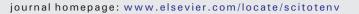
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The effects of continentality, marine nature and the recirculation of air masses on pollen concentration: *Olea* in a Mediterranean coastal enclave



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HIGHLIGHTS

GRAPHICAL ABSTRACT

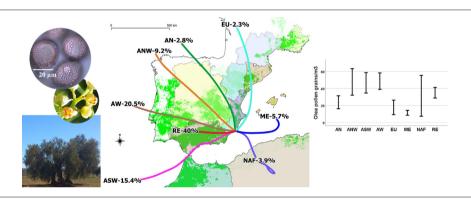
- Back trajectories explained variations in events with high *Olea* pollen concentrations.
- The highest Olea pollen concentrations were produced with regional air masses.
- Long-distance transport caused *Olea* pollen peaks in a coastal, peninsular location.
- Marine effects during advections reduced *Olea* pollen concentrations.

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ABSTRACT

Olea pollen concentrations have been studied in relation to the typology of air masses, pollen grain sources and marine nature during advections in a coastal enclave in the south-eastern Iberian Peninsula. Since Spain is the world's leading olive producer, and olive growing extends throughout the Mediterranean basin, this location is ideal for the study of long-distance transport events (LTD) during the main pollen season (MPS). The air masses were classified using the calculation of 48-h back trajectories at 250, 500 and 750 m above ground level using the HYSPLIT model. After that, the frequency of LDT events from Africa and Europe was found to be 8.7% of the MPS days. In contrast, regional air masses were found in 38.6% of the MPS days. This was reflected in pollen concentrations, with significantly higher concentrations (p-value <0.05) on days with regional air masses compared to days with European air masses. Regarding the source areas, the importance of nearby sources with intense olive cultivation was confirmed (i.e., Andalusia). This proximity was relevant beyond the attenuations observed when the advections acquired a marine nature as the air mass back trajectories moved over the sea (p-value <0.001). The review of air mass typologies, source areas and pollen concentrations resulted in establishing peak dates and the detection of LDT associated with these peak dates. Distortions in the typical path of each air mass back trajectories varied the pollen load that every type of air mass could originally contain.

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Abbreviations: AN, North Atlantic; ANW, Northwest Atlantic; AW, West Atlantic; NAF, North African; ME, Mediterranean; EU, European; RE, Regional; ASW, Southwest Atlantic. * Corresponding author.

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1. Introduction

Olive cultivation in southern Europe and North Africa is profuse given the benefits of the Mediterranean climate to the physiology of the species (Ramos-Román et al., 2019). According to Rodríguez Pleguezuelo et al. (2018), 5,000,000 ha are allocated to agriculture in the Mediterranean basin alone, with Spain being the world's leading olive producer. Therefore, it constitutes a flagship species in its agroecosystem (Delcourt et al., 2019). Despite the fact that consumption of its oil and fruit is beneficial (Cicerale et al., 2010), the crop has an undesired impact due to the proven allergenic potential of its pollen (Moreno-Grau et al., 2016). For this reason, it is a relevant species in Mediterranean pollen calendars (Galera et al., 2018), in which pollen grains can be autochthonous or allochthonous, given their ability to travel thousands of kilometres from the flowering trees (Aguilera et al., 2015). As the olive tree is mainly an anemophillic species, the particularities of the air masses in which pollen grains are transported determine not only their viability but also the path they take before landing on a surface, ideally, the gynoecium. Low relative humidity causes dehydration and the anthers to release pollen grains (Kozlowski and Pallardy, 2002). Long-distance transport (LDT) is dependent on the greater levels of humidity and lower temperatures of the upper layers of the atmosphere, and lower vapour pressure with respect to the humidity of the grains favours their viability (Viner and Arritt, 2010). In recent years, different papers have used tools to model air mass back trajectories (Sommer et al., 2015) and backward dispersion, including particle cross section calculations (De Weger et al., 2016; Monroy-Colín et al., 2020) for aerobiological monitoring. It has been possible to trace the routes of the allochthonous pollen grains of different taxa, such as Ambrosia, Artemisia, Amaranthaceae, Betula, Juniperus, Olea or Quercus (Negral et al., 2017). In many cases, the air mass back trajectories have moved over continental territory (Van de Water et al., 2003). In others, these air mass back trajectories with allochthonous pollens have moved, at least partially, over bodies of seawater (Makra et al., 2016) and, in the extreme case of small islands, almost completely over seawater (Romero et al., 2003).

A peninsula is a magnificent geographical configuration for measuring the influence of continental and marine air masses on pollen loads. This occurs when a coastal area of the peninsula is surrounded by pollen sources in the peninsular territory and the neighbouring territories outside the peninsula. In this case, the variations in the pollen concentrations of a taxon will reflect the influence of continentality or a marine nature on the pollen sampler, in addition to the logical phenological variations resulting from latitudinal and altitudinal disparity (Aguilera and Ruiz Valenzuela, 2009). The pollen grains of the olive tree are a good subject for this study, taking into account their cultivated, feral or ornamental specimens located throughout the Iberian Peninsula (IP) (Vargas and Talavera, 2012). Since it is a species that is cultivated throughout the Mediterranean basin, the study of the influence of marine air masses on the pollen concentration in a sample located on the Spanish Mediterranean coast is possible. Gustafsson (1998) used the sticky properties of sea salt particles in his work with the Cour pollen trap. Therefore, the incorporation of marine constituents on pollen grains on the Portuguese coast was expected (Duque et al., 2013). These authors detected Cl^- , Na^+ and Mg^{2+} , which are tracer species of marine salinity (Negral et al., 2008), coating the surface of the pollen. Since relative humidity increased the concentration of these ions, it was possible to establish the relationship between pollen grains, ions, water uptake by pollen grains and relative humidity. This may be important in the phenology of species such as the olive tree, in which an alteration of flowering and other reproductive functions have been verified under conditions of stress caused by NaCl (Koubouris et al., 2015).

The sedimentation of pollen grains is also conditioned by the synoptic charts (i.e., meteorological maps describing the state of the atmosphere over a large area at a given moment, American Meteorological Society, 2021) that determine wind flows. Monsoons are capable of depositing large amounts of pine pollen grains over large areas of the northern waters of the South China Sea (Sun and Li, 1998). Airborne pollen can also settle close to its source. The lack of strong synoptic charts leads to the aging of the air masses with recirculation (Prtenjak et al., 2012). The corresponding air mass back trajectories form loops when approaching their destination, which facilitates the resuspension of particulate matter (PM) in the absence of physical obstacles or ground cover (Negral et al., 2012). Along these lines, pollen grains have been identified as the main source of organic matter in settling dust from arid areas of Australia (Boon et al., 1998). Topographic features join the variability of synoptic charts, making the circulation and, consequently, the transport of bioaerosols more complex. Viner et al. (2017) reported how topography determined pollen deposition in a complex island environment. Indeed, topography and prevailing synoptic charts are relevant in the dispersal of a species. In a study of olive pollen grains in Sierra Nevada (mountain range in southern Spain) it was possible to verify that the positive North Atlantic Oscillation phase, associated with arid and warm conditions in the western Mediterranean basin, coincided with regressive periods of olive cultivation (Ramos-Román et al., 2019).

