5	30.3	17.3	20.4	12.6	31.2	11.1
June	2	1	19.2	11.9	18.3	15.7	14.9	11.6	21.1	11.1
		3	26.2	12.9	26.6	19.6	23.4	12.3	28.5	12.4
July	2	1	27.2	10.6	31.1	16.8	25.4	9.5	28.8	9.9
		2	24.9	10.1	25.0	15.3	20.0	8.9	26.9	10.0
August	1	2	35.5	9.1	36.9	16.5	28.7	12.6		
September	1	2	12.8	5.8	11.6	10.5	12.3	6.5	15.9	4.0
October	4	2	24.8	5.8	19.7	14.3	22.8	8.1	21.4	8.3
		3	17.3	7.3	11.9	13.4	14.5	7.2	12.6	6.5
		3	19.9	11.4	15.9	16.3	16.9	10.3	14.7	10.3
November	2	1	21.3	13.8	16.7	16.2	19.3	10.4	16.8	10.6
		2	30.5	20.5	27.5	24.4	31.5	18.7	24.1	17.1
December	4	1	27.8	18.7	24.7	22.3	25.4	16.5	22.1	14.6
		8	42.8	35.5	42.7	39.3	42.7	31.6	35.2	29.7
		6	44.6	23.3	42.8	27.9	35.2	17.4	38.7	22.9
		1	32.8	18.9	31.9	28.1	27.4	16.5	27.1	19.0
		1	23.0	13.8	20.7	18.2	18.3	12.1	18.8	13.4
2016										
January	1	3	26.2	17.9	24.4	24.1	19.8	13.3	19.7	14.0
February	2	1	25.5	16.7	22.9	21.7	18.3	12.3	19.5	14.9
		2	97.1	28.9	102.5	35.4	84.4	27.2	90.9	34.5
March	0									
April	1	1	7.2	7.0	9.3	9.3	8.8	11.3	10.5	20.3
May	2	2	8.0	6.4	7.9	8.6	9.7	5.0	9.8	2.3
		3	6.9	4.9	6.9	7.1	7.2	3.4	7.3	1.6
June	1	1	20.3	11.5	28.5	15.7	15.3	9.0	20.8	5.9
July	2	4	19.2	10.8	19.4	13.6	11.8	8.7	18.9	7.6
		1	40.5	13.3	46.9	18.4	36.2	14.6	42.2	10.9
August	2	2	25.9	11.1	27.0	18.3	9.7	9.5	25.7	8.1
		3	29.5	16.4	29.7	22.1	15.4	13.3	30.6	13.1
September	3	1	49.9	18.6	51.7	29.1	25.6	11.9	48.9	16.3
		2	38.0	20.1	40.0	25.4	15.7	11.1	33.1	15.3
October	2	1	26.7	15.3	28.1	21.5	25.9	12.3	24.6	13.2
		1	23.6	19.5	24.8	23.3	25.2	13.5	23.1	14.4
November	2	5	21.2	21.0	24.9	24.7			23.0	16.5
		4	36.9	28.5	27.1	25.3			26.3	17.4
		4	13.0	12.4	12.9	15.9	15.2	8.7	10.7	8.0

**Table 6** (continued)

	No. of events	No. of days	Arco Ladrillo		Rubia		Vega Sicilia		Puente Poniente	
			PM10	PM2.5	PM10	PM2.5	PM10	PM2.5	PM10	PM2.5
December	5	2	27.8	22.8	27.0	27.9	29.3	19.9	22.9	14.4
		4	16.1	14.9	14.9	17.5	15.2	10.3	14.4	12.1
		3	24.2	23.5	20.2	23.8	14.8	13.8	21.5	21.1
		1	13.4	13.4	14.3	16.8	11.8	10.9	13.2	13.4
		1	15.0	13.1	14.2	18.5	14.2	12.9	13.8	13.8

**Fig. 5** 5-d backward air mass trajectories computed by HYSPLIT at 2500 m (circles), 1500 m (squares), and 750 m (triangles), reaching the measuring area at 12 GMT; February 21 (left) and 22 (right)

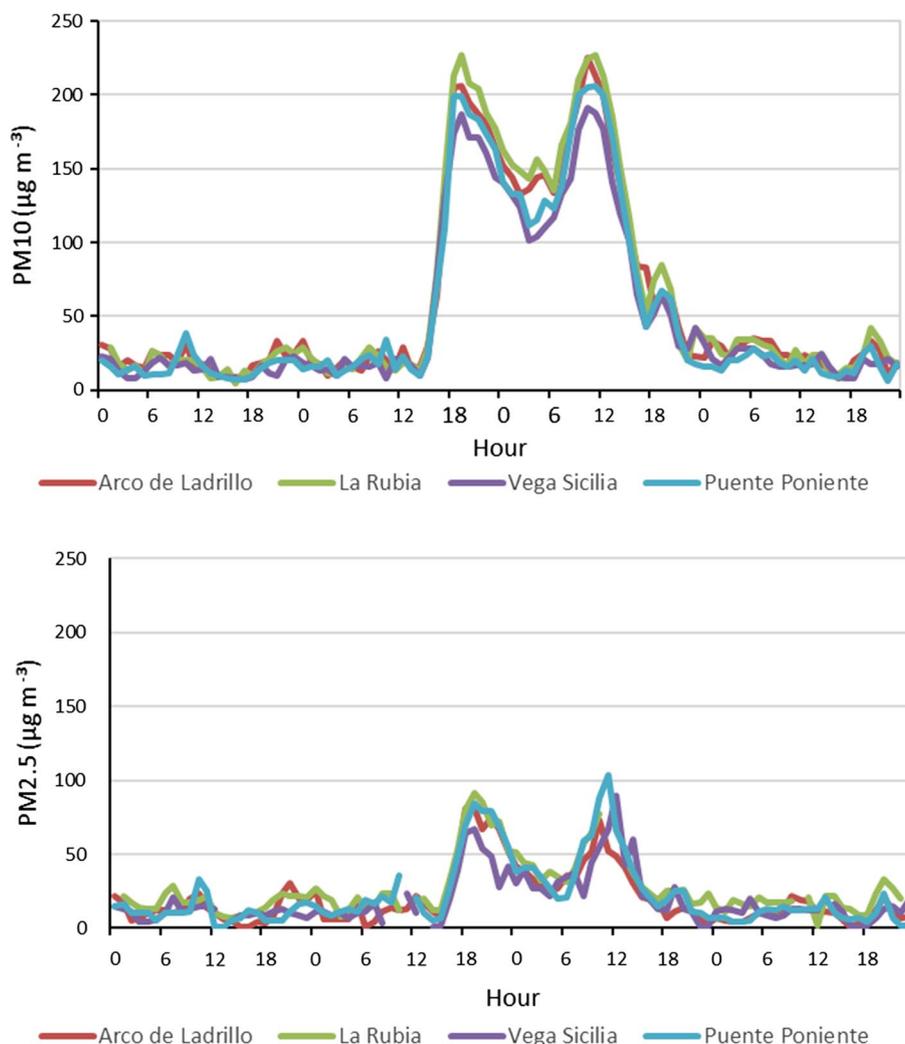
way to distinguish the contribution of potential sources of interference and to avoid concentration excess.

