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Links between recent trends in airborne pollen concentration, meteorological parameters and air pollutants

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ABSTRACT

Biogenic aerosols may play an active role in various diseases. Pollutant gases and bioaerosols coexist in the atmosphere with the possibility of interacting with each other increasing their adverse impacts on human health. The study of long-term trends and the correlation between the pollen concentration from selected taxa (especially those related to allergies) and both the main atmospheric pollutants and the meteorological parameters has enabled us to identify the main factors that affect pollen concentration in the atmosphere. This study analyzes the long-term trend in CO, NO, NO_2, PM_{10}, SO_2 and O_3 from 1997 to 2016 and *Fraxinus*, Poaceae and *Populus* pollen concentrations in the city of León from 1994 to 2016. In general, there is a significant decreasing trend in atmospheric pollutant concentrations and a significant increasing trend in *Fraxinus* pollen concentrations. In addition, the influence of air pollutants and climatic factors on pollen concentrations and pollination period duration was studied using the Spearman correlation, showing that the flowering and pollination periods depend largely of the weather conditions before these periods and are influenced by air pollutant concentrations.

1. Introduction

In the atmosphere there are several pollutants that cause a negative impact on human health and the environment. The main air pollutants are carbon monoxide (CO), nitrogen oxides (NO_x), particulate matter (PM), which includes bioaerosols (pollen, fungal spore, bacteria, viruses, etc.) and non-biological particles, sulfur dioxide (SO_2) and ozone (O_3). These pollutants, gases and particles, coexist in the same medium, the atmosphere, with the possibility of interacting with each other and increasing their adverse impact due to synergistic effects (Monsalve et al., 2013). Thus, the exposure to O_3 is related to inflammatory effects in the respiratory tract, and the exposure to CO, NO_2, PM and SO_2 has been associated with cardiopulmonary mortality, pulmonary edema, problems in the central nervous system, and respiratory and cardiovascular hospital admissions (Bernstein et al., 2004; Brunekreef and Holgate, 2002; Curtis et al., 2006; Kampa and Castanas, 2008). The presence of high levels of bioaerosols in the atmosphere is usually linked to allergic respiratory diseases (Buters et al., 2012, 2015).

On the other hand, the presence of high levels of air pollutants may also affect the environment. For example, aerosols may scatter or absorb solar and infrared radiation (Calvo et al., 2010; Ren-Jian et al., 2012) and NO_x is the main precursor of acid rain, which may cause the acidification of soils, lakes, and streams, and accelerate the corrosion of buildings and monuments (Tang et al., 2005).

For many years, efforts have been made in Europe to reduce the levels of emission of air pollutants. In 1996 the Directive 96/62/CE about the Evaluation and Management of Ambient Air Quality was passed. This was the first standard adopting fixed criteria, objectives

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Abbreviations: APIn, Annual Pollen Integral; CO, carbon monoxide; CO_2, carbon dioxide; DP, days with precipitation; DS, days with snow; EGR, exhaust gas recirculation valve; LPG, liquefied petroleum gas; MPS, main pollen season; NEI, National Emissions Inventory; NO, nitric oxide; NO_2, nitrogen dioxide; NO_x, nitrogen oxides; O_3, ozone; P, accumulated precipitation; PM, particulate matter; PM_{10}, particulate matter (diameters $< 10\ \mu m$); REA, Spanish Aerobiology Network; RH, relative humidity; SO_2, sulphur dioxide; SPIn, seasonal pollen integral; T, temperature; T_{Max}, maximum temperature; T_{Min}, minimum temperature; VOC, volatile organic compounds.

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and evaluation techniques for air quality. From this one, in the following years, different Directives have been developed including Directives 1999/30/CE, 2000/69/CE, 2002/3/CE and 2004/107/CE. All these Directives were incorporated to the one currently in force, Directive 2008/50/EC relative to Ambient Air Quality and a Cleaner Atmosphere in Europe.

In Spain, the legal basis relative to the Protection of the Atmospheric Environment dates from 1972, when the Law 38/1972 was passed. Subsequently, this law was adapted to the European environmental needs for better prevention of the harmful effects of atmospheric pollutants over human health and the environment, and was integrated into the Royal Decrees 833/1975, 1073/2002, 1796/2003 and 812/2007. These Royal Decrees were derogated and integrated into the current Spanish regulation about Air Quality and Atmosphere Protection (Law 34/2007) and Improvement of Air Quality (Royal Decree 102/2011).

Long-term studies of air pollutants are essential to evaluate the effectiveness of the implementation of national and/or international environmental policies. In the past decade, several authors have reported a general and progressive decrease in air pollutant concentrations, attributable to different actions taken by local governments and international organizations to reduce emissions (Aziz et al., 2016; Ebelt et al., 2001; Guerreiro et al., 2014; Karanasiou et al., 2014; Querol et al., 2014). Studies focusing on the long-term trend of airborne pollen concentrations have been of great interest in recent years due to the increase in allergies, especially in urban and industrialized areas. In addition, airborne pollen is considered a sensitive indicator of plants to climate change (Clot, 2003; D'Amato et al., 2015, 2016; Oteros et al., 2015).

Although there are many studies on air pollutant concentration trends, there are few on pollen concentration trends (e.g. Galán et al., 2016; Sofiev et al., 2015) and those relating both topics are scarce. Atmospheric pollution has a direct effect on the physical, chemical and biological properties of pollen grains, and the change registered in the concentrations of air pollutants in the past decade and the impact of environmental regulations on the levels of air pollutants can be related to pollen concentrations and allergenic proteins which are potentially responsible for the increase in allergic diseases. According to Sénéchal et al. (2015), atmospheric pollutants may have several effects on pollen: i) increasing their potential health hazards; ii) alteration of the physicochemical characteristics of the pollen surface, iii) change in the allergenic potential, and iv) decrease in viability and germination. For example, high levels of CO_2 may be related to an increase in the airborne pollen concentration in large cities and, as a consequence, to an increase in respiratory allergies (Albertine et al., 2014; Rogers et al., 2006; Schmidt, 2016; Sharma et al., 2014). Evidence demonstrates that there is an interaction between air pollution and plant-derived respiratory disorders (Beggs, 2010; Bielory et al., 2012; D'Amato et al., 2000, 2014,2015). This interaction is influenced by several factors such as the type of air pollutants, plant species, climatic factors, chemical interactions, etc. (Beggs, 2004; D'Amato et al., 2007; Reinmuth-Selzle et al., 2017).

Due to the different levels of air pollutant concentrations and the numerous types of vegetation in different regions of Spain, and because of the lack of data on the connection between air pollutants and pollen concentration in the northwest of the Iberian Peninsula, it is interesting to assess this connection on the basis of long-term trends in the city of León. León, situated in the NW of the Peninsula, is an ideal place for this study because of its unique characteristics: it has a wide variety of vegetation associated with respiratory allergies and there is a Spanish Aerobiology Network (REA) station, with a historical database of pollen levels of several species in the city, together with an extensive database on air quality.

The aim of this study is to evaluate the trend in *Fraxinus*, Poaceae and *Populus* pollen concentration (related to allergies) as well as atmospheric pollutants in León (Spain) over the last two decades. The study also seeks to explore the correlation between the concentration of pollen and both the main atmospheric pollutants and the meteorological parameters. Additionally, the location of the main sources of atmospheric pollutants and pollen from the selected taxa in León will also be investigated.

2. Material and method

2.1. Study zone

The study was carried out in the city of León, located in the northwest of the Iberian Peninsula (42° 36′ N, 05° 35′ W and 838 m above sea level), between 1997 and 2016. The climate is Mediterranean with continental features. The mean seasonal weather conditions obtained from the National Agency for Meteorology (www.aemet.es) are presented in Table 1.

The population of the city of León and the nearby municipalities (San Andrés del Rabanedo, Villaquilambre, Valverde de la Virgen and Onzonilla) has grown, according to the data presented by the National Institute for Statistics (www.ine.es), from 176,333 inhabitants in 1998 to 185,393 inhabitants in 2016. With regard to the number of cars, the regional government of the Junta de Castilla y León registered an increase in the province of León from 301,365 vehicles in 2005 to 347,174 vehicles in 2015 (www.estadistica.jcyl.es). Besides, the General Agency for Traffic (www.dgt.es) reports that the vehicles registered are grouped as follows: buses (0.2%), trucks (8.1%), mopeds (4.6%), vans (8.0%), motorcycles (6.3%), private cars (68.4%) and others (4.3%). Fig. 1 shows the classification of these vehicles by type of fuel (diesel, gasoline and electric) from 2010 to 2016.

Table 1
Maximum, minimum and mean values of temperature (T), relative humidity (RH), days with precipitation (DP) and days with snow (DS) by season in the city of León.

Season	Months	T (°C)	T_{Min} (°C)	T_{Max} (°C)	RH (%)	DP (mean precipitation)	DS
Winter	January, February, March	5.2	0.4	10.0	74	6.4 (39 mm)	2.9
Spring	April, May, June	12.9	6.7	19.1	61	4 (44 mm)	0.3
Summer	July, August, September	18.6	11.5	25.7	56	3.3 (27 mm)	0
Autumn	October, November, December	7.6	3.3	12.0	79	8.1 (62 mm)	1

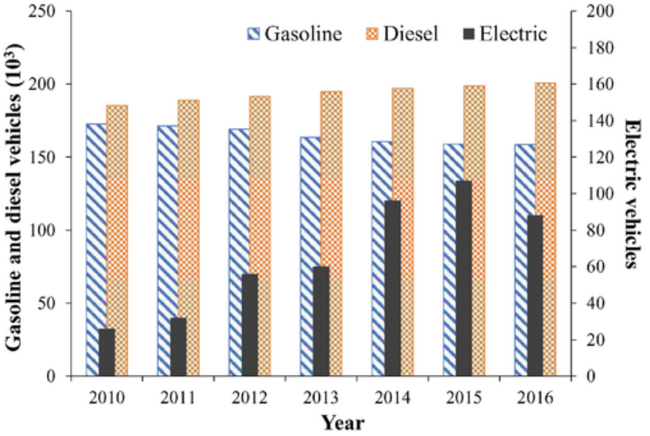

Fig. 1. Number of electric, gasoline and diesel vehicles in the province of León (data from the Spanish General Agency for Traffic -DGT).

The mountains located north of the province (about 30 km far from the city) are an important area of origin of bioaerosols because of their numerous forests, with many types of vegetation, whose pollination contributes to a high pollen concentration. In addition, deciduous forests, with poplars (*Populus* L.) and ash trees (*Fraxinus* Tourn. ex L.), are present in numerous valleys and as ornamental flora. A variety of grasses (Poaceae Barnhart) and other herbaceous plants are also found in grasslands and meadows (Del Río González, 2005).

These pollen types (*Fraxinus*, Poaceae and *Populus*) have been chosen for analysis because grass pollen (16–40 μm size) is the main cause of respiratory allergies in the world (D'Amato et al., 2010) and also in León. The other two pollen types correspond to pre-spring flowering species with high incidence in pollinosis in the area. *Fraxinus* (pollen size: 15–24 μm) belongs to the Oleaceae family, one of the most allergenic types in Mediterranean regions and whose incidence is increasing in the city of León due to cross-reactivity with the rest of the family's species (Lombardero et al., 2002). *Populus* (pollen size: 22–30 μm) is a taxon not always well identified as responsible for pollinosis, as its symptoms coincide with those from viral conditions characteristic of this season.

2.2. Sampling and data bases

Atmospheric pollen concentration was analyzed using a Hirst volumetric trap (Hirst, 1952), placed on the terrace of the Faculty of Veterinary Sciences of the University of León (42° 36′ 50″ N, 5° 33′ 38″ W, 846 m) from 1994 to 2016 (Fig. 2). The daily and hourly optical microscopic counts were carried out using the method recommended by the Spanish Aerobiological Network (Galán Soldevilla et al., 2007). The average daily and hourly pollen concentrations for *Fraxinus*, Poaceae and *Populus* were expressed as pollen grains per cubic meter of air. The Main Pollen Season (MPS) was defined by Galán et al. (2017) as the period when the atmosphere contains significant concentrations of pollen and the Seasonal Pollen Integral (SPIn) as the integral over time of pollen concentration expressed as pollen day m^{-3}. There are different methods to define the main season start and end (Jato et al., 2006; Pfaar et al., 2017), depending on the main goal of the study. For our work, the consecutive days representing 95% of the SPIn, were selected to establish the MPS, starting on the day on which the accumulated value reaches 2.5% and ending on the day when 97.5% of the SPIn is reached (Andersen, 1991). This criterion has been widely used in many works (Chiesa and Toletti, 2004; Recio et al., 2010; Rojo et al., 2015).

The study of the evolution of pollen concentration was carried out in three periods of four months each (January–April, May–August, and September–December). These three groups will allow us to define for each species the flowering period. The other two periods will be considered as previous to the flowering period.

For this study, we have used the data provided by the Air Quality Network of the Junta de Castilla y León (www.servicios.jcyl.es/esco/), from January 1997 to December 2016 for the traffic station León1, located in San Ignacio de Loyola Avenue (05° 35′14″W 42° 36′14″N), for the air pollutants CO, NO, NO_2, PM_{10}, SO_2 and O_3 (Fig. 2). This station is located in an urban residential area with medium traffic. The main sources of pollution in León are vehicular emissions and residential devices used for heating, especially in the cold months (Oduber et al., 2018).

The meteorological parameters (mean temperature, relative humidity, minimum temperature, maximum temperature, wind direction, wind speed and accumulated precipitation) during the study period (1994–2016) were obtained from the database of the National Agency for Meteorology (www.aemet.es). The atmospheric pollutant emission levels were drawn from the National Emissions Inventory of the Spanish Ministry of Agriculture, Nature and Food Quality (MAGRAMA- www.magrama.gob.es) and the data on the production and consumption of energy in the province of León were taken from the website of

Fig. 2. Map of the city of León.

the regional government Junta de Castilla y León (www.energia.jcyl.es).

2.3. Statistics treatment

The statistical treatment has been carried out using R software (www.R-project.org) with the Openair package (Carslaw, 2015; Carslaw and Ropkins, 2012) and SPSS (IBM Statistics Software V. 24). The trend calculations were carried out using Theil-Sen methodology and non-parametric Mann-Kendall tests (Hipel and McLeod, 2005; Kendall and Gibbons, 1990) for a significance level of $p < 0.001$. The correlation among pollen concentrations, pollutant concentrations and meteorological parameters (temperature, relative humidity and rain) was made using the nonparametric Spearman's rank correlation method.

3. Results and discussion

3.1. Trends in air pollutant concentrations and meteorological parameters

The Mann Kendall Trend test was applied to the meteorological parameters and the pollutant concentrations registered by the traffic station León1. Regarding the meteorological parameters, no significant trends were observed during the study period.

The application of the Mann Kendall Trend test to pollutant concentrations showed the following results (Fig. 3):

- A significant decreasing trend of CO concentrations beginning around 2006 (−0.07 mg m-3 year-1).
- The NO concentrations show a significant decrease (−1.73 µg m-3 year-1), and the Man-Kendall test shows that this trend begins around 2007.
- The NO2 concentrations show a significant decrease (−1.78 µg m-3 year-1), from 2002.
- A significant decreasing trend of PM10 concentrations in the study period (2.35 µg m-3year-1) beginning around 2004.
- A statistically significant decrease in SO2 concentrations (−2.08 µg m-3 year-1), beginning around 2007.
- O3 concentrations show a significant decrease beginning around 2000 (−1.38 µg $m^{-3}year^{-1}$).

The general decrease in air pollutant emissions is probably the result of a series of measures taken in different sectors such as industrial, public electricity and road transport, so that from 2008 the recorded values were within the permissible limit values. The CO and SO_2 concentrations decreased by 85% each, NO and NO_2 concentrations dropped by 66 and 55%, respectively, PM_{10} decreased by 76% and O_3 by 31%. Several papers have found similar results in other cities. For example, in Germany, Ebelt et al. (2001) reported a reduction in the NO and SO_2 concentrations between 1990–1999 (91% and up to -50%, respectively). Aziz et al. (2016) showed a decreasing trend in SO_2 levels (−70%), CO (−32%), NO_x (−39%) and PM_{10} (−48%), but an increasing trend in O_3 (+79%), in Bangkok between 2000 and 2015. The same pattern was found by Guerreiro et al. (2014) in the study of 38 European cities between 2002 to 2011, with a general decrease of SO_2 (between 34 and 50%), NO_x (between 23 and 27%) and CO (between 27 and 32%) concentrations. In different cities of Spain Querol et al. (2014) reported a decrease in the CO (up to −6.4% $year^{-1}$), SO_2 (up to −7.7% $year^{-1}$), NO_2 (up to −3.7% $year^{-1}$) and PM_{10} (up to −5.1% $year^{-1}$) concentrations between 2001 to 2012, which are comparable to those found in this study (−5.01, −5.32, −3.39 and −4.11% $year^{-1}$, respectively). Karanasiou et al. (2014) also reported a decreasing trend in SO_2, NO_2, PM_{10} and CO concentrations in Barcelona (−0.21, −0.65, −2.2 µg m^{-3} $year^{-1}$, -0.02 mg m^{-3} $year^{-1}$, respectively) during the period of 2003–2010, a decrease of the SO_2 and CO concentrations (−0.97 µg m^{-3} $year^{-1}$ and −0.11 mg m^{-3} $year^{-1}$, respectively) in Madrid between 2001 and 2009, and a decrease in PM_{10} concentrations (−2.11 µg m^{-3} $year^{-1}$) in Huelva (2003–2010). A more detailed analysis of each sector can be found in the supplementary material.

3.2. Long-term trends SPIn and MPS

The seasonal pollen integral, SPIn, varied between 38 (year 2000) and 732 (year 2015) pollen day m^{-3} for *Fraxinus*, between 1625 (year 2009) and 7072 (year 2000) pollen day m^{-3} for Poaceae and between 296 (year 2004) and 2992 (year 2012) pollen day m^{-3} for *Populus*. The

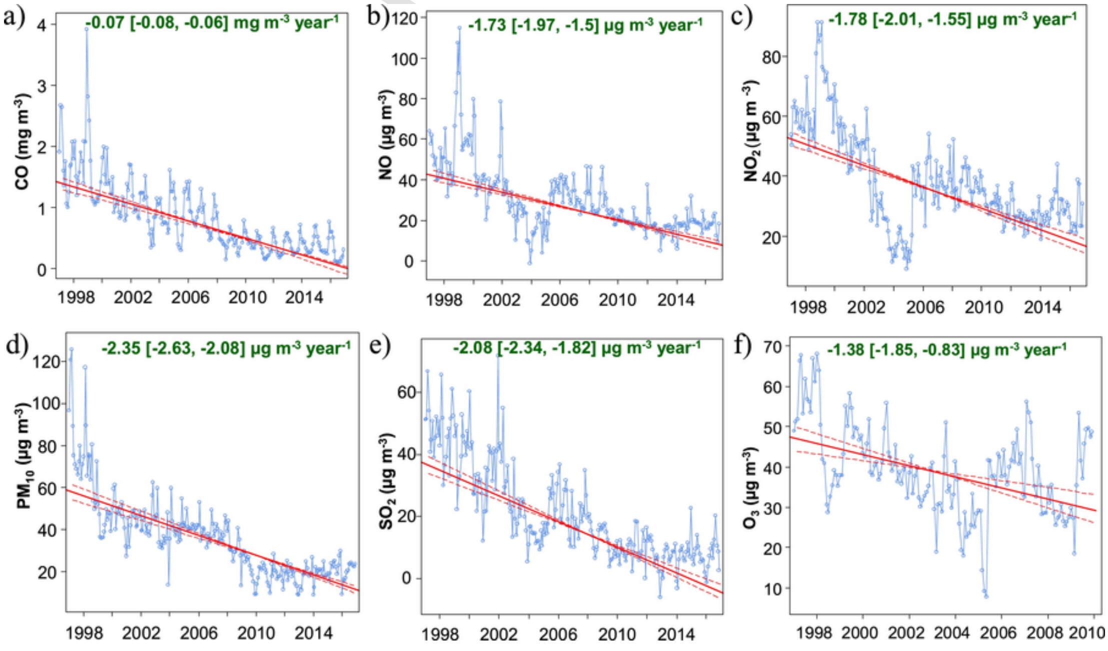

Fig. 3. Long-term trends of a) CO, b) NO, c) NO_2, d) PM_{10}, e) SO_2 and f) O_3 concentrations, in the traffic station León1. The solid red line shows the estimated lineal trend and the dashed red lines show the 95% confidence intervals for the trend. The overall trend is shown at the top and the 95% confidence intervals in the slope. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

Mann Kendall trend test was applied to the SPIn. The results showed that only *Fraxinus* had a statistically significant increasing trend in pollen concentration (beginning in 2006), with an increase of 10 pollen day m^{-3} $year^{-1}$ ($p < 0.01$).

In a previous study carried out in different cities of Spain (Córdoba, Granada, Barcelona and Orense) between 1994–2013, Galán et al. (2016) reported similar results. They found that the *Fraxinus* SPIn had a positive significant trend in all cases. In some European cities, the increasing trend observed for SPIn of several pollen has been attributed to the increase in temperatures in the past decade (Ziello et al., 2012). However, in León we observed a significant decreasing trend in the minimum temperatures (−0.07 °C $year^{-1}$, $p < 0.01$) from 1994 to 2016, and no significant trend in the mean or maximum temperatures that can be linked to the trend observed in the *Fraxinus* SPIn. The increasing trend in the pollen concentrations may be due to the fact that many ornamental trees planted in the city in the past two decades have reached maturity and therefore their floral and pollen production is very abundant. The decrease in the minimum temperatures also allowed for a more suitable vernalization and, in consequence, optimum flowering.

In the case of Poaceae and *Populus* SPIn, they have a non-significant positive and negative trend, respectively. Some authors report different trends for these two pollen types SPIn in other cities. Damialis et al. (2007) observed a significant positive trend for Poaceae and a significant negative trend for *Populus* SPIn in Greece. These trends are linked to a rise in the temperatures and changes in the local pollen due to the urbanization in the area of study. In Spain, Galán et al. (2016) show that Poaceae SPIn has a significant negative trend in Santiago, Badajoz and León and a positive one in Orense and Cartagena, showing clear differences due to the geographical distribution of species. The different trends observed could be mainly attributed to local human activities, and changes in rainfall patterns. These behaviors suggest different endogenous processes of adaptation to the local climate for the different taxa.

Fig. 4 shows the monthly evolution of pollen concentration during the study period. This allows us to identify the pollination period for

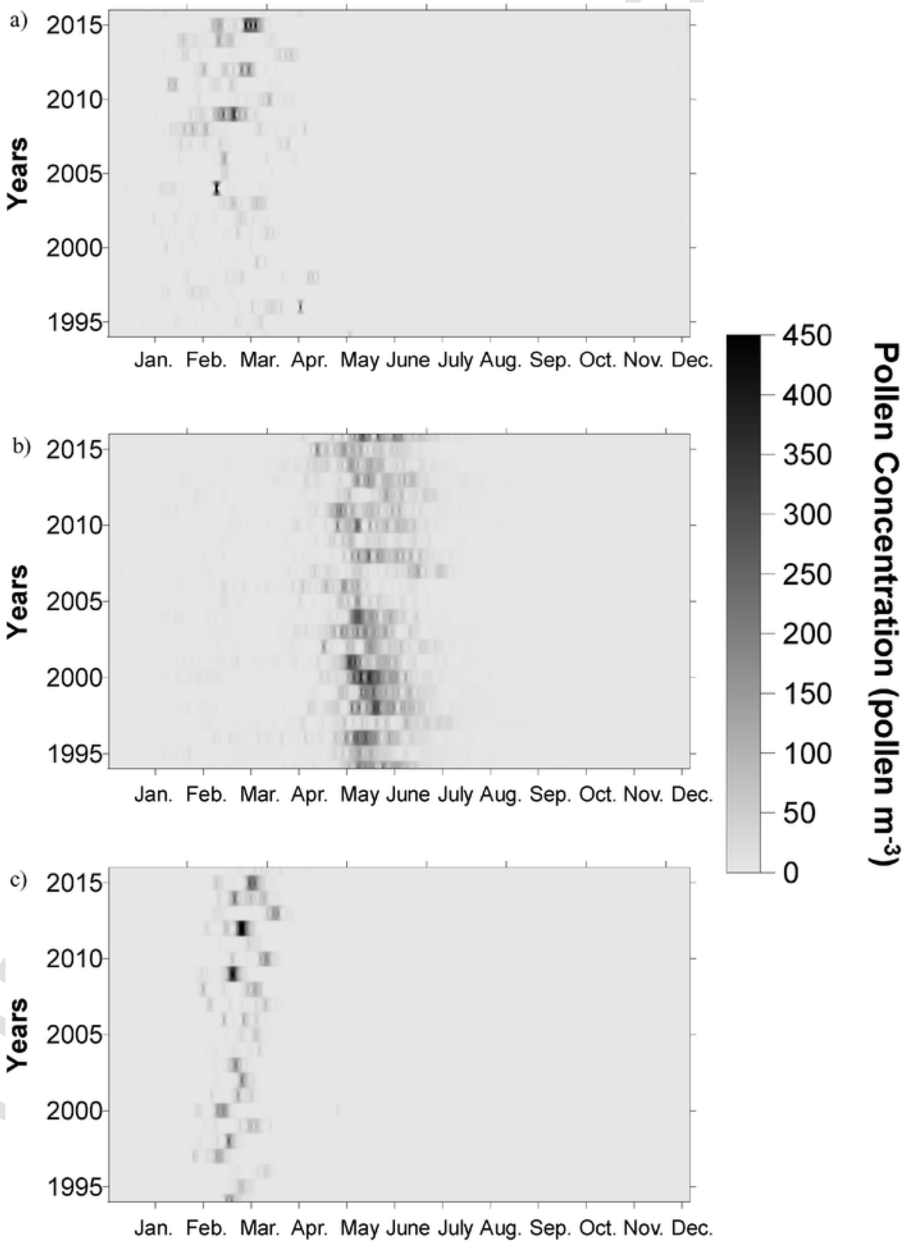

Fig. 4. . Evolution of the daily pollen concentration between the years 1994 and 2016 for a) *Fraxinus* (concentrations × 5), b) Poaceae and c) *Populus.*

each species and its changes over the years. The MPS of the *Fraxinus* starts at the end of January and ends in mid-April (average duration of 77 days, minimum of 32 days in 2005 and 2015 and maximum of 136 days in 2016); the MPS of Poaceae starts at the end of March and ends in the first days of August (average duration of 136 days, minimum of 89 days in 2015 and maximum of 195 days in 1997). Regarding *Populus,* the flowering season begins in the first days of March and ends in April (average duration of 37 days, minimum of 17 days in 1994 and maximum of 89 days in 2000). No trends in the advance or in the delay of the main pollen season and in the number of days with presence of pollen have been observed in any of the three taxa analyzed. However, some authors, such as Recio et al. (2010), reported an advance of 2 days year^{-1} and a delay of 7 days year^{-1} in the study of the MPS trend, between 1992–2007, of Poaceae in Malaga, Spain. These authors reported that the increase in rainfall at the beginning of spring delays the start of MPS, most probably because the water content of the soil favors vegetative development and inhibits early flowering. Moreover, the end of pollination is affected by the increase in the minimum temperature in spring due to the withering of the flowers and dehydration, especially, of the herbaceous plants, which can dry quickly when there is low availability of water. The meteorological conditions before the flowering period have an important influence on this process and in the release of pollen. The beginning of the main pollen season in these plants is related to changes in temperatures, which must reach a level that allows the end of the vernalization period of the plants (Galán et al., 2016; Puc and Bosiacka, 2011).

In Toledo, Spain, García-Mozo et al. (2006) reported that the flowering period for *Fraxinus*, Poaceae and *Populus* started in February, April and January, respectively. In Córdoba Velasco-Jiménez et al. (2013) observed that the flowering period of Poaceae and *Populus* started in April and February and ended in June and April, respectively, coinciding with the results found in this study. García-Mozo et al. (2006) found in Toledo that the start of the summer and the increase in the temperatures coincide with the decreasing pollen concentration (most abundant taxas Cupressaceae, *Quercus*, Poaceae, *Populus*, *Olea*, Urticaceae, *Platanus*, *Pinus* and *Ulmus*). They observed that in Toledo the autumn is the season with lowest pollen concentrations, as very few species flower in this season and rainfall cleans the atmosphere (scavenging). The same behavior was observed in the present study in the city of León.

3.3. *Correlation of pollen concentration with meteorological parameters and air pollutants*

3.3.1. *Correlations between pollen concentration and meteorological parameters*

The Spearman coefficients were computed between pollen concentration parameters (SPIn and MPS) and meteorological parameters (mean temperature, relative humidity, minimum temperature, maximum temperature and accumulated precipitation) for *Fraxinus*, Poaceae and *Populus*, following previous studies: Grinn-Gofroń and Bosiacka, (2015); Plaza et al. (2016); Puc, (2012); Rathnayake et al. (2017); Rojo et al. (2015); Sabo et al. (2015); Tassan-Mazzocco et al. (2015); Vara et al. (2016). The calculations were made on an annual and a four-month basis. This way we obtained the correlation coefficients before and during the flowering period (Table 2).

In León, the SPIn of *Fraxinus* and *Populus* have not a significant correlation with the meteorological parameters. However, previous studies carried out in Northern Serbia and in the city of León have reported a significant correlation between different types of APIn (*Annual Pollen Integral*) pollen and relative humidity (Fernández-Gonzáles et al., 1993; Sabo et al., 2015). Variations in relative humidity affect the physiology of plants and, consequently, their phenology. These variations can also influence the emission and dispersion of grains of pollen to the atmosphere, either because the anthers are not dry enough to release the pollen or because the pollen, once in the air, becomes partially hydrated and scatters hardly. Consequently, the correlation between relative humidity and SPIn depends mainly on the plant phenology. The morphology of pollen, that is, the shape and characteristics of its wall, hardly influence these correlations. In addition, Makra et al. (2014) have reported that the relative humidity has an important influence on dry climate conditions, where the low availability of groundwater is replaced in part by relative humidity.

Table 2
Spearman coefficients between pollen concentration parameters (SPIn and MPS) and meteorological parameters (mean temperature (T), relative humidity (RH), minimum temperature (T_{Min}), maximum temperature (T_{Max}), and accumulated precipitation (P)) for *Fraxinus*, Poaceae and *Populus*.

	Fraxinus		Poaceae		*Populus*	
	SPIn	MPS	SPIn	MPS	SPIn	MPS
Annual						
T	0.02	−0.02	−0.08	0.21	0.03	**−0.45***
T_{Min}	−0.36	0.06	−0.20	0.06	−0.20	**−0.44***
T_{Max}	0.31	−0.03	0.11	0.24	0.08	−0.11
RH	−0.16	0.19	0.38	−0.20	−0.29	0.08
P	−0.38	0.14	0.36	−0.05	0.00	−0.17
January-April						
T	−0.05	0.19	0.08	0.17	−0.05	−0.20
T_{Min}	−0.21	**0.44***	0.28	0.12	−0.23	−0.08
T_{Max}	0.28	−0.13	−0.25	0.26	0.23	−0.15
RH	−0.10	**0.51***	0.27	−0.23	−0.28	0.30
P	−0.15	**0.59****	**0.52***	−0.23	−0.16	0.19
May- August						
T	−0.07	−0.17	0.04	−0.17	0.00	−0.34
T_{Min}	−0.33	−0.13	0.04	−0.17	−0.14	−0.37
T_{Max}	0.15	−0.09	−0.01	−0.26	0.04	−0.17
RH	−0.12	0.13	0.32	−0.04	−0.34	0.14
P	−0.13	0.15	**0.44***	−0.22	−0.17	−0.14
September-December						
T	−0.04	−0.11	−0.39	0.41	0.01	−0.14
T_{Min}	−0.20	−0.33	−0.30	**0.42***	0.04	**−0.45***
T_{Max}	0.24	0.09	−0.37	0.32	0.05	0.20
RH	−0.02	−0.38	0.06	−0.17	0.20	−0.41
P	−0.24	−0.40	−0.15	0.14	0.24	−0.26

** $p < 0.01$.
* $p < