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Airborne *Quercus* pollen in SW Spain: Identifying favourable conditions for atmospheric transport and potential source areas

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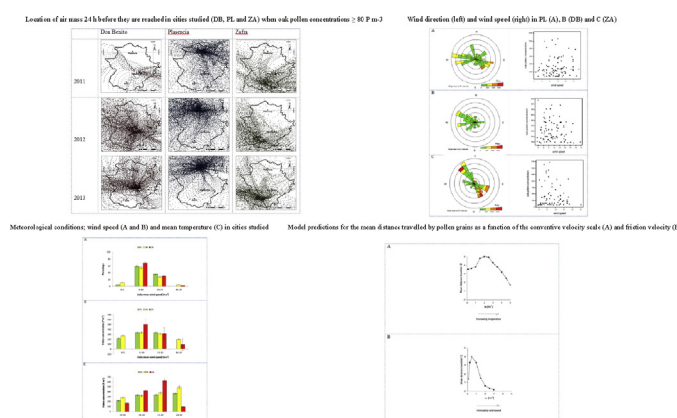
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HIGHLIGHTS

- Calculation of distance to which *Quercus* pollen can be transported in 24 h period
- Analysis of the air mass transport using the HYSPLIT model
- Analysis of *Quercus* pollen concentration measured at 3 sites for a 3-year period
- Production of *Quercus* tree inventory maps
- Comparison of experimental data with pollen dispersion model outputs

GRAPHICAL ABSTRACT



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ABSTRACT

The pollen grains of *Quercus* spp. (oak trees) are allergenic. This study investigates airborne *Quercus* pollen in SW Spain with the aim identifying favourable conditions for atmospheric transport and potential sources areas. Two types of *Quercus* distribution maps were produced. Airborne *Quercus* pollen concentrations were measured at three sites located in the Extremadura region (SW Spain) for 3 consecutive years. The seasonal occurrence of *Quercus* pollen in the air was investigated, as well as days with pollen concentrations $\geq 80 \text{ P m}^{-3}$. The distance that *Quercus* pollen can be transported in appreciable numbers was calculated using clusters of back trajectories representing the air mass movement above the source areas (oak woodlands), and by using a state-of-the-art dispersion model. The two main potential sources of *Quercus* airborne pollen captured in SW Spain are *Q. ilex* subsp. *ballota* and *Q. suber*. The minimum distances between aerobiological stations and *Quercus* woodlands have been estimated as: 40 km (Plasencia), 66 km (Don Benito), 62 km (Zafra) from the context of this study. Daily mean *Quercus* pollen concentration

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Back-trajectory analysis
Bioaerosol transport

can exceed $1,700 \text{ P m}^{-3}$, levels reached not less than 24 days in a single year. High *Quercus* pollen concentration were mostly associated with moderate wind speed events ($6\text{--}10 \text{ m s}^{-1}$), whereas that a high wind speed ($16\text{--}20 \text{ m s}^{-1}$) seems to be associated with low concentrations.

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1. Introduction

The “dehesas” (Spanish) or “montados” (Portuguese) are currently the most common forms of Mediterranean forests derived ecosystems in the SW of the Iberian Peninsula. They comprise pastures and tree species mainly belonging to the genus *Quercus* (oak). Five species are the most representative in the Extremadura region: *Quercus ilex* subsp. *ballota* (= *Q. rotundifolia*), *Q. suber*, *Q. pyrenaica*, *Q. coccifera* and *Q. faginea*. In the Iberian Peninsula, 12 native *Quercus* species have been described (Amaral Franco, 1990). Occasionally, other species are planted as ornamentals or they appear rarely as spontaneous, mainly as relictual, for instance *Q. robur*. Production of *Quercus* spp. (hereafter *Quercus*) pollen occurs during spring in Spain (García-Mozo et al., 2002). *Q. ilex* (holm oak) has a considerable ecological and economic importance (García-Mozo et al., 2007). *Q. suber* (cork oak) landscapes have a high level of biodiversity, being an important source of income derived from cork production and play a key role in ecological processes (Vessella and Schirone, 2013).

Quercus is an anemophilous genus producing high quantities of pollen grains (Tormo-Molina et al., 1996) that are suitable for atmospheric dispersion (Hernández-Ceballos et al., 2011a) and with different airborne pollen dynamics depending on the influence of geography and meteorological conditions (Hernández-Ceballos et al., 2015). Studies have examined the influence of weather conditions on the distribution of the seasonal pollen curve (Grewling et al., 2014; Jung and Choi, 2013; Kasprzyk et al., 2014) and pollen release and dispersal (Jato et al., 2015). *Quercus* pollen allergens are considered to be a cause of pollinosis in Europe (Egger et al., 2008; Jato et al., 2014; Jato et al., 2015) and India (Bist et al., 2005). Whilst in the USA, *Quercus* pollen has been related to asthma morbidity (Darrow et al., 2012). In Extremadura, sensitization to *Quercus* pollen has been established for 21% of allergic patients (Cosmes Martín et al., 2005).

The amount of airborne *Quercus* pollen recorded at a site is influenced by the local tree density, pollen production, the distance between the source and the pollen trap and atmospheric transport processes (Jato et al., 2015; Schueler and Schlünzen, 2006). Climatic limitations and soil properties partly explain the regional distribution of *Quercus* in southern Spain (Urbieta et al., 2011). Also in relation to amount of airborne pollen, Oteros et al. (2015) proposed a new methodology for modelling the dynamics of airborne particles, called the Concentric Ring Method (CRM), based on the emission surface around a point that is correlated with the total amount of airborne particles.

The temporal distribution of airborne *Quercus* pollen has been analysed throughout the day (Hernández-Ceballos et al., 2015), the year (Pérez-Badía et al., 2013), comparatively between two samplers in height (Charalampopoulos et al., 2013; Fernández-Rodríguez et al., 2014c) and distance (Fernández-Rodríguez et al., 2014b). It is usually not known whether airborne *Quercus* pollen grains are sampled from nearby or more remote sources. Back-trajectory analysis is a useful tool for studying the origin of airborne biological particles, and several papers have recently combined trajectory calculations with records of airborne pollen concentrations (Fernández-Rodríguez et al., 2014a; Hernández-Ceballos et al., 2011b, 2015). For these analyses, two models have been employed in aerobiological studies, e.g.: the hybrid single particle lagrangian trajectory model (HYSPLIT) (Fernández-Rodríguez et al., 2014a; Skjøth et al., 2012) and Weather Research and Forecasting-Chemistry (WRF-Chem) (Hernández-Ceballos et al., 2014; Skjøth et al., 2015).

This study investigates airborne *Quercus* pollen in SW Spain with the aim identifying favourable conditions for atmospheric transport and potential sources areas. This was achieved by a combination of statistical analysis of atmospheric pollen concentrations and meteorological data, atmospheric modelling using HYSPLIT and an atmospheric dispersion model, as well as spatial analysis of oak tree distribution using 2 different inventory maps performed in ArcGIS.