Table 8 shows the results of the mean PM10 and PM2.5 concentrations using the remaining data in each case once the different restrictions were applied for each measuring station and trajectory height according to the procedure presented in the “Materials and Methods” section. The range of mean concentrations obtained at the AL, LR, and PP measuring sites with regard to all heights, was around  $3 \mu\text{g m}^{-3}$  and  $2 \mu\text{g m}^{-3}$  for PM10 and PM2.5 particle fractions, respectively (except  $4.5 \mu\text{g m}^{-3}$  for PM10 at the PP site at 750-m height). However, at the VS station, a lower PM10 range was obtained, around  $2 \mu\text{g m}^{-3}$  and a higher one for PM2.5, approximately  $3 \mu\text{g m}^{-3}$  at 750 m height. In general, extreme mean values were obtained for the 40th percentile and mean  $\pm 3\sigma$  restrictions. The criterion of excluding Local

Conditions and European trajectories led to lower average concentrations within heights.

Excluding the extreme average values obtained when different criteria were applied, the average concentrations are similar at all heights for each station. The results obtained for the PM10 particle fraction show baseline values of around  $15 \mu\text{g m}^{-3}$  in the area around the AL, LR, and PP stations, and  $2 \mu\text{g m}^{-3}$  lower at the VS sampling site. Results are within the range found at other times (1996–2000) in regional background stations (Querol et al. 2004b),  $15\text{--}20 \mu\text{g m}^{-3}$ . The differences between the stations were more evident for the PM2.5 particle fraction,  $8.5$ ,  $7.7$ , and around  $7 \mu\text{g m}^{-3}$  at the AL, VS, and PP stations, respectively. Consequently, a mean value of  $7.7 \mu\text{g m}^{-3}$  can be considered as the background level for the area of these measuring stations. However, the highest average value,  $12.9 \mu\text{g m}^{-3}$ , was obtained at LR station and was influenced by anthropogenic emissions, such as traffic.

**Fig. 6** Hourly concentration evolution of PM<sub>10</sub> and PM<sub>2.5</sub> from 20 to 23 February 2016



**Table 7** Mean concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> during the February 21–22 episode, 2016

Day/concentration ( $\mu\text{g m}^{-3}$ )	Arco Ladrillo		La Rubia		Vega Sicilia		Puente Poniente	
	PM <sub>10</sub>	PM <sub>2.5</sub>						
February 21, 2016	68.8	28.0	72.7	34.5	61.4	23.2	63.4	32.0
February 22, 2016	125.5	29.9	132.3	36.3	107.3	31.3	116.6	37.1
Episode mean	97.1	28.9	102.5	35.4	84.4	27.2	91.0	34.5

## Conclusions

Analysis of PM<sub>10</sub> and PM<sub>2.5</sub> particle concentrations recorded over 2 years, 2015–2016, at four air quality stations in an urban area, Valladolid, on the northern Spanish plateau, revealed that they did not exceed air quality regulations (current annual limit for PM<sub>10</sub>,  $40 \mu\text{g m}^{-3}$  and for PM<sub>2.5</sub>,  $25 \mu\text{g m}^{-3}$ ). The overall mean value for the PM<sub>10</sub> fraction was approximately  $17.5 \mu\text{g m}^{-3}$  at most stations. Greater variability was found for the PM<sub>2.5</sub> particle fraction with mean values ranging between 8.9 and

$14.8 \mu\text{g m}^{-3}$  at the PP and LR stations, respectively. The highest monthly means were recorded in autumn and winter and did not exceed  $26.2 \mu\text{g m}^{-3}$  for the PM<sub>10</sub> fraction. The lower dispersive capacity of the atmosphere in this period was a significant controlling factor. Minimum average values were obtained in spring: April and May. The PM<sub>2.5</sub> fraction shows a similar evolution to that of PM<sub>10</sub>. The maximum average value obtained in December varied between sampling stations and was between approximately 25 and  $15 \mu\text{g m}^{-3}$  at the LR and VS stations, respectively. Spring minimum values reached  $10 \mu\text{g m}^{-3}$ . Hourly maximum PM<sub>10</sub> values ranged between 191 and  $227 \mu\text{g m}^{-3}$

**Table 8** Mean PM10 and PM2.5 concentrations after applying different data selection criteria for each measuring station

Mean concentration Data excluded/height (m)	PM10 ( $\mu\text{g m}^{-3}$ )			PM2.5 ( $\mu\text{g m}^{-3}$ )		
	750	1500	2500	750	1500	2500
<b>Arco Ladrillo</b>						
Calms and AF						
LC and EU	14.3±8.3	14.5±9.4	14.7±7.8	8.0±5.6	8.2±5.8	8.6±6.0
Mean ± 3 $\sigma$	16.4±8.2	16.1±8.0	15.8±7.8	9.9±6.2	9.7±6.0	9.5±5.9
Mean ± 1.5 $\sigma$	15.5±7.1	15.2±6.9	14.4±6.2	8.8±4.7	8.6±4.6	8.3±4.3
1% < data < 90%	14.9±6.4	14.6±6.2	14.3±6.0	8.6±4.4	8.4±4.2	8.2±4.1
Mean ± 1.5IR	15.8±7.4	15.5±7.2	15.1±7.0	8.9±4.8	8.7±4.7	8.4±4.4
40th percentile	13.6±4.4	13.2±4.2	13.0±4.1	7.9±3.3	7.7±3.0	7.5±3.0
Baseline	15.1	15.0	14.6	8.6	8.5	8.4
<b>La Rubia</b>						
Calms and AF						
LC and EU	14.4±8.2	14.5±9.4	14.7±7.7	12.3±5.7	12.6±5.8	13.0±6.2
Mean ± 3 $\sigma$	16.3±8.2	16.1±8.0	15.7±7.7	14.5±6.6	14.2±6.3	13.9±6.2
Mean ± 1.5 $\sigma$	15.4±7.1	15.0±6.7	14.5±6.0	13.1±4.8	13.0±4.8	12.7±4.5
1% < data < 90%	14.8±6.2	14.9±6.1	14.2±5.9	13.0±4.6	12.9±4.5	12.6±4.2
Mean ± 1.5IR	15.7±7.4	15.3±7.1	15.0±6.9	13.3±5.1	13.2±5.0	12.8±4.7
40th percentile	13.4±3.8	13.0±3.6	12.8±3.6	12.3±3.3	12.0±3.0	11.9±2.9
Baseline	15.1	14.9	14.6	13.1	13.0	12.8
<b>Vega Sicilia</b>						
Calms and AF						
LC and EU	12.4±7.0	12.8±8.2	12.9±6.8	7.2±4.6	7.5±5.1	7.8±5.3
Mean ± 3 $\sigma$	14.4±7.1	14.0±6.7	13.5±6.2	8.9±5.2	8.6±5.0	8.3±4.6
Mean ± 1.5 $\sigma$	13.4±5.7	13.1±5.5	12.4±4.9	8.0±3.9	7.8±3.7	7.6±3.5
1% < data < 90%	12.9±5.1	12.7±4.9	12.3±4.6	7.7±3.5	7.6±3.3	7.4±3.2
Mean ± 1.5IR	13.5±5.9	13.3±5.7	12.7±5.2	7.9±3.7	7.8±3.6	7.6±3.4
40th percentile	13.2±3.6	11.5±2.9	11.4±2.9	6.1±2.8	6.8±2.0	6.7±1.9
Baseline	13.3	13.0	12.6	7.7	7.7	7.6
<b>Puente del Poniente</b>						
Calms and AF						
LC and EU	14.0±7.7	14.2±8.9	14.5±7.7	6.4±5.3	6.6±5.5	6.9±5.6
Mean ± 3 $\sigma$	16.4±8.4	16.0±8.0	15.4±7.4	8.2±5.9	8.0±5.7	7.7±5.5
Mean ± 1.5 $\sigma$	15.0±6.6	14.7±6.4	14.1±5.9	7.1±4.3	6.9±4.2	6.6±4.0
1% < data < 90%	14.7±6.1	14.4±5.9	14.0±5.6	7.0±4.1	6.8±3.9	6.5±3.8
Mean ± 1.5IR	15.3±6.9	14.9±6.6	14.5±6.3	7.2±4.4	7.0±4.3	6.8±4.1
40th percentile	11.8±3.2	12.8±3.5	12.6±3.4	7.1±2.3	5.9±2.6	5.7±2.6
Baseline	14.8	14.6	14.3	7.1	6.8	6.7

IR interquartile range

Trajectory origins: AF African, EU European, LC local conditions

and between 97 and 131  $\mu\text{g m}^{-3}$  for PM2.5 at the sampling sites, with the highest values being obtained at the LR station. Daily patterns showed the highest levels of particles at two intervals, one in the morning, 7:00–9:00 GMT, and the other in the evening, 20:00–22:00 GMT, mainly related to anthropogenic activity. The mean value of PM10 concentrations was approximately 21  $\mu\text{g m}^{-3}$  in the morning. PM2.5 mean concentrations varied between 17  $\mu\text{g m}^{-3}$  at the LR station and 12  $\mu\text{g m}^{-3}$  at the remaining stations. The nocturnal peak for both fractions was slightly

lower and covered a wide range. The lowest concentrations occurred at night and during the afternoon. A similar controlling factor to that mentioned above concerns changes in weekly concentrations, causing an increase on weekdays and a significant decrease at weekends. Mean PM10 concentration on weekdays was around 18  $\mu\text{g m}^{-3}$  at most of the sampling sites. However, means reached up to 16  $\mu\text{g m}^{-3}$  at weekends. The VS sampling site provided the lowest mean values. PM2.5 concentration means ranged between 9.3 and 15.7  $\mu\text{g m}^{-3}$  on weekdays, whereas they

were lower at weekends: 8.3 and 13.8  $\mu\text{g m}^{-3}$ . The highest and lowest values were obtained at the LR and PP stations, respectively.

The PM<sub>2.5</sub>/PM<sub>10</sub> ratio ranged between 0.49 obtained at the PP station and 0.95 at the LR sampling site, which had the highest presence of the finest particles. Considering all the stations, an average of 0.67 was obtained, similar to that presented in other studies for this area of Spain.

Air masses of African origin over the sampling site showed variability in altitude and, mainly at high altitudes, contributed to increasing particulate matter concentrations. However, the Atlantic origin was related to lower mean concentrations. In general, Saharan dust outbreaks lasted 2–3 days, although one lasted 8 days in December 2015 and others lasted 4–5 days in April 2015 and July, October–December 2016. Most events were characterised by a mean concentration between 20 and 30  $\mu\text{g m}^{-3}$  for PM<sub>10</sub>, with an overall mean of 25  $\mu\text{g m}^{-3}$ . The corresponding value for the PM<sub>2.5</sub> concentration was 15  $\mu\text{g m}^{-3}$ , with higher values mainly in autumn–winter. The episode on February 21–22, 2016, presented backward trajectories at 2500-m height with African origin that entered the Peninsula from the southeast. Results were consistent with high pressures affecting the Peninsula, which favoured the entry of warm African air masses. Average concentrations over this 2-day episode were between 84 and 102  $\mu\text{g m}^{-3}$  for the PM<sub>10</sub> fraction and 27 and 35  $\mu\text{g m}^{-3}$  for the PM<sub>2.5</sub> fraction. Finally, different procedures were used to provide representative baseline conditions for the measuring site. Background concentrations estimated for PM<sub>10</sub> were between 13  $\mu\text{g m}^{-3}$  at VS station and 15  $\mu\text{g m}^{-3}$  at the other stations. Although greater differences were found between stations for PM<sub>2.5</sub>, a mean value of 7.7  $\mu\text{g m}^{-3}$  could be considered, except for the LR station, which had the highest background concentration, 12.9  $\mu\text{g m}^{-3}$ .

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## Compliance with Ethical Standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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