In the literature review by Fröhlich-Nowoisky et al. (2016), the authors stated the need to improve the quantification of bioaerosol emissions and their interactions with atmospheric transport. Although bacterial counts have been addressed as a function of air masses (Romano et al., 2019; Uetake et al., 2019), we have not found studies focussed on the effect of the typology of air masses carrying pollen grains, i.e., its continental or marine nature. In order to determine this influence and the effect of air mass recirculation, this paper carries out a 22-year retrospective analysis of pollen concentrations and the cataloguing of air masses in a peninsular coastal enclave potentially receptive to autochthonous and allochthonous grains from the same taxon: *Olea*.

2. Materials and methods

2.1. Study area

The city of Cartagena (Spain) is located in the southeast of the IP, on the Mediterranean coast (37° 37′ 1" N; 0° 58′ 59" W). It belongs to the Region of Murcia and is enclosed between the parallels 37° 20' 20" and 38° 45' 15" N and meridians 0° 42' 2" and 2° 20' 0" W. The city is the capital of a district called the Campo de Cartagena, a coastal plain, with a slight incline towards the southeast which is surrounded by mountainous elevations except in the coastal area, where a jagged, steep coast is located to the south and southwest. To the east, the Menor Sea, 180 km², is separated from the Mediterranean Sea by La Manga spit (Romero Díaz and Belmonte Serrato, 2011). The climate consists of high temperatures and little rainfall, producing the marked characteristics of aridity: weak average thermal oscillation; long, hot summers with severe droughts; short, mild winters; long autumns with mild temperatures; and short springs. It is located in the driest area in Europe, from which the expansion of sub-desert steppes begins (Andrés Sarasa, 1986). According to the Köppen-Geiger climate classification, it is classified as BSh (dry, steppe, warm), with an average temperature of 18.2 °C (average temperature in January, 11.0 °C, August, 26.0 °C, with an annual range of 15.0 °C). The mean accumulated annual rainfall is 296 mm. The bioclimatic floors Infra-Mediterranean and Thermo-Mediterranean are present across its municipal area (Rivas-Martínez et al., 2017). Vegetation is highly humanimpacted in the Campo de Cartagena. Herbaceous crops occupy 23,759 ha (22,213 irrigated and 546 rainfed) and 14,680 ha correspond to woody crops (9548 irrigated and 5132 rainfed), with olive groves occupying an area of 463 ha (77 irrigated and 424 rainfed) (Ministry of Ecological Transition and Demographic Challenge, 2019).

2.2. Sampling and Olea pollen concentrations

Aerobiological samples were collected using a Hirst-type trap, with an aspiration volume of 10 L/min (Lanzoni VPPS 2000), located on the roof of the Cartagena railway station, 10 m from the ground. The procedure for sampling, handling and the identification and quantification of the samples adhered to that described in the standard EN 16868 (2019). In summary, the trap has a rotating drum that moves at 2 mm/h, on which a transparent Melinex tape is placed. The tape is covered with adhesive silicone. The air is sucked in from the bottom and enters through the front, always facing downwind as it is equipped with a weather vane. This configuration forces the air to change direction so that the particles from a certain equivalent aerodynamic diameter cannot adapt to the change in the direction of the gas flow. They collide on the surface of the head and remain adhered. The sampling, performed continuously, lasts for seven days. At the end of this period, the head is replaced with another blank one. The removed head is transferred to the laboratory, where the tape is cut into daily sections, mounted on slides with glycerogelatin (colourless at the base with fuchsin at the top), and protected with a coverslip. After a settling period, the tape is analysed under an optical microscope (Olympus BH2, equipped with plan-achromatic optics). Four transverse transects are read with an immersion 50× optical zoom lens; the total surface analysed must be >10%. Pollen and fungal types are identified, and the corresponding count is made. A conversion algorithm is used to calculate daily concentrations, expressed as pollen grains/m³. The main pollen season (MPS) was calculated for every year (1993-2014), eliminating the beginning and end of the flowering days that account for 2.5% of Olea pollen every year (MPS, 95% of the annual pollen integral, API) (Andersen, 1991).

2.3. Air mass origin and Olea pollen sources

The air masses that reached the study area were catalogued following the proposal for the IP by Font Tullot (2000). This classification consists of seven types, grouped according to the thermal nature and the marine or continental origin of the mass. In accordance with previous studies (Negral et al., 2012, 2020), a new type was included to define the slowing down of advections upon arrival, causing recirculation over the peninsular territory and providing the mass with a regional nature. Therefore, the types of masses were: Marine Arctic, coded with the acronym AN since it reaches the IP from the North Atlantic; Marine Polar, coded as ANW, reaching the IP from the Northwest Atlantic; Marine Subtropical, coded as AW, which reaches the IP from the West Atlantic; Marine Tropical, coded as ASW since it reaches the IP from the Southwest Atlantic; Continental Tropical, coded as NAF since it reaches the IP from North Africa; Continental Tropical, with considerable permanence over the Mediterranean, coded as ME; Continental Polar, coded as EU, which reaches the IP after travelling over the European continent; and, finally, when the mass remains over the IP with limited movement and acquiring a regional nature, it was coded as RE (Negral et al., 2008). Every day of the study was coded according to one of these eight types of air masses. The code was assigned in accordance with the air mass back trajectories. The procedure to calculate these air mass back trajectories was as follows: Throughout the 22 years, 1202 days were counted in the MPS. Discounting the days with problems in the trap, the result was 1188. Of these, 1167 days of the MPS in Cartagena were finally taken, corresponding to those on which it was possible to calculate the 48-h isentropic air mass back trajectories reaching Cartagena. The 48-h period was chosen to ensure a strict criterion establishing confidence in the LDT of airborne pollen grains and to reduce errors due to longer-lasting air mass back trajectories. The graphic representation was obtained for three heights: 250, 500 and 750 m above ground level (m agl) between the days of the MPS 03/ 21/1993 and 12/31/2014. In the event that the air mass back trajectories at different heights were not coincident at their origin, the air mass back trajectory at the lowest height and its behaviour when approaching the destination was the reference. The selection of the height of the air mass back trajectories for pollen monitoring varies in the scientific literature between 10 and 6000 m (Negral et al., 2017). Since Olea pollen grains are about 20–30 µm in diameter (Laaribi et al., 2017), lower heights were chosen than those for monitoring PM10 transport in the IP (Negral et al., 2012), but higher than the 10 m used for monitoring Olea grains at a short distance in the IP (Hernandez-Ceballos et al., 2014). Eventually, other heights (1500 and 2500 m agl) and longer periods (up to 120 h) were introduced into the model to obtain the air mass back trajectories. This was done to explore some interesting situations involving Olea pollen concentrations and the behaviour shown by the air mass back trajectories. The air mass back trajectories for Cartagena were calculated using the HYSPLIT model of the National Oceanic and Atmospheric Administration (Stein et al., 2015). This model allows users to choose the vertical motion method among vertical velocity, isobaric and isentropic models. We opted for the latter, as in a comparison among different methods, isentropic models showed lower deviation statistics (Harris et al., 2005). According to the length of the study period, different meteorological databases were used to feed the model. Detailed information on every database regarding, among other things, the domain, the mesh or the periodicity of the meteorological information can be consulted at the ECMWF website (https://www.ecmwf.int/en/forecasts/datasets/reanalysisdatasets/era-interim) and the Nacional Oceanic and Atmospheric Administration website (https://www.ready.noaa.gov/archives. php). Specifically, for every period, the following were used: 1st ECMWF Era Interim (European Centre for Medium-Range Weather Forecasts): 03/21/1993-12/31/1996; global domain; 80 km spatial resolution; 60 levels in the vertical, with the top level at 0.1 hPa; four analyses per day at 00, 06, 12 and 18 UTC. 2nd NCEP FNL (National Centers for Environmental Prediction, Final Operational Global Analysis): 01/01/1997-31/12/2004; global domain; 1 degree latitude-longitude (360 by 181); the upper level FNL data are output on the following mandatory pressure surfaces: 1000, 925, 850, 700, 500, 400, 300, 250, 200, 150, 100, 50, and 20 hPa; 4 times a day, i.e., at 00, 06, 12, and 18 UTC. 3rd NCEP GDAS (National Centers for Environmental Prediction, Global Data Assimilation System): 01/ 01/2005-31/12/2014; global domain; 1 degree latitude-longitude (360 by 181); 23 levels in the vertical, with the top level at 20 hPa; 4 times a day, i.e., at 00, 06, 12, and 18 UTC.

In addition to cataloguing air masses, the potential geographic sources of Olea pollen grains were taken into account on their path to the trap. Therefore, 14 potential geographic sources were defined where these pollen grains could be released before reaching Cartagena (Fig. 1): Murcia, understood throughout the document as the homonymous region (code 1); Andalusia (2); Plateau, including the Spanish regions Castile-La Mancha, Castile and Leon, Extremadura and Madrid (3); Aragon (4); Catalonia (5); Balearic Islands (6); Valencia, understood throughout the document as the Valencian region (7); Algeria (8); Morocco (9); Occitania (10); Alpes Cote (11); Corsica (12); Sardinia (13); and Tunisia (14). These source areas were assumed to be large enough to maintain the presence of Olea during the 22-year study period. The definition of the source areas was not intended to cover changes in land uses. In the event that air mass back trajectories were located over the sea, they were considered to have contributed a differentiating effect to that of the original source area of Olea pollen grains, in which case the new source area code was denoted by placing a "2" in front of each of the 14 source area codes. Given the location of Cartagena with respect to some potential source areas, the air mass back trajectories of these areas always typified the air mass with a marine nature. This was the case for the potential geographic sources of the Balearic Islands (26), Algeria (28), Morocco (29), Corsica (212), Sardinia (213) and Tunisia (214). Finally, if the air mass back trajectory moved over the Mediterranean before reaching Cartagena, the source area was coded as Mediterranean (215). When searching for the presence of LDT, the steps to elucidate whether pollen grains may be compatible

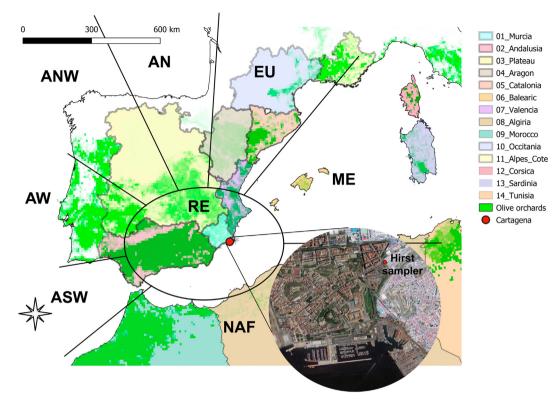


Fig. 1. Olive orchard distribution by source areas for *Olea* grains reaching Cartagena and air mass origin: North Atlantic (AN), Northwest Atlantic (ANW), West Atlantic (AW), North African (NAF), Mediterranean (ME), European (EU), Regional (RE) and Southwest Atlantic (ASW). Own elaboration using IPUMS Terra (Ruggles et al., 2018) and Google Maps [®].

with a pattern of LDT were: 1) the definition of air masses compatible with LDT (i.e., NAF and EU); 2) the delimitation of surrounding source areas (i.e., source areas in the IP) and far source areas (i.e., Mediterranean islands, source areas in France or countries in Africa); 3) a revision of the source areas passed over by the air mass back trajectories (i.e., far source areas); and 4) a revision of concentrations in the context of the days before and after (i.e., justification of saw-tooth shaped concentrations in the timeline).

2.4. Database analysis

The data obtained in Section 2.1. were incorporated into the aerobiological database, completed with the rainfall data provided by the State Meteorological Agency (AEMET). Information on the origin of air masses and sources of Olea pollen was incorporated into this database, as indicated in Section 2.2. The database, initially in Excel, was transformed into spv, in the licensed software package SPSS v.26 (IBM Corp. 2019). For this study, the database covering 1993-2014 was used. The normality of the distribution of Olea pollen concentrations was studied using the Kolmogorov-Smirnov test. Given that large sample sizes do not imply the rejection of ANOVA due to non-compliance with normality (Schmider et al., 2010; Blanca et al., 2017), the 1-factor ANOVA was applied to define the statistical differences between the subsets formed by types of air masses and source areas. Subsequently, the Tamhane test was used to reveal the differences, in twos, between subsets. The homogeneity of the variances had previously been contrasted using the Levene test. The error bar graph for the mean (95% confidence interval) was obtained to provide visual information.

3. Results and discussion

3.1. Olea pollen concentrations and air mass origin

The distribution of *Olea* pollen grains in the MPS had a *p*-value <0.001 for the Kolmogorov-Smirnov normality test, rejecting the null

hypothesis of normal distribution. Transformation into a normal distribution was not possible despite using logarithms for the pollen concentration. After ruling out homocedasticity with the Levene test (p-value <0.001), we opted for the assumption of no homogeneous variances in the 1-factor ANOVA and for the Tamhane test for subsequent differences between different types of air masses.

The frequency of MPS days according to air mass types is presented in Table 1. Further detail about the context of the aerobiological and climatic situation of the area can be consulted elsewhere (Galera et al., 2018). The highest probability of finding *Olea* pollen grains during the MPS occurred when the air masses were RE (38.6%), therefore, with geographically limited transportation. The greatest pollen grain deposition in the vicinity of the sources was expected (Matthias and Giesecke, 2014; Sommer et al., 2015). In contrast, the lowest frequency of air masses with *Olea* pollen concentrations was for AN (4.0%); which since it is a long-haul marine Arctic advection, would arrive from the north, with long residence times over areas with low olive density

Table 1	
Olea pollen grains in the MPS according to air mass back trajectories (1993–2014).	

Origin	Number of days (percentage ^a)	Pollen grains on peak date, grains/m ³ (peak date: mm/dd/yyyy)	Daily mean; total pollen grains, grains/m ³ (percentage ^b)
AN	47 (4.0%)	113 (05/22/1999)	23.8; 1117 (2.8%)
ANW	76 (6.5%)	334 (05/06/1997)	47.7; 3625 (9.2%)
AW	166 (14.2%)	427 (05/21/2006)	48.6; 8075 (20.5%)
ASW	130 (11.1%)	390 (05/26/2010)	46.6; 6062 (15.4%)
NAF	49 (4.2%)	561 (05/23/2009)	31.3; 1533 (3.9%)
ME	197 (16.9%)	206 (05/12/2013)	11.3; 2232 (5.7%)
RE	450 (38.6%)	648 (05/08/2011)	35.0; 15,734 (40.0%)
EU	52 (4.5%)	155 (05/23/1999)	17.7; 918 (2.3%)

North Atlantic (AN), Northwest Atlantic (ANW), West Atlantic (AW), Southwest Atlantic (ASW), North African (NAF), Mediterranean (ME), Regional (RE) and European (EU).

^a Percentage of total number of days in the MPS between 1993 and 2014.

^b Percentage of total pollen grains in the MPS between 1993 and 2014.

(Moreno-Grau et al., 2016). The air mass back trajectories with the highest concentrations corresponded to ANW, AW, ASW and RE. They presented significant differences (p-value <0.001) from the lowest concentrations of ME air mass back trajectories. Important differences were also shown (p-value <0.05) between concentrations of those Atlantic air mass back trajectories and those of EU air mass back trajectories (Fig. 2, Table S1 of the Supplementary Material). ANW, AW, ASW and RE air masses pass over olive cultivation areas (Rojo and Pérez-Badia, 2015) before they arrive to Cartagena. Therefore, greater pollen concentrations would have been expected. Even though the air masses could have experienced powerful Atlantic advection, this did not prevent them from remaining enough time over the olive groves of Andalusia and the Iberian plateau and loading up with pollen grains on their way to their destination (Rojo et al., 2016; Fernández-Rodríguez et al., 2020). Conversely, ME air masses, the second most frequently recorded (16.9%), did not have the opportunity to load up with pollen grains on their way. The great variability in the concurrent concentrations with NAF air mass back trajectories is worth noting, although they were infrequent (4.1%).

Those days with NAF and EU air mass back trajectories are interesting as potential indicators of the presence of allochthonous pollen grains due to intense advections. These air masses may transport these pollen grains from distant enclaves; that is, from Africa and other European countries with regards to the position of Cartagena. As these air masses only accounted for 8.7% of the MPS days, potential LDT events were not very probable. In line with this, García-Mozo et al. (2017) noticed the limited impact that the African air masses causing dust outbreaks had on *Olea* pollen concentrations in Andalusia, despite coinciding with the flowering periods of the species. However, for autochthonous pollen grains, these were days when air mass back trajectories were RE.

Having identified the days of interesting events, such as the peaks/ maxima for every type of air mass, they should be placed into the context of *Olea* pollen concentrations on the days before and after said peaks. For instance, in a sequence of days with air masses of the same type and with similar meteorological conditions, any fluctuation in the levels could reflect a deviation from the expected path for the air mass type. As the day of maximum pollen concentration for every type of air mass should be an exponent of that air mass, the days in the database with maximum concentrations for the eight types of air masses are analysed below (Table 1).

3.1.1. North Atlantic (AN)

On 05/22/1999 (113 pollen grains/m³) the maximum pollen concentration occurred with AN air masses. Three days earlier, 05/19/1999, the previous date with AN air masses, the pollen concentrations had been less than half (53 pollen grains/m³). Between both dates, the air masses were ANW, 05/20/1999 (315 pollen grains/m³) and 05/21/1999 (203 pollen grains/m³), with the annual peak being the first of

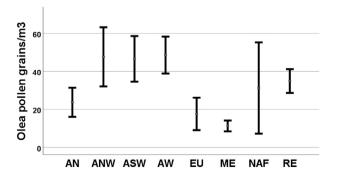


Fig. 2. Error bar graph for the mean (95% confidence interval, CI) *Olea* pollen grains/m³ in Cartagena according to the air mass back trajectories (1993–2014): North Atlantic (AN), Northwest Atlantic (ANW), Southwest Atlantic (ASW), West Atlantic (AW), European (EU), Mediterranean (ME), North African (NAF) and Regional (RE).

these two. As can be seen in Fig. S1 (Supplementary Material), the advections from ANW had longer residence times over peninsular olive areas than the advections originating from AN. In addition, in the north-western IP, olive trees have been an enhanced crop in recent years, which is having an influence on local aerobiological concentrations (Garrido et al., 2020). The dextrorotatory turning of the origin of the air masses from the annual peak to the maximum in AN on 05/22/1999 can be explained as a consequence of a downward transition in terms of *Olea* pollen loads, producing a transfer from more heavily loaded air masses to others less heavily loaded with these pollen grains.

3.1.2. Northwest Atlantic (ANW)

The day of maximum concentration with ANW air masses occurred on 05/06/1997, coinciding with the annual peak date. From 05/04/1997 to 05/08/1997, the sequence of air masses was: RE (36 pollen grains/m³), AW (236 pollen grains/m³), ANW (334 pollen grains/m³, peak date), ANW (125 pollen grains/m³) and AN (33 pollen grains $/m^3$). The evolution of the air masses (Fig. S2) and pollen concentrations would suggest that the concentrations on the peak date were caused by non-regional contributions, corresponding to the crops on the plateau of the IP. Thus, two days before the peak, the air masses had limited movement, remaining over the Mediterranean despite crossing the olive-growing areas near the trap. On 06/05/1997, the air masses demonstrated more powerful advection from the Atlantic, crossing the IP from west to east. This had an impact on a subsequent load of Olea pollen grains throughout the southern strip of the IP. However, this load was greater a day later, when the masses turned to ANW. This would imply that the greatest Olea contributions on the peak date came from the southern plateau, although slightly to the north of the Andalusian crops. The day after the peak date, the air masses remained ANW, although they stayed for less time in the south of the Iberian plateau, culminating the next day with AN air masses when the pollen concentrations fell by an order of magnitude with regard to the peak date. In short, the ANW advection could potentially be the cause of huge Olea contributions to Cartagena due to recharging, not so much in the olive groves of Andalusia but in those to the north (Fernández-Rodríguez et al., 2020).

3.1.3. West Atlantic (AW)

The maximum Olea pollen concentration with AW masses was detected in 2006, in an ascending sequence of pollen concentrations beginning 05/18/2006 (13 pollen grains/m³) with NAF air masses. Olea pollen concentrations increased by an order of magnitude the following two days with RE masses (112 and 187 pollen grains/ m^3 , respectively), reaching the relative maximum with AW masses on 05/21/2006 (427 pollen grains/ m^3), and decreasing the two subsequent days 05/22/2006 and 05/23/2006, with ASW masses up to two orders of magnitude (309 and 2 pollen grains/m³, respectively). From Fig. 3 we can intuit that even though the two-day air mass back trajectories did not move over the Atlantic on the day of the relative maximum, they came from the ocean, which was verified by extending the temporal calculation of the air mass back trajectories up to 5 days (Fig. S3). It can also be seen that the great recharge of the maximum on 05/21/2006 occurred between 6:00 am-6:00 pm on 05/20/2006, when the air mass back trajectories moved very close to ground level, incorporating pollen grains from the olive growing areas of eastern Andalusia. In conclusion, unlike ANW air masses, the increases in Olea pollen concentrations in Cartagena could be explained by recharges in the olive groves of Andalusia with AW air masses (Gabaldón-Leal et al., 2017).

3.1.4. North African (NAF)

The maximum *Olea* pollen concentration with NAF air masses occurred on 05/23/2009. From 05/20/2009 to 05/24/2009, the pollen concentrations were 50, 43, 37, 561 and 60 pollen grains/m³, respectively, when air mass back trajectories were NAF (Fig. S4). This boost of one

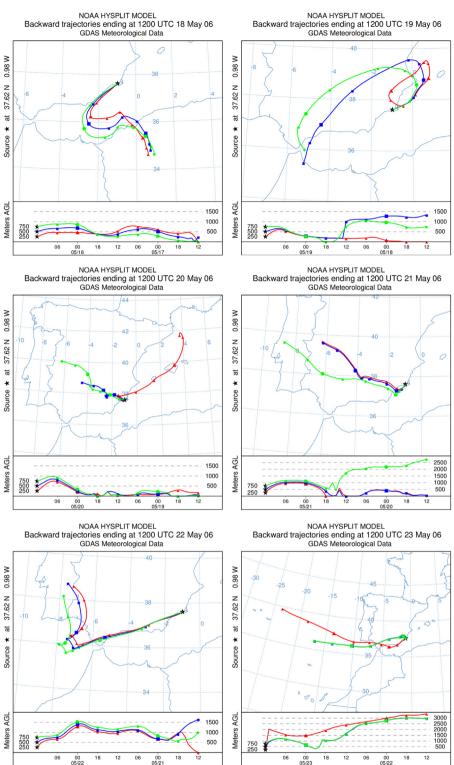


Fig. 3. Air mass back trajectories on 05/21/2006, with the maximum *Olea* pollen concentration with West Atlantic (AW) air masses; the previous days 05/18/2006 (North African, NAF) and 05/20–21/2006 (Regional, RE); and the following days 05/22–23/2006 (ASW).

order of magnitude on 05/23/2009 cannot be explained by the origin of the air mass since during all those days they had the same NAF classification. On 05/20/2009, 05/21/2009 and 05/23/2009, the air masses left the African continent through north-western Algeria, where, in the city of Oran, the *Olea* pollen concentrations represented the third most frequent pollen type (Kiared et al., 2017). In contrast, on 05/22/2009 and 05/24/2009, the air masses left the African continent from northern

Morocco, a country where *Olea* cultivation covers the entire territory except the Atlantic coastal strip. In the Moroccan city of Tétouan, the year 2009 registered the second highest sum value of daily pollen concentrations from the beginning of the MPS to the peak date; the period covered 2008–2014 (Achmakh et al., 2020). A possible contribution from African sources cannot be ruled out in the Cartagena concentrations. However, 21 mm of rainfall was collected on the day with 561 pollen

grains/m³. The air mass back trajectories indicated that the air masses remained for more than 24 h, recirculating over eastern Andalusia and Murcia, where they loaded up with Olea pollen grains. To elaborate on this hypothesis, the days 05/18/2009 and 05/19/2009 were reviewed. They showed concentrations of 565 and 242 pollen grains/m³, respectively. These two days the air mass back trajectories corresponded to RE masses, with the annual peak occurring during the first day, which, in turn, was a relative maximum of those days with RE air mass back trajectories (Fig. S5). On 05/18/2009, the air mass back trajectories had formed loops, surpassing eastern Andalusia. These trajectories were calculated at 120 h (Fig. S6), with the air masses slowing down and remaining for a longer period of time over areas of intensive olive cultivation in Andalusia. Therefore, the maximum Olea pollen concentrations with NAF air masses were attributed to a recharging of pollen grains when passing through olive cultivation areas of the IP rather than being caused by loading on the African continent.

3.1.5. Mediterranean (ME)

The maximum Olea pollen concentrations with ME air masses occurred on 05/12/2013 (206 pollen grains/m³), which also coincided with the annual peak. Comparing the levels of this peak date with the other peak dates in the database (1993-2014), the peak in 2013 corresponded to the 40.9th percentile. In principle, an even lower percentile could be expected for marine air masses since they lack any source of Olea pollen grains. In Palma, on the western Mediterranean island of Mallorca, olive cultivation is important. Olea was the second most abundant pollen type between 2004 and 2010 (Boi and Llorens, 2013). The API remained between 1450 and 5760 pollen grains/m³, compared to 675 to 3691 pollen grains/m³ for Cartagena during the same period. Including the two days before and after the peak date, the sequence of air masses was (Fig. S7): RE 05/10/2013 (86 pollen grains/m³), RE 05/11/2013 (184 pollen grains/m³), ME 05/12/2013 (206 pollen grains/m³), RE 05/13/2013 (20 pollen grains/m³) and RE 14/05/2013 (21 pollen grains/m³). Since the ME air mass is between days whose masses were RE, two possible hypotheses could justify the fact that the peak appeared precisely with ME masses instead of RE. First, in line with the levels before and after the maximum, the regional contributions are not very high, so the peak could be due to an even more geographically restricted contribution of origin than the regional one; that is, local. This hypothesis is based on the load of Olea pollen grains present during the limited continental route until they reach the trap. This cannot be ruled out since, as Bilińska et al. (2019) observe, large variations in pollen concentrations can occur between stations 4 km apart. Second, the peak could be due to a fall out of Olea pollen grains on the peak date. This fall out could be caused by notable levels of pollen grains previously injected into the upper layers of the atmosphere which settle on the peak date in the trap. This assessment is consistent with the air mass back trajectories calculated for higher altitudes (750, 1500 and 2500 m agl) and for 120-h intervals (Fig. S7).

3.1.6. Regional (RE)

On 05/08/2011, the maximum *Olea* pollen concentrations with RE air masses occurred. This day was also the peak date of the year and the global maximum of the entire study period (648 pollen grains/m³). The context of the peak date was RE masses for four days, reaching the maximum value on the third day. The pollen concentrations were 45 pollen grains/m³ on 05/06/2011, 51 pollen grains/m³ on 05/07/2011, 648 pollen grains/m³ on 05/08/2011 and 90 pollen grains/m³ on 05/09/2011. In this sequence, the jump in the order of magnitude of pollen concentrations from the second to the third day is striking as the air masses were of the same type. As can be seen in Fig. 4, the air mass back trajectories during these four days had the characteristics of the RE air mass type: loops, breaks, recirculation, or slowdowns when reaching their destination. The aging and stagnation of air masses in the IP is documented in the literature comparing air quality loss due to

LDT and biogenic emissions from forests (Gómez et al., 2020). Not only are meso-scale circulations able to justify the formation of photo-oxidant pollutants, but they also explain the injection of pollutants to the mid-troposphere in association with mountain ranges (Millán et al., 1996). This recurrent mechanism is caused by the development of the Iberian Thermal Low on sunny days, which are common during hotter seasons (Pay et al., 2019). This heat concludes with daytime convention acting as a reservoir of pollutants; subsequently, sea breezes may drive the latter inland (Gangoiti et al., 2002, 2006). However, only on the third day, the air masses remained over continental territory for practically 48 h, charging precisely in eastern Andalusia. In terms of the coastal location of Cartagena, it is possible to distinguish between the eminently continental RE masses, which caused the global maximum, versus the RE marine air masses. The latter could be responsible for the decline in pollen concentrations. In a study of bacterial bioaerosol in Tokyo, local sources were always the most prominent, although a variation and low bacterial contribution was also documented when the air masses came from the Pacific (Uetake et al., 2019).

3.1.7. European (EU)

The maximum pollen concentration on days with EU air mass back trajectories occurred on 05/23/1999 (155 pollen grains/m³). The sequence of air masses from three days before, with the peak date on 05/20/1999 (315 pollen grains/m³), until 05/27/1999, when the levels dropped to 1 pollen grain/m³ (ANW, ANW, AN, EU, RE, ME, ME, ME from May 20th to 27th, 1999) Figs. S1 and S8, could explain the gradual fall in pollen concentrations in the middle of which the maximum was detected: there was an eastward change in the origin of the air masses. In other words, the trap recorded the evolving concentration of a great amount of pollen grains from the plateau and Murcia towards a low concentration due to the washing effect of Mediterranean marine air masses. Similarly, in a study carried out in Italy on the effects of the transport of air masses on bioaerosol, the richness and biodiversity of bacterial communities decreased more with marine air masses than with continental ones (Romano et al., 2019). In addition, these authors verified that several hours with the air masses over the continent were enough to modify the nature of the bioaerosol.

3.1.8. Southwest Atlantic (ASW)

In 2010, the peak date occurred on 05/26/2010, coinciding with the highest pollen concentration with ASW masses. These air masses flowed through southern Andalusia to reach Cartagena (390 pollen grains/m³). The previous day the masses had been ME, with a clear marine effect on *Olea* pollen concentrations (10 pollen grains/m³). The day after the peak date, 05/27/2010 (217 pollen grains/m³), the air masses also travelled through Andalusia, but changed to RE. The decrease of more than 100 pollen grains/m³ was repeated on 05/28/ 2010 (114 pollen grains/m³), also with RE air masses. When reviewing the back trajectories (Fig. S9), we observe that the air masses arrived from the sea, as on 05/27/2010, or remained over the surface of the sea, as on 05/28/2010. Both of these situations would have toned down pollen concentrations. The effect of marine air masses in contrast to inland air masses has already been observed on the northern coast of Spain on Pinus pollen concentrations: when the air masses arrived from the sea, pollen concentrations dropped. However, if the masses came from the interior, they drove pollen grains from reforested areas (Jato et al., 2000). Meanwhile, on 05/28/2010, when the air masses entered the continent, the latitudes and enclaves they moved over were significantly different from those of the peak date: the air mass back trajectory at 750 m agl travelled through northern Andalusia while the air mass back trajectories at 250 and 500 m agl came from northern Murcia. In summary, ASW air masses were responsible for high levels of Olea pollen concentrations as a consequence of their dragging pollen grains from Andalusia (Galán et al., 2005). Once again, the route of the masses through Andalusia, a prominent olive growing area, may not faithfully

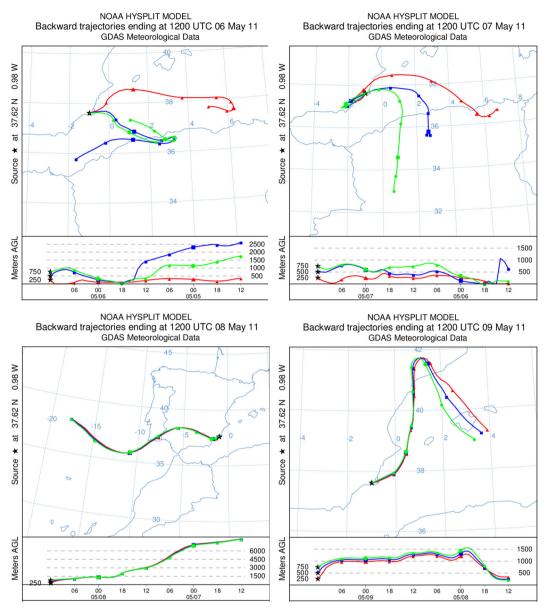


Fig. 4. Air mass back trajectories on 05/08/2011, with the maximum Olea pollen concentration with Regional (RE) air masses, with the previous days (05/06–07/2009) and the following day (05/09/2011), whose air mass back trajectories were also RE.

transfer the pollen load if the masses remain over the surface of the sea before reaching the trap.

3.2. Potential Olea pollen grain source areas and the marine effect on pollen concentrations

When revising the days in the MPS, 19 categories with *Olea* pollen grains appeared, disaggregated by source area and their possible marine nature (Table 2). The most frequent codes, around 10% of the days or more, corresponded to areas close to Cartagena: Andalusia, Marine Andalusia, Plateau and Mediterranean. As expected, this points to the greater probability of pollen grains being incorporated in areas close to the receptor than their coming from LDT (Bourgeois et al., 2001). This is in line with the air mass typology, with the greatest frequency of RE air masses during the MPS, which has already been discussed. The infrequent events, below 2% of the days, corresponded to codes from remote origins: Marine Alpes Cote, Corsica, Tunisia, Marine Occitania and Sardinia, all of which are marine in nature. Therefore,

under the prevailing conditions of atmospheric dynamics during the *Olea* MPS in Cartagena, the emitting areas at the greatest distance from the trap were the most unlikely.

When *Olea* pollen concentrations are reviewed together with their source areas, the importance of proximity to the receptor is again noted. Thus, the highest concentrations according to the types of source areas occurred for the most frequent and nearby source areas, also adding the area corresponding to Murcia. The impact of the pollen coming from Andalusia and Plateau on Cartagena, whether the carrier masses moved over the sea or not, was indisputable. Thus, there were significant differences (*p*-value <0.05) between the highest concentrations on days catalogued in these three source areas and the other ten source areas (Fig. 5, Table S2). Among those with a significantly lower contribution are some corresponding to LDT events: the Balearic Islands, Morocco, Marine Occitania, Corsica and Sardinia. Considering the concentration gradient in the deposition of pollen grains from the source (Raynor et al., 1974), if the LDT phenomena occurred, it was quantitatively much less impacting than the contributions from source areas

Table 2

Olea pollen grains in the MPS according to source areas (1993–2014).
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Source	Number of days	Pollen grains on peak date, grains/m ³	Daily mean; total pollen grains, grains/m ³			
	(percentage ^a)	(peak date:	(percentage ^b)			
		mm/dd/yyyy)				
Murcia	57 (4.9%)	648 (05/08/2011)	48.9; 2787 (7.1%)			
Andalusia	248 (21.3%)	390 (05/26/2010)	43.0; 10,672 (27.2%)			
Plateau	115 (9.9%)	334 (05/06/1997)	47.7; 5480 (13.9%)			
Valencia	43 (3.7%)	183 (04/29/1997)	30.3; 1301 (3.3%)			
Marine Murcia	36 (3.1%)	112 (05/19/2006)	19.0; 683 (1.7%)			
Marine	168 (14.4%)	565 (05/18/2009)	54.2; 9101 (23.2%)			
Andalusia						
Marine	20 (1.7%)	277 (05/04/2011)	45.6; 911 (2.3%)			
Plateau						
Marine Aragon	27 (2.3%)	240 (05/20/2003)	25.0; 675 (1.7%)			
Marine	33 (2.8%)	155 (05/23/1999)	14.4; 475 (1.2%)			
Catalonia						
Balearic	112 (9.6%)	159 (05/16/2003)	14.7; 1649 (4.2%)			
Islands						
Marine	50 (4.3%)	73 (06/04/2004)	17.2; 861 (2.2%)			
Valencia						
Algeria	58 (5.0%)	561 (05/23/2009)	28.0; 1626 (4.1%)			
Morocco	23 (2.0%)	63 (06/07/2012)	18.6; 427 (1.1%)			
Marine	16 (1.4%)	86 (05/13/2014)	13.7; 219 (0.6%)			
Occitania						
Marine Alpes	13 (1.1%)	109 (06/02/2005)	16.9; 220 (0.6%)			
Cote						
Corsica	6 (0.5%)	21 (04/25/2006)	12.2; 73 (0.2%)			
Sardinia	18 (1.5%)	16 (05/03/2008)	5.6; 100 (0.3%)			
Tunisia	9 (0.8%)	31 (05/30/2005)	10.3; 93 (0.2%)			
Mediterranean	115 (9.9%)	242 (05/19/2009)	16.9; 1943 (4.9%)			

Marine entails that the air mass back trajectories were located over the sea before reaching their destination.

^a Percentage of total number of days in the MPS between 1993 and 2014.

^b Percentage of total pollen grains in the MPS between 1993 and 2014.

closer to the IP. Likewise, significant differences appeared in the low pollen concentrations of the Mediterranean "source area". It should be noted that all these source areas with a low contribution were marine in nature. This is despite being, as in the case of the Balearic Islands, source areas with higher *Olea* pollen loads than those registered in Cartagena itself (Boi and Llorens, 2013). Given this marine pattern, all the days were grouped in marine source areas (N = 704) to compare them with those days that lacked a marine effect (N = 463) (Fig. S10): The result was clear, with a marine nature coinciding with the lowest concentrations (ANOVA: p-value <0.001) despite being more frequent events. This situation is in line with the marine effect on pollen concentrations in Australia (De Morton et al., 2011) and would help to explain the low concentrations in Cartagena compared to other enclaves of the IP (Elvira-Rendueles et al., 2019). It was possible to affirm the preponderance of the marine factor as an attenuator of the

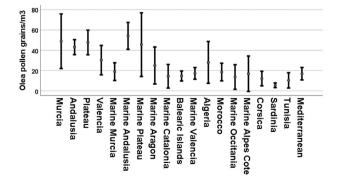


Fig. 5. Error bar graph for the mean (95% confidence interval, Cl) *Olea* pollen grains/m³ in Cartagena according to source zones (1993–2014).

Olea pollen concentrations in Cartagena, beyond the potential contributions of LDT.

In accordance with this approach, it could be expected that the source area where Cartagena itself is located, i.e., Murcia, had a great impact on pollen concentrations. Although the API in the Murcia source area was the fourth highest among all source areas (Table 2), the great dispersion of concentrations on days with this source possibly explained the lack of significant differences from corresponding source areas with LDT: Mediterranean islands, source areas in France or countries in Africa. In part, the significant dispersion in the Murcia concentrations could be explained by the fact that the release of pollen grains is less intense at the beginning and end of flowering (Aguilera and Ruiz Valenzuela, 2009). With low emissions of pollen grains, concentration is low. It seems more plausible that few pollen grains from nearby trees reached the trap, rather than few grains from distant trees. It is more likely that grains from farther away would have settled prior to the transporting air mass reaching the trap. In other words, the high dispersion of concentrations in the source area of Murcia is due to the fact that it is easier to capture low sources of pollen grains from olive trees in Murcia than the low emissions from olive trees in more distant areas.

With the attenuating marine nature, we also expect that the Marine Murcia source will have lower pollen concentrations than its nonmarine source area. Indeed, the pollen concentrations were lower, although without being statistically significant. Apart from the aforementioned dispersion of pollen concentrations in Murcia, the absence of a clear descent with the marine effect in a source area so close to the trap could be explained by the effect of the sea breeze, which would provoke the pollen caused by air recirculation to be retrained (Gassmann et al., 2002). In regard to this, a parallelism might be observed between airborne pollen grains and other pollutants such as ozone. For instance, sea breezes have provoked intensive ozone episodes along the Spanish Mediterranean coast (Querol et al., 2016).

To delve deeper into marine effects and the anomaly of high pollen concentrations in two source areas near Cartagena, i.e., Marine Andalusia and Marine Plateau, the source areas were segregated into four groups: No marine (N = 463), Marine Andalusia (N = 168), Marine Plateau (N = 20) and Other marine (N = 516) (Fig. S11 and Table S3). The pollen concentrations of Other marine were significantly lower (*p*-value <0.001) than those of No marine and Marine Andalusia. As indicated, the pollen concentrations of Marine Andalusia were quantitatively guite relevant. This indicates the great influence the contributions of the Andalusian olive groves had, in spite of the marine factor: even if the masses acquired a marine nature, the pollen concentrations would not be significantly attenuated. Speed, the relative moment when the air masses move over the sea and the time during which the masses remain over the sea are decisive in attenuation: a 12-h stay over the sea from the beginning of the calculation of the air mass back trajectories would not be the same as if the 12 h make up the last hours of the path before reaching the trap. In the first case, the air masses could be intensely loaded over the continental territory before reaching the trap; that is, the marine effect would be less recent. Taking into account the proximity to Andalusia, another conclusion was that intense advections would not be necessary to drag notable pollen loads towards Cartagena.

A paradox of high pollen concentrations could occur when they coincide with the eminently marine source area Mediterranean. On 05/ 19/2009 (242 pollen grains/m³), the highest pollen concentration with the Mediterranean source area appeared. Such a high value, the 98.6th percentile of 22 years, is striking for being the only source area defined as purely marine. The air mass back trajectories showed sinuous paths of only 200 km for 48 h (Fig. S12). The graphical representation showed that all air masses arrived by sea, although the corresponding air mass back trajectories of the lowest air masses had an inbound direction 180° from that of the highest air mass. This contrast of directions and gradient weakness could cause the elevation of *Olea* pollen concentrations. Low gradients in wind flows accompanied by local thermal L. Negral, S. Moreno-Grau, M.D. Galera et al.

circulations have been responsible for high pollen concentrations in other countries in the Mediterranean basin (Prtenjak et al., 2012).

3.2.1. Simultaneous use of back trajectory analysis and pollen concentrations during LDT

The difficulty of isolating pure LDT events in the pollen concentrations was caused by the high probability of grains being from nearby sources. Furthermore, sequences of more than two consecutive days in the MPS with the same distant source were very rare in Cartagena. Below these lines, two events are presented to exemplify LDT from African and European countries, the latter of which caused the annual peak date.

3.2.1.1. LDT from Algeria: 05/08/1998-05/10/1998. From 05/08/1998 to 05/10/1998, the air masses that arrived in Cartagena were NAF, while the source area was Algeria (Fig. 6). Pollen concentrations were 6, 6

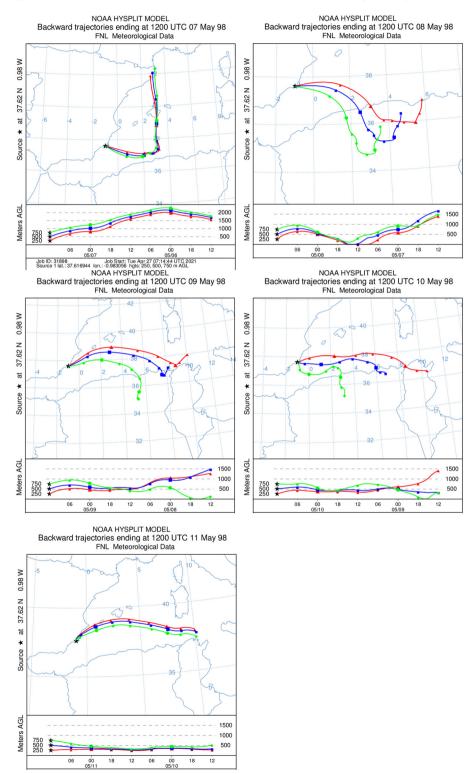


Fig. 6. Air mass back trajectories between 05/08/1998 and 05/10/1998, with LDT from Algeria and North African (NAF) air masses; the previous day, 05/07/1998, from the Balearic Islands with Regional (RE) air masses; and the following day, 05/11/1998, from Marine Murcia with Mediterranean (ME) air masses.

with 10 pollen grains/m³. These values were higher than the previous day with 1 pollen grain/ m^3 , with RE and Balearic Islands, and the day after with 5 pollen grains/m³, from ME and Marine Murcia. The increase in pollen concentrations during these three days cannot be justified by contributions close to the receptor since on 05/07/1998, the concentration was modest. In fact, 1 mm of rainfall was recorded on both 05/10/ 1998 and 05/11/1998, which could reduce transported pollen grains. African transport is visualised in the representation of the air mass back trajectories: the masses ran very close to ground level on their journey over Algerian territory during different periods. It was only over this territory that the air masses approached such low levels. During this event, the marine nature of the Algerian source was not an obstacle to diluting the LDT contribution since the source areas had a marine nature both immediately before and after the event. In other words, the fact that the increase in concentration occurred during the three central days points to an LDT phenomenon in the set of five days, all of which have marine source areas.

3.2.1.2. LDT from Sardinia, Tunisia, Balearic Islands and Alpes Cote: 05/29/ 2005 to 06/02/2005. The position of Cartagena allowed the concatenation of LDT from different source areas to be detected. On 05/29/2005 (10 pollen grains/m³), a multi-day LDT began with ME air masses and a Sardinian source area. The event exceeded one hundred pollen grains (Fig. S13). The day before, 05/28/2005 (7 pollen grains/m³), the air masses had also been ME, but without moving over any territory for 48 h before arriving to Cartagena, consequently being in the framework of the Mediterranean source zone. On 05/30/2005, despite 3 mm of rainfall, the levels rose to 31 pollen grains/m³, changing the masses to NAF with a Tunisian source.

On 05/31/2005, 17 mm of rain fell, which may have caused the slight decrease to 23 pollen grains/m³ with ME masses and the Balearic Islands source area. On 06/01/2005 (107 pollen grains/m³), the air masses turned to EU, with the lowest air mass back trajectory staying very close to ground level over the south of France; however, the source zone was the Balearic Islands, as it was the nearest territory the air mass back trajectories moved over.

On 06/02/2005, the annual peak date occurred (109 pollen grains/m³); the air masses returned to ME, not moving over any territory from the Marine Alpes Cote source area. Finally, on 06/03/2005 (5 pollen grains/m³), the levels indicated that the source area was Mediterranean with the same ME air masses, which explained the one order of magnitude drop in *Olea* pollen concentrations. It seems likely that this peak was caused by allochthonous pollen contributions since it occurred 10 days before the end of the MPS. Furthermore, the latitudinal difference between Cartagena and the south of France could explain a delayed flowering in specimens from that country.

4. Conclusions

Air mass back trajectories have been satisfactorily used as a tool for the classification of the air masses that transported Olea pollen grains and for tracing the source area where these grains were released before arriving to Cartagena. The fact that the city is a coastal enclave to the southeast of the IP, geographically surrounded by Olea pollen emitting sources, facilitated the study of the marine effect on carrier air masses and the LDT phenomena. The frequency with which air masses compatible with the LDT phenomena appeared in Olea pollen concentrations was 8.7% of the MPS days between 1993 and 2014. This percentage corresponded to intense advections from NAF or EU, explaining the low probability of LDT of olive pollen in Cartagena. In contrast, RE air masses were the most frequent: 38.6% of the days in the MPS. When comparing this frequency with Olea pollen concentrations, it was significantly lower (p-value <0.05) when the masses were EU compared to when they were RE. Broad dispersion of pollen concentrations with NAF air masses made it impossible to conclude significant differences from the concentrations of other air masses. There were important differences (p-value <0.001) between pollen concentrations with RE and ME air masses. The impact of the Atlantic air masses, with the exception of AN, was reflected in dragging from the olive-growing areas of the Iberian south, where the world's leading olive producers are located.

When examining source areas of Olea pollen grains in conjunction with air mass back trajectories, the importance of the Andalusian source was highlighted, attributing 27.2% of the total grains in the MPS. In the case of Marine Andalusia, 23.2% of pollen grains were added. Despite the high values of the Marine Andalusian source area, the set of sources that moved over marine waters had significantly lower concentrations than the set of sources of a non-marine nature (p-value <0.001). This could be due to the fact that the sea was not a source of pollen grains and because pollen grains gradually settled during the time that the air masses remained over the sea. Given this, other phenomena, such as the higher relative humidity of the air masses and the incorporation of marine features which alter the aerodynamic behaviour of pollen (for instance, sea salt), could also be responsible for the decrease in pollen concentrations. Further research is needed to investigate the performance of these marine agents in attenuating the concentration of bioaerosol

Marine factors had more impact on pollen concentrations than on pollen grains from remote sources corresponding to LDT. However, the definition of source areas led to the detection of LDT from Africa and Europe. The 2005 peak date was attributed to LDT from southern France. This observation was consistent with the late date at which it occurred: just 10 days before the end of the MPS, in harmony with the delayed latitudinal flowering of a northern country.

The study justified the variations of pollen concentrations around the days of maximum concentration for every type of air mass. Distortions in air mass paths depending on the type of air mass explained unexpected alterations in pollen concentrations on consecutive days. Recirculation and loops of air mass back trajectories varied the pollen load that each type of air mass could originally have. This suggests that cataloguing air masses alone was not sufficient to explain the fluctuations in pollen concentrations. This concern should be addressed during conditions of prevailing sea breezes that may impact local pollen concentrations. Among other factors, it is worth mentioning the different time spent over source areas, the evolution of the air mass in the vicinity of the destination and the moment when a marine nature was acquired.

CRediT authorship contribution statement

- L. Negral: Conceptualization, writing-original draft, investigation
- S. Moreno-Grau: Methodology, investigation, writing-review
- M.D. Galera: Analytic tools, investigation

B. Elvira-Rendueles: Data acquisition and airborne pollen data curation, investigation

- I. Costa-Gómez: Methodology, investigation
- **F. Aznar:** Investigation
- **R. Pérez-Badia:** Writing–review & editing
- J.M. Moreno: Funding, Writing–review & editing, supervision

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi. org/10.1016/j.scitotenv.2021.147999.

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