2. Material and methods

2.1. Sampling sites and oak tree resources

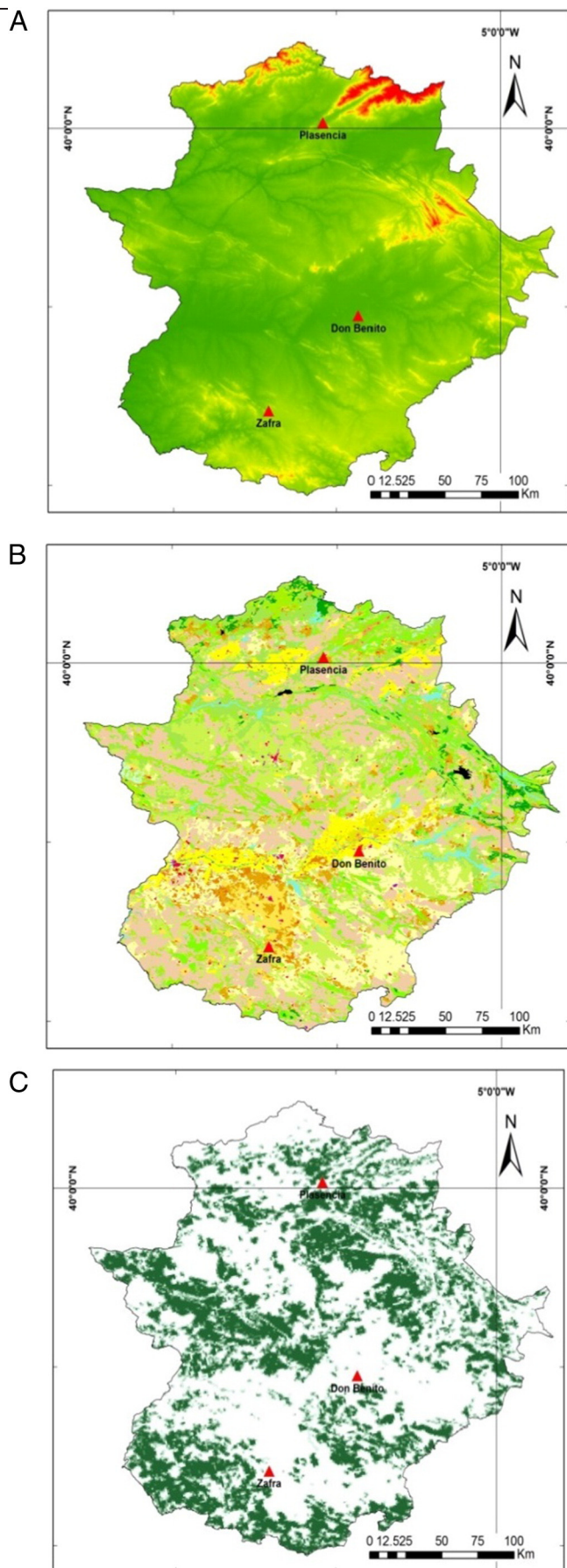
Extremadura is an agricultural region ($41,635 \text{ km}^2$) in South-West Spain, with a population of 151,565 (NSI, 2015). Atmospheric sampling was undertaken in three cities located latitudinally in the region, i.e., Plasencia (PL) in the north, Don Benito (DB) in the middle, and Zafra (Z) in the south (Fig. 1A–C). The maximum distance of 182 km separated PL and ZA. The respective heights above sea level are 415 m, 280 m and 508 m (Fig. 1A). Around PL there are holm oak and cork oak trees and extensive grazing. The most frequent land use around DB is irrigated crops (e.g. fruit trees, corn, tomato, rice). In the vicinity of ZA there is an influence of livestock production, olive trees and holm-oak trees (Fig. 1B).

An indication of major *Quercus* pollen sources was prepared in the present study using Corine Land Cover 2006 v. 17 dataset for the studied area (EC, 2013). Three classes were jointly presented (i.e., 244, 311 and 313), which corresponded to agro-forestry areas, broad-leaved forest and mixed forest, respectively. Distribution maps of 3 oak species (*Q. pyrenaica*, *Q. ilex* subsp. *ballota*, *Q. suber*) were based on raster data sets kindly made available by Dr Ángel M. Felicísimo (Felicísimo et al., 2004). These maps were constructed using predictive modelling techniques based on the statistical relations between the spatial patterns of presences/absences of the species and environmental variables such as climate, slope, elevation, etc. Guisan and Zimmermann (2000) and Mateo et al. (2011) provide a comprehensive review of these models and methodology. The inventory maps were gridded using the CLC 2006 projection by aggregating the grid cells to $5 \text{ km} \times 5 \text{ km}$ using similar procedures after Sadyś et al. (2014) and Skjøth et al. (2012).

2.2. Climate and meteorological data

The Extremadura region has a Mediterranean climate with a continental influence, annual mean temperatures range between 16°C and 17°C (PL 16.1°C , DB 17.1°C and ZA 15.8°C). The highest temperature is reached in the summer, mainly in July and August. Total annual rainfall varies depending on latitude (PL 694 mm, DB 460 mm and ZA 619 mm). Average data for the 1961–1990 period was provided by the Regional Meteorological Center (AEMET, 2016). Prevailing winds are from the West (Silva-Palacios et al., 2007) in a region that has a nearly flat topography.

Hourly meteorological data for PL, DB and ZA were provided by the AEMET (Agencia Estatal de Meteorología). Distance between meteorological stations and Hirst type 7-day recording volumetric traps (Hirst, 1952) was 2.24 km (PL), 0.95 km (DB) and 0.53 km (ZA).



2.3. *Quercus* pollen data

Daily mean (00:00–23:59 h) and hourly airborne *Quercus* pollen concentrations were observed from 2011 to 2013 using the Hirst type air sampler (Hirst, 1952). In the case of DB, only data of 2012 and 2013 have been taken into account. This is because the sampler was situated in the nearby town of Santa Amalia during the first six months of 2011, and there were significant gaps in the study period. The traps were situated on the terraces of: (1) Hospital Virgen del Puerto (43° 10' N, 2° 25' W) at 16 m (PL), (2) Donoso Cortés Secondary School (38° 58' N, 5° 50' W) at 6 m (DB), (3) and Hospital Zafra (38° 25' N, 6° 25' W) at 6 m (ZA). Standardised data management procedures were used, similar to those described by the Spanish Aerobiology Network (REA) (Galán et al., 2007). Slides were examined along two longitudinal transects, which equates to 7.7% of the slide examined. Daily average *Quercus* pollen concentrations and hourly concentrations were expressed as particles per cubic metre of air ($P\ m^{-3}$) following Comtois (1998).

The main pollen seasons (MPS) of *Quercus* were defined using the 90% method, whereby the season started when 5% of the total annual catch was achieved and ended when 95% was reached (Nilsson and Persson, 1981). The characteristics of *Quercus* pollen seasons were described by the start and end dates of the airborne pollen season, the duration (number of days), the date and value of the maximum daily mean *Quercus* pollen concentration, and the sum of daily average airborne *Quercus* pollen concentrations recorded in the season (Seasonal Pollen Index – SPI).

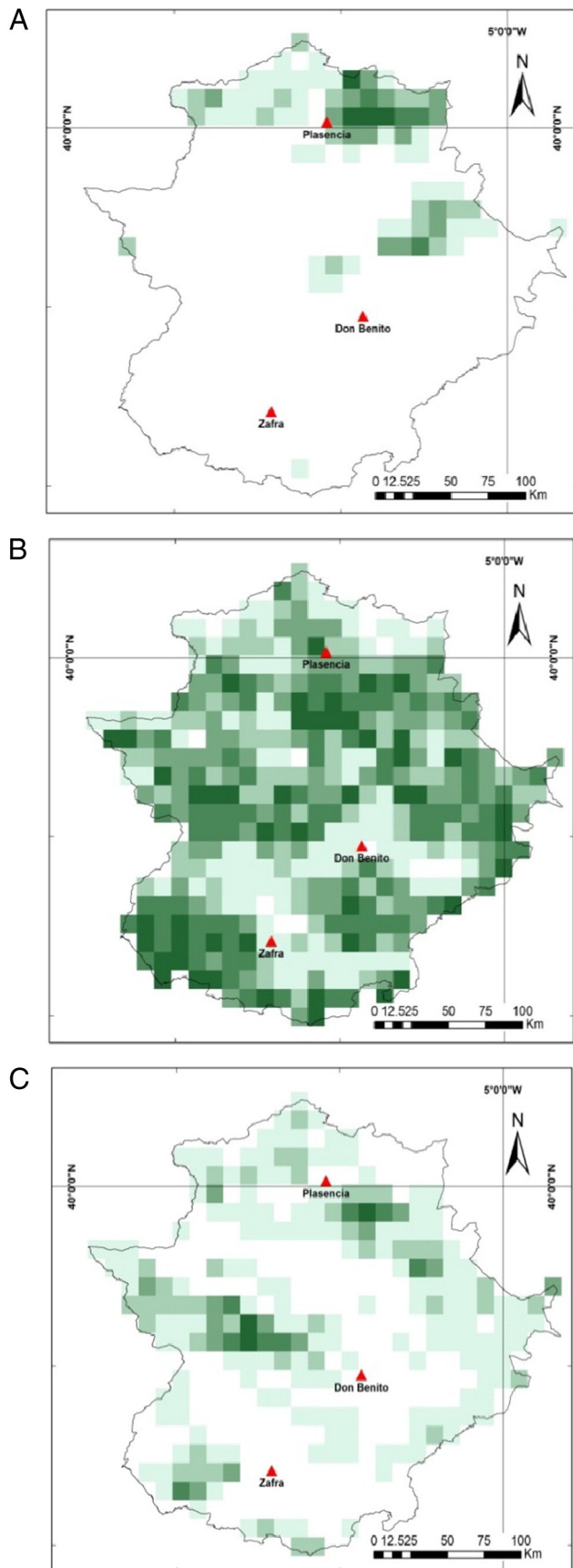
The mean diurnal variation of all days with a daily average airborne *Quercus* pollen concentrations $\geq 80\ P\ m^{-3}$ recorded in PL, DB and ZA were selected for detailed examination. The threshold of $\geq 80\ P\ m^{-3}$ was purely arbitrary and helped in removing days with low daily average values, whilst still allowing a large enough dataset for analysis. It is similar to the threshold levels estimated for other allergenic tree pollen types de Weger et al. (2013) and references therein.

2.4. Back-trajectory analysis

The air mass transport above PL, DB and ZA was examined using back-trajectories computed by the HYSPLIT model (Draxler and Hess, 1998; Draxler and Rolph, 2014; Luo et al., 2010; Rolph, 2014) for all days when *Quercus* pollen concentrations $\geq 80\ P\ m^{-3}$ were observed. The number of days selected was: PL $n = 122$, in DB $n = 83$, and in ZA $n = 86$. The weather data used for the trajectory calculation came from the Global Data Analysis System (GDAS) archives, which is maintained by the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL). The chosen methodology has certain limitations relating to uncertainty: (1) the quality of meteorological data (GDAS archives); (2) the intrinsic ambiguity that is linked to individual back-trajectories. To overcome this uncertainty “clusters” of back-trajectories were examined, i.e. a group of trajectories computed in a particular year and specific sampling site, merged together and analysed jointly. Such clusters of trajectories have previously been used, in combination with observations, in a number of other studies on bioaerosols (Fernández-Rodríguez et al., 2014a; Hernández-Ceballos et al., 2011b; Makra et al., 2010; Skjøth et al., 2009). This is a well-established procedure and thus the uncertainties related to individual trajectories will have a limited impact on the final results.

Air mass trajectories were plotted 24 h back in time, with 1 h interval, at 500 m above ground level (Fernández-Rodríguez et al., 2015). A further step required matching the location of the clustered back-

Fig. 1. Location of the aerobiological stations in the Extremadura region, SW Spain: (A) elevation above ground level based on the Digital Elevation Model (<http://srtm.csi.cgiar.org>); (B) land cover based on the CLC2006 (EC, 2013); (C) forest cover based on the CLC2006 (EC, 2013). Spatial resolution: 90 m (b), 100 m (c) and 1 km² (d).



trajectories with *Quercus* species distribution and overall wind pattern, characteristic for each study site. This examination was performed with an hourly resolution. Once the time frame was estimated, the mean velocity of the air masses was calculated using the tools available in ArcGIS (v. 10.2.1); thus, enabling a precise calculation of the distance travelled by the air masses carrying large quantities of airborne oak pollen grains that were observed in PL, DB, ZA (Sadyś et al., 2015). We also have taken into consideration the time of natural pollen release by the oak trees to verify their potential for the airborne transport during a 24-h time frame.

2.5. Pollen dispersion modelling

An in-house state-of-art pollen dispersion model was used to simulate the dispersal of *Quercus* pollen grains. By invoking the well-mixed condition Rotach et al. (1996) formulated a Lagrangian stochastic (LS) model for the simulation of passive body dispersal in atmospheric boundary layers with stabilities ranging from ideally-neutral to fully-convective. Model predictions are in good agreement with data collected in laboratory-scale experiments and in the field (Gryning and Lyck, 1984; Willis and Deardorff, 1976). A detailed description of this LS model, its numerical implementation and the accompanying parameterization of required meteorological inputs can be found in (Rotach et al., 1996). Here we adopt this model for the simulation of *Quercus* pollen grain dispersal in convective boundary-layers with wind shear. These boundary-layers are characterised by two velocity scales: the friction velocity u_* (square root of the surface stress divided by the density of the air) and the convective velocity scale $w_* = (-H \cdot Z)^{1/3}$, where H is the surface buoyancy flux and Z the boundary-layer height.

2.6. Statistical analysis

Initial data screening was performed using descriptive statistics and normality test (Shapiro-Wilk test). Spearman's rank test was used to examine the relationship between the occurrence of *Quercus* pollen in the air and meteorological parameters, except for the association between pollen concentration and wind direction which was investigated using circular statistics as described in (Sadyś et al., 2015). Hourly records were used for this purpose. Statistical testing was performed in SPSS (v.15.0), GenStat (v. 17.0) and Microsoft Excel (2010) software.

3. Results

3.1. Inventory of potential *Quercus* pollen sources

Forest cover based on the CLC2006 (EC, 2013) as an indication of major *Quercus* pollen sources is presented in Fig. 1C. According to the Spanish National Forest Inventory (MMA-DGB, 2007) three main species of *Quercus* dominate the current vegetation in Extremadura (Fig. 2). *Q. ilex* subsp. *ballota* in the lowlands and low mountainous areas (<500 m) with 30.4% of the surface (12,670 km²), *Q. suber* in mountainous areas with heights in the range 500–900 m, with 5.1% of the surface (2,127 km²), and *Q. pyrenaica* in the mountainous areas with the highest heights (900–1600 m), with 2.3% of the surface (945 km²) (Felicísimo et al., 2004). The other *Quercus* species, *Quercus ilex* subsp. *ilex*, *Q. petraea* and *Q. robur* are not frequently recorded and appear as complementary elements in the three main of current climatophilous vegetation apart from river landscapes (Fig. 2).

Fig. 2. Distribution of *Quercus* in the Extremadura region, SW Spain: (A) *Q. pyrenaica*; (B) *Q. ilex* subsp. *ballota*, (C) *Q. suber*. Applied spatial resolution was 1 km². The colour scale corresponds to the *Quercus* distribution expressed in percentage from lowest (1–20%, light green) to the highest (81–100%, very dark green). Distribution maps are not shown for *Quercus ilex* subsp. *ilex*, *Q. petraea* and *Q. robur*, because tree densities were very low. (For interpretation of the references to color in this figure legend, the reader is referred to the online version of this chapter.)

Table 1

Characteristics of the main pollen seasons (MPS) of airborne *Quercus* pollen in the Extremadura region, SW Spain (2011–2013). Plasencia (PL), Don Benito (DB), and Zafra (ZA).

City and year	MPS start date	MPS end date	Length (days)	Peak day	Peak daily average concentration ($P\ m^{-3}$)	SPI	Average daily value ($g\ m^{-3}$)	Days with airborne pollen concentration $\geq 80\ P\ m^{-3}$
PL 2011	01 April	18 May	48	18 April	808	8266	172	31
PL 2012	05 April	04 Jun	61	11 May	1058	19827	325	52
PL 2013	14 April	10 Jun	58	17 April	1181	13606	235	39
PL mean			56			13899	250	41
DB 2011 ^a	-----	-----	-----	-----		-----	-----	-----
DB 2012	01 April	10 Jun	71	17 May	587	13482	190	46
DB 2013	08 April	01 Jun	55	17 April	875	10043	183	28
DB mean			63			11762	187	37
ZA 2011	30 March	11 May	43	09 April	1639	15156	352	31
ZA 2012	10 April	08 Jun	60	11 May	1729	13939	232	32
ZA 2013	06 April	26 Jun	82	17 April	785	6417	78	23
ZA mean			62			11837	192	29

^a Pollen trap was moved from Santa Amalia to Don Benito and as a result there is not a continuous record for this site in 2011.

3.2. Temporal variations in airborne concentrations of *Quercus* pollen

The maximum concentrations were recorded mainly in the months of April, in 2011 and 2013, and in May in 2012 (Table 1). The mean SPI for each site were PL 13,899 grains, DB 11,762 grains and ZA 11,837 grains. The average daily values at each site ranged from 172 to 325 $P\ m^{-3}$ in PL, from 183 to 187 $P\ m^{-3}$ in DB, and from 78 to 352 $P\ m^{-3}$ in ZA, depending on the year analyzed, with means of 250, 187 and 192 $P\ m^{-3}$, respectively. The highest daily average airborne *Quercus* pollen concentrations were in PL on the 17th of April of 2013 (1,181 $P\ m^{-3}$), DB on the 17th of April of 2013 (875 $P\ m^{-3}$) and ZA on the 11st of May of 2012 (1,729 $P\ m^{-3}$) (Table 1). The length of the MPS ranged from 43 days (Zafra in 2011) to 82 days (Zafra in 2013).

The highest concentrations were recorded on two days; the 11th of May 2012 for PL and ZA, and on the 17th of April 2013 for the three cities studied. The results of Spearman's rank test for the MPS showed that (Table 2): (a) statistically significant positive correlations between airborne *Quercus* pollen concentrations and maximum temperature and mean temperature in all three stations, minimum temperature in DB, and wind direction in ZA; (b) statistically significant negative correlations were found between airborne *Quercus* pollen concentrations and rainfall in all three sites and relative humidity in PL. According to the

linear-circular association test 3 statistically significant positive correlations were obtained for wind direction. The results of Spearman's rank test for the days with concentrations $\geq 80\ P\ m^{-3}$ show statistically significant positive correlations between airborne *Quercus* pollen concentrations and maximum (PL and ZA), mean (DB and ZA) and minimum (DB and ZA) temperature, while negative correlations were found for relative humidity (ZA). The linear-circular association test indicated 2 statistically significant positive correlations between airborne *Quercus* pollen concentrations and wind direction (Table 3).

3.3. Analysis of the air mass transport, wind direction and wind speed

Fig. 3 shows the origin of the air mass 24 h before they reached the study sites (PL, DB, ZA) and the location of trajectory points. The time (expressed in percent) air masses spent inclusively over the territory of the Extremadura region varied and, on average, was equal to 30% (PL), 34% (DB) and 29% (ZA).

Table 4 shows the distance travelled by the air masses between sampling points and the closest large woodland areas, when airborne *Quercus* pollen concentrations $> 80\ P\ m^{-3}$ were recorded. The results show that the closest sources of airborne *Quercus* pollen are estimated as being 40 km (PL), 66 km (DB), and 62 km (ZA) for the specification of this study.

Table 2

Spearman's rank test and correlation coefficient values between airborne *Quercus* pollen concentrations and meteorological parameters recorded during the MPS (2011–2013). Statistically significant values are highlighted in bold; r – correlation coefficient, p – probability, n – number of observations.

		Plasencia	Don Benito	Zafra
T_{max}	r	0.381	0.283	0.263
	p	0.000	0.001	0.000
	n	167	126	185
T_{mean}	r	0.292	0.303	0.239
	p	0.000	0.001	0.001
	n	167	126	185
T_{min}	r	0.073	0.262	0.095
	p	0.347	0.003	0.197
	n	167	126	185
Rainfall	r	-0.264	-0.250	-0.198
	p	0.001	0.007	0.011
	n	162	116	164
RH	r	-0.451	-0.147	-0.122
	p	0.000	0.099	0.099
	n	167	126	185
Wind Speed	r	0.097	-0.003	-0.064
	p	0.214	0.973	0.385
	n	167	122	185
Wind direction ^a	r	0.023	0.065	0.174
	p	0.771	0.475	0.018
	n	167	122	185

^a Linear-circular association test was used.

Table 3

Spearman's rank test and correlation coefficient values between airborne *Quercus* pollen concentration and meteorological parameters, recorded on days with pollen concentration $\geq 80\ P\ m^{-3}$, between 2011 and 2013. Statistically significant values are highlighted in bold; r – correlation coefficient, p – probability, n – number of observations.

		Plasencia	Don Benito	Zafra
T_{max}	r	0.233	0.209	0.375
	p	0.010	0.074	0.000
	n	122	74	86
T_{mean}	r	0.178	0.245	0.382
	p	0.050	0.035	0.000
	n	122	74	86
T_{min}	r	0.046	0.233	0.275
	p	0.612	0.046	0.010
	n	122	74	86
Rainfall	r	-0.030	-0.169	-0.052
	p	0.743	0.172	0.676
	n	122	67	68
RH	r	-0.151	-0.104	-0.281
	p	0.096	0.378	0.009
	n	122	74	86
Wind speed	r	0.068	-0.084	0.032
	p	0.458	0.475	0.771
	n	122	74	86
Wind direction ^a	r	0.009	0.056	-0.044
	p	0.923	0.636	0.688
	n	122	74	85

^a Linear-circular association test was used.

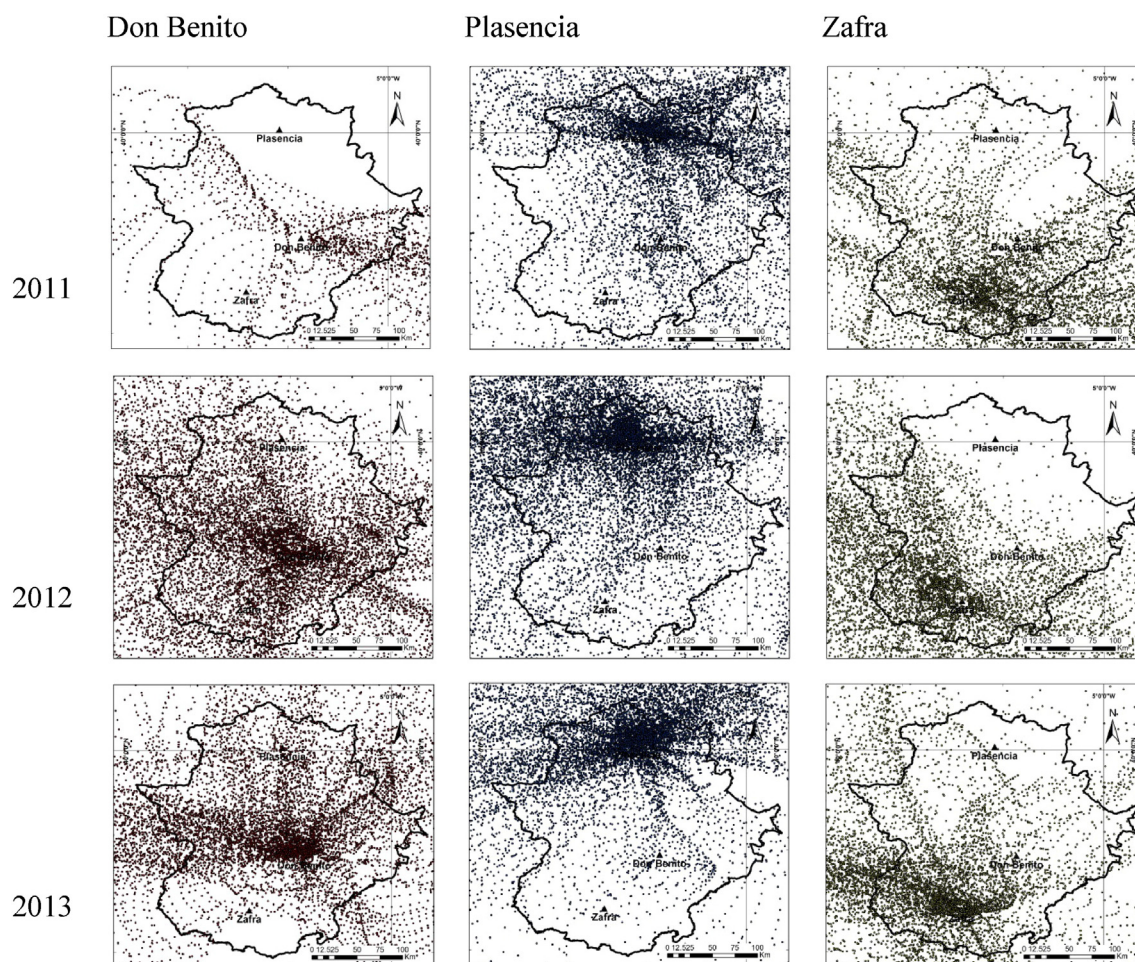


Fig. 3. Location of air mass 24 h before they reached Don Benito, Plasencia and Zafra cities, during the 3 year studied period, when oak pollen concentrations $\geq 80 \text{ P m}^{-3}$ were recorded.

Results of the descriptive circular statistics for the local wind direction measured at 3 aerobiological stations are given in Table S1. There was a statistically significant difference in the overall mean wind direction recorded at each station in 2011 and 2012. This has been confirmed by 2 out of 3 tests, which were used to examine the data uniformity around the circle ($p < 0.05$; probability test of randomness and probability Rayleigh test of uniformity). In 2011 a dominance of eastern winds was observed, while in 2012 western winds were prevalent when *Quercus* pollen concentrations $\geq 80 \text{ P m}^{-3}$ were detected (Fig. 4, Table S1).

Out of the 3 sites under investigation, the highest daily mean pollen concentrations were found in ZA ($> 1,500 \text{ P m}^{-3}$). The wind carrying large amounts of pollen originated from two opposite directions, i.e., NW and SSE. In the case of PL and DB, airborne *Quercus* pollen concentrations increased when wind was blowing from W-WNW and ESE-SSE arcs, respectively (Fig. 4). The same figure also presents an overview on the wind speed measured simultaneously. The impact of changes in wind speed on high atmospheric concentrations of *Quercus* pollen are shown in Fig. 5. Overall, the greatest quantities of pollen were observed

when the wind speed was within the range of $6\text{--}10 \text{ m s}^{-1}$ (Fig. 4) and higher wind speed ($> 14 \text{ m s}^{-1}$) seem to be associated with lower concentrations although the number of data points was limited. Similarly, the greatest quantities of airborne *Quercus* pollen were observed when the temperature was between 21 and 25°C (Fig. 4). Lower concentrations were observed when temperatures were $\leq 20^\circ\text{C}$ and $\geq 26^\circ\text{C}$. Figs. 5 and 6 show that pollen dispersal is most effective in terms of distance on warm, windy days and is less effective during purely convective (hot ambient temperatures and no breeze) or neutrally stable conditions (cool ambient temperatures and strong winds).

3.4. Pollen dispersion modelling

The Lagrangian stochastic model predicts that the mean distance travelled by pollen grains is a maximum on warm days with a light breeze (Fig. 6), i.e., under conditions that might occur in the spring in temperate latitudes. Model predictions for dispersal kernels (distributions of travel distances under a variety of atmospheric conditions) can be found in Reynolds (2011). Exponential and power-law contact distributions represent different atmospheric conditions.

4. Discussion

Quercus pollen is widely recorded and abundant among other airborne pollen types in the Iberian Peninsula, e.g. in Badajoz, Estepona, Évora, Huelva, Malaga, Salamanca (Caeiro et al., 2007; Fernández-Rodríguez et al., 2014b; Rodríguez-de la Cruz et al., 2010, 2008; Trigo et al., 2000). Trigo et al. (2000) evaluated the overall contribution of *Quercus* pollen in Estepona and Malaga as 14.98% and 10.87%,

Table 4

The distance travelled by the air masses between sampling points and the closest large woodland areas, with airborne concentrations of *Quercus* pollen $\geq 80 \text{ P m}^{-3}$ were recorded.

Distance	Plasencia			Don Benito			Zafra		
	2011	2012	2013	2011	2012	2013	2011	2012	2013
Min [km]	70.4	40.4	45.1	66.3	132	147.4	61.9	72.3	96.3
Max [km]	142.6	83.4	93.5	207.7	194.1	177.8	176.6	142.1	142.6

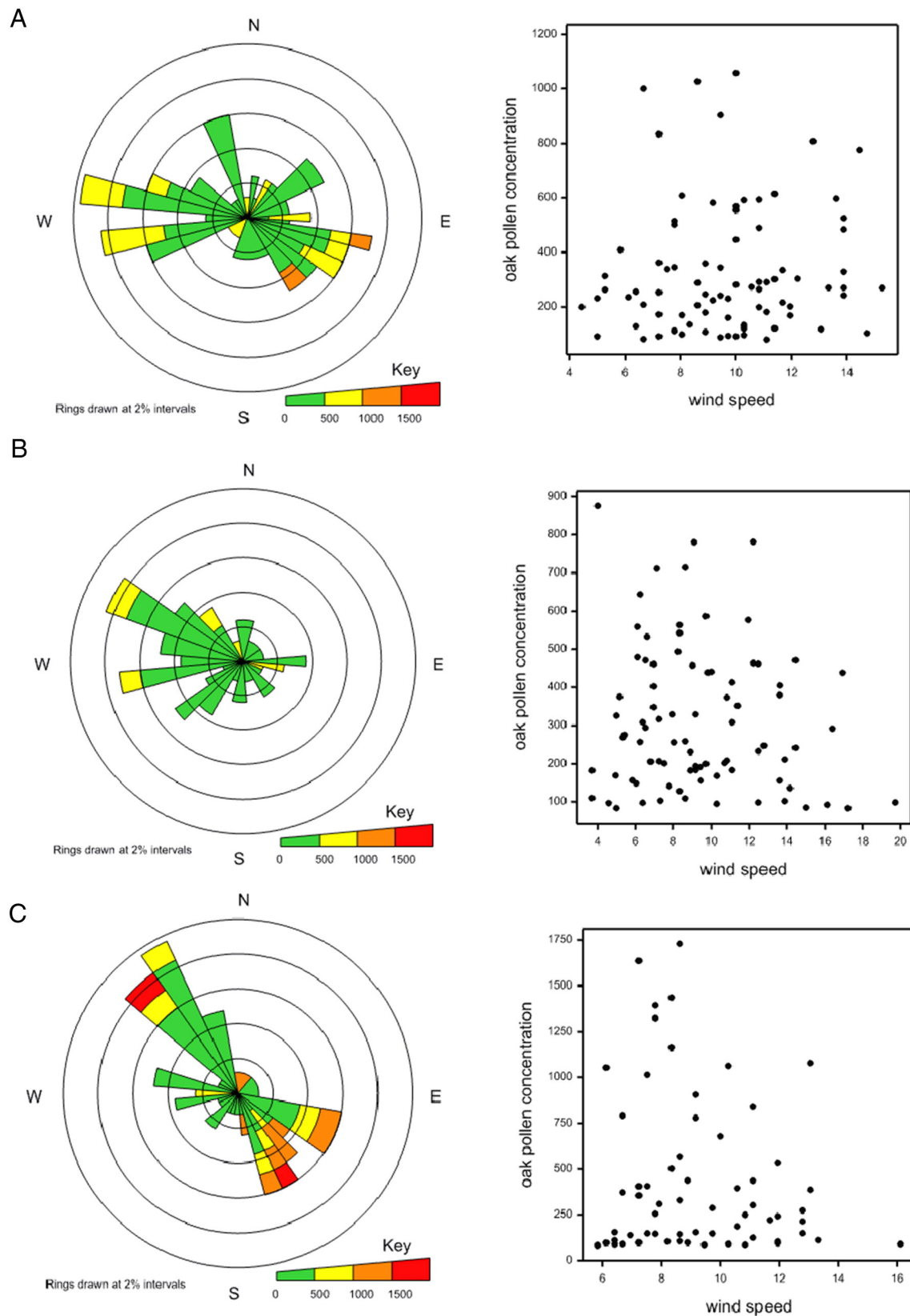


Fig. 4. A–C. Wind direction (left hand side) and wind speed (m s^{-1}) (right hand side) recorded during days with *Quercus* pollen concentrations $\geq 80 \text{ P m}^{-3}$ for 2011–2013 at: (A) Plasencia; (B) Don Benito; (C) Zafra. The applied interval was equal to 30° , while colours indicate airborne *Quercus* pollen concentrations from low (green) to high (red). Wind speed data for the year 2013 were not available for Plasencia and Zafra, thus a 2-year data were pooled together for these sites.

respectively. However, much greater values were found by Maya-Manzano (2015) for the 3 cities under investigation, i.e., 46.7% (PL), 31.4% (DB), and 40.1% (Zafra). Daily average values, similar to the

ones in this study (Table 1), were recorded by (Pérez-Badia et al., 2010) in Toledo (31 P m^{-3}). Lower values have been obtained at other sites such as Madrid (17 P m^{-3}) (Bustillo et al., 2006); Toledo

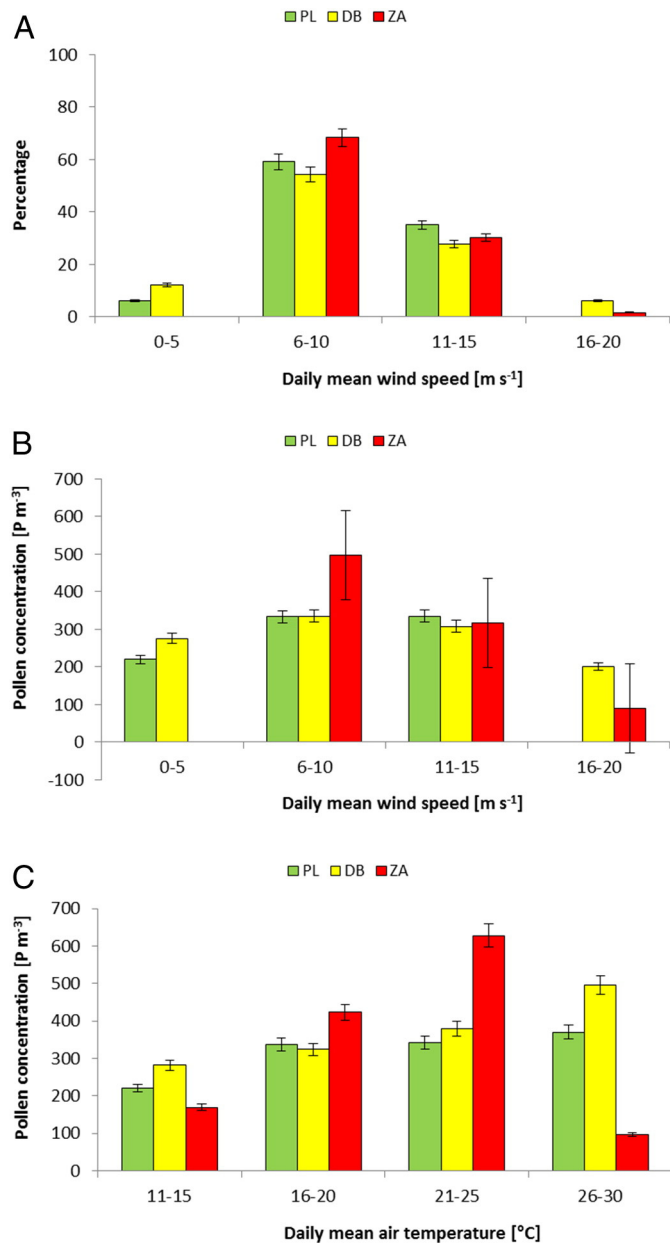


Fig. 5. The influence of meteorological conditions on atmospheric *Quercus* pollen concentrations at Plasencia (PL), Don Benito (DB) and Zafra (ZA) for 2011–2013: (A) Dominant wind speed (m s^{-1}) recorded during days with airborne *Quercus* pollen concentrations $\geq 80 \text{ P m}^{-3}$ and expressed as a percent. The error bars show 5% margin in data variation. Wind speed data for 2013 were not available for PL and ZA, and thus only 2-years data were used for these sites (see Fig. 4). (B) Daily average airborne *Quercus* pollen concentrations as a function of mean wind speed. (C) Daily average airborne *Quercus* pollen concentrations as a function of mean air temperature.

(20 P m^{-3}) (García-Mozo et al., 2006); and Nerja, Málaga (15 P m^{-3}) (Docampo et al., 2007). With regard to the maximum airborne concentrations of *Quercus* pollen recorded in Zafra in 2011 ($1,639 \text{ P m}^{-3}$) and 2012 ($1,729 \text{ P m}^{-3}$), observed values exceeded the highest concentrations registered in the Galicia region (NW Spain) throughout the 20-year period (Jato et al., 2015).

Such high concentrations of *Quercus* pollen could be explained by a very high rate of pollen production by *Quercus* trees and the occurrence of large woodland areas in the vicinity of the aerobiological stations (Fig. 1). A study conducted by Tormo et al. (1996) determined that *Quercus ilex* subsp. *ballota* produces one of the largest quantities of

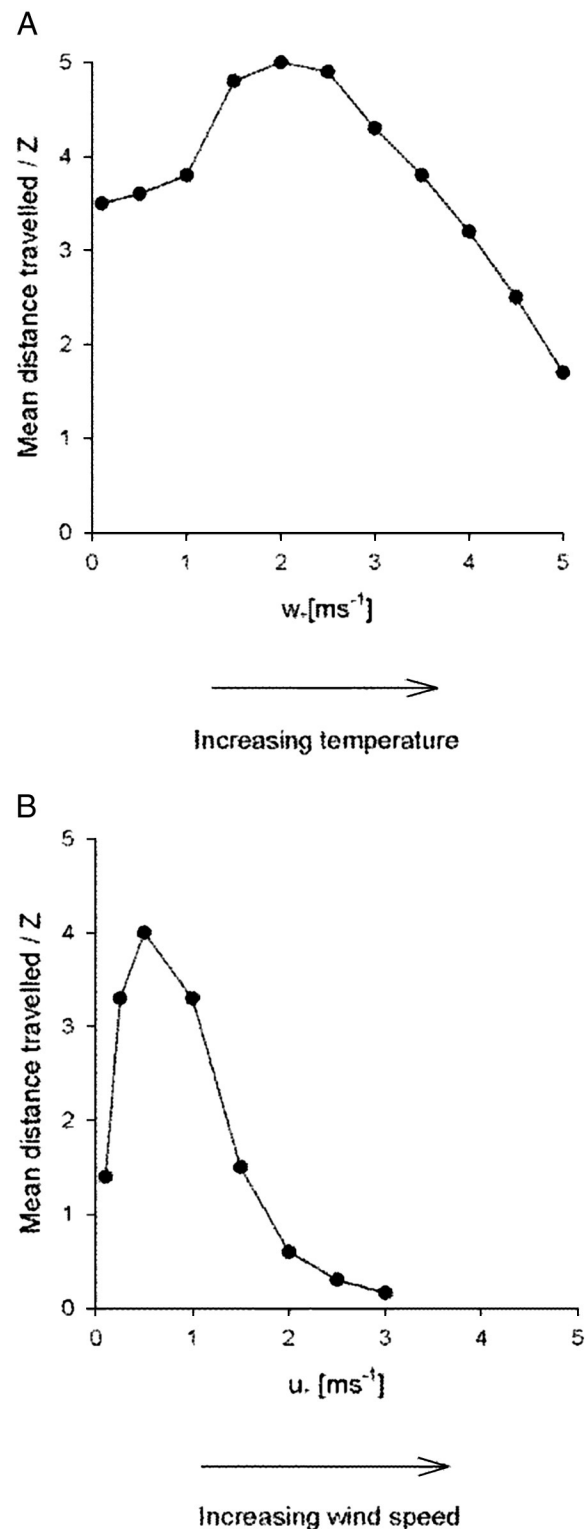


Fig. 6. Model predictions for the mean distance travelled by pollen grains after becoming airborne at a height $0.001Z$ (where Z is the height of the boundary layer) and before landing: (A) As a function of the convective velocity scale (w_*). Predictions are shown for a friction velocity scale $u_* = 0.5 \text{ m s}^{-1}$ which corresponds to mean wind speeds (averaged over the depth of the boundary-layer) ranging from 6.9 m s^{-1} ($w_* = 0 \text{ m s}^{-1}$) to 2.2 m s^{-1} ($w_* = 5 \text{ m s}^{-1}$). (B) As a function of the friction velocity (u_*). Predictions are shown for a convective velocity scale $w_* = 1 \text{ m s}^{-1}$. The predicted mean distance travelled is a maximum when $u_* \approx 0.5 \text{ m s}^{-1}$ which equates to a mean wind speed of 5.5 m s^{-1} .

pollen within a group of 10 anemophilous tree taxa, reaching 550.90×10^8 pollen grains per meter of tree crown diameter. It is important to note that the inflorescences of *Quercus* grow at the edge of the tree crown enabling pollen to be released and dispersed freely in contrast to other broadleaved tree taxa (Tormo et al., 1996). Gómez-Casero et al. (2004) used a different method to calculate *Quercus* pollen production per single tree when examining 120 trees from 4 species. The authors found the *Quercus suber* ($240.46\text{--}550.45 \times 10^8$) to have the highest productivity and *Q. coccifera* ($41.99\text{--}56.01 \times 10^8$) the lowest. Other investigated species, i.e., *Q. ilex* ssp. *ballota* and *Q. faginea*, produced a maximum of 244.96×10^8 and 227.31×10^8 pollen grains per single tree, respectively.

Species such as *Quercus ilex* ssp. *ballota* and *Q. faginea* flower in Spain between mid-February and the end of March. In contrast, *Q. suber* and *Q. coccifera* flower later, i.e., from the beginning of April until mid-May and from an early May to the end of May, respectively (Boavida et al., 2001). The flowering period usually lasts between 11 and 15 days, and *Quercus* pollen can be present in the atmosphere for up to 20–30 days longer (Jato et al., 2007).

Galan et al. (1991) showed that *Quercus* pollen concentrations recorded in Córdoba peaked in the afternoon and in the evening (16:00–21:00 h) and minimum concentrations were witnessed at night and early in the morning (02:00–08:00 h). In this study, hourly concentrations of airborne *Quercus* pollen peaked between 13:00 and 15:00 h at PL, but later in both DB and ZA (15:00–17:00 h). The minimum concentrations were recorded at night, with the lowest concentrations from 04:00 to 07:00 h. These are similar to the results presented by Fernández-Rodríguez et al. (2014b) in Badajoz, with peaks from 12:00 to 16:00 h.

After heat requirements for the onset of flowering have been met (García-Mozo et al., 2002), meteorological parameters directly influence the release of pollen grains from the anthers that dehisce while drying out in order to enable their dispersal throughout the air (Frenguelli and Bricchi, 1998). It was found that:

Sunny and warm weather generally corresponds with the highest pollen production (Hyde, 1950a, b).

Relative humidity plays an important role, since high humidity contributes to hydration of pollen grains, resulting in an increased likelihood of pollen grain aggregation and an increase in their weight.

The occurrence of rainfall causes a “washing off” effect in the atmosphere, resulting in a decrease in pollen concentrations (Hall, 1992). However, it should be noted that a convective rain shower can eliminate the pollen plume on its way to a site, which will still report perfect weather conditions.

Orlanski (1975) proposed definitions for scales of atmospheric processes, i.e., meso-gamma (2–20 km), meso-beta (20–200 km) and meso-alfa (200–2,000 km). Wind direction and wind speed can affect the pollen distribution in many different ways, emphasizing that high wind speed can move the pollen to meso-beta or even meso-alfa scale, also could result in discharge of large amounts of pollen. However, high wind speed can aid the release of large quantities of pollen, re-suspend pollen that has fallen to the ground, or dilute the amount of pollen in the atmosphere (O'Rourke, 1990). In our study, we found that the wind speed within the range of $6\text{--}10 \text{ m s}^{-1}$ increased airborne *Quercus* pollen concentrations measured at all 3 sites (Fig. 4). Furthermore, a high wind speed ($16\text{--}20 \text{ m s}^{-1}$) seems to be associated with low concentrations (Fig. 5), which could be due to the factors cited previously. Moreover, high wind speeds can also decrease the efficiency of the volumetric trap (Hirst, 1952).

The Extremadura region is characterized by relatively low variance in topography (Fig. 1), and so air masses are not disturbed in their flow over long distances. These observations are in agreement with the linear-circular association test, which only detected occasionally statistically significant relationships between high atmospheric *Quercus* pollen concentrations and local wind speed (Table 2). Moreover, Fig. 4A shows that *Quercus* pollen concentration recorded in PL and ZA

originated from two opposite directions (W-SE and NW-SE), while in DB from one direction (W). *Quercus* pollen was transported from a source located to the west, regardless of the overall dominance of eastern winds recorded in 2011 (Figs. 4 & S1). This information is important in terms of the *Quercus* pollen forecast produced for these cities, but more years of study are still necessary for improving the accuracy of this information.

This study shows that increasing the surface heat flux, promotes thermals that in turn promote the successful lift-off of pollen grains from the plant canopy (woodlands). However, when the situation is anti-cyclonic and geostrophic wind all but disappears, the free convection can form and most pollen grains will tend to rise and fall within a column of air above the launch site, rather than disperse in a downwind direction. Similarly, increasing the wind speed promotes dispersal, but too much speed suppresses the creation of thermals (updrafts) that keep the pollen suspended. The minimum distances between aerobiological stations and *Quercus* woodlands have been estimated as: 40 km (Plasencia), 66 km (Don Benito), 62 km (Zafra) from the context of this study. Jato et al. (2002) reported a distance of 10–30 km using a combination of airborne pollen data and phenological observations made on *Quercus robur* and *Q. pyrenaica* in Ourense (NW Spain). A longer distance of potential *Quercus* pollen transport, equal to 40 km, was suggested by Hernández-Ceballos et al. (2015). The authors examined atmospheric *Quercus* pollen concentrations recorded using air samplers located in 2 sites in southern Spain and modelled air mass movement during the pollination period with the aid of the HYSPLIT model (Hernández-Ceballos et al., 2015). Finally, Dyakowska (1959) calculated theoretical dispersal distance of *Quercus robur* in still air as 65 km, taking into account the average grain size equal to $27.7 \mu\text{m}$ and 3.96 cm s^{-1} settling velocity. Later on as discussed by the author, applied formula was underestimating the actual dispersal range because variables, such as wind speed and wind direction during the pollination period and the height above ground level at which pollen was released, had not been included in the calculations. Nevertheless, estimated value of 65 km was found in agreement with results derived from this study.

5. Conclusions

- Daily mean *Quercus* pollen concentration can exceed $1,700 \text{ P m}^{-3}$ in the area and period studied.
- The number of days with high atmospheric concentrations of *Quercus* pollen varied between years and sites, but was not less than 24 days in a single year.
- High airborne *Quercus* pollen concentrations in PL, DB and ZA are mostly associated with moderate wind speed events, when wind velocity is within the span of $6\text{--}10 \text{ m s}^{-1}$, whereas that a high wind speed ($16\text{--}20 \text{ m s}^{-1}$) seems to be associated with low concentrations
- In PL and ZA *Quercus* pollen is transported from two independent sources, while in DB airborne *Quercus* pollen originates from a single source.
- Based upon the species distribution maps, it can be concluded that the greatest contributions towards the total amounts of *Quercus* pollen recorded in PL, DB and ZA were made by *Quercus ilex* subsp. *ballota* and *Q. suber*. The potential contribution of *Q. pyrenaica* was found to be of less importance in PL.
- The minimum distances between aerobiological stations and *Quercus* woodlands have been estimated as: 40 km (PL), 66 km (DB), 62 km (ZA) from the context of this study.

Conflict of interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2016.07.094>.

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Supplementary materials to the article

Airborne *Quercus* pollen in SW Spain: identifying favourable conditions for atmospheric transport and potential source areas

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Table S1 Results of descriptive circular statistics for local wind direction, when *Quercus* pollen concentration was recorded in days with $\geq 80 \mu\text{m}^{-3}$.

Site		Year	
		2011	2012
Don Benito	Number of observations	9	46
	Mean direction	119.37°	275.15°
	Circular standard deviation	42.22°	55.44°
	Mean resultant length	0.762	0.626
	Skewness	-2.298	0.087
	Kappa estimate	1.717	1.628
	Prob. test of randomness	0.011	0.000
	Prob. Rayleigh test of uniformity	0.003	0.000
	Chi-square von Mises with 9 df	10.060	8.710
	Prob. Chi-square von Mises	0.346	0.465
Plasencia	Number of observations	31	52
	Mean direction	92.38°	289.29°
	Circular standard deviation	63.05°	87.87°
	Mean resultant length	0.546	0.309
	Skewness	0.473	-0.258
	Kappa estimate	1.311	0.649
	Prob. test of randomness	0.001	0.000
	Prob. Rayleigh test of uniformity	0.000	0.007
	Chi-square von Mises with 9 df	10.560	41.160
	Prob. Chi-square von Mises	0.307	0.000
Zafra	Number of observations	31	32
	Mean direction	151.00°	288.51°
	Circular standard deviation	78.57°	66.36°
	Mean resultant length	0.391	0.511
	Skewness	-0.238	1.145
	Kappa estimate	0.849	1.195
	Prob. test of randomness	0.000	0.000
	Prob. Rayleigh test of uniformity	0.008	0.000
	Chi-square von Mises with 9 df	42.640	39.960
	Prob. Chi-square von Mises	0.000	0.000

* df - degrees of freedom

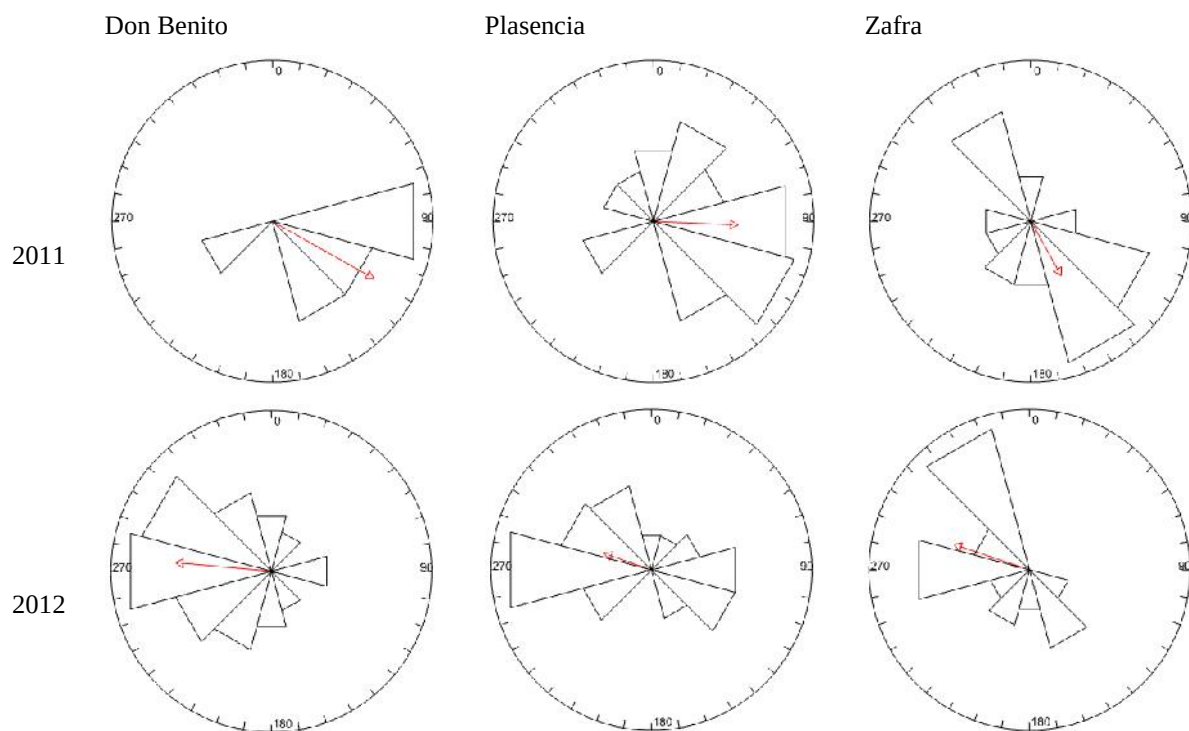


Fig. S1 Local wind direction recorded during days with concentration $\geq 80 \mu\text{g m}^{-3}$. The applied interval was equal to 30° , while arrows indicate mean wind direction. Data for the year 2013 were not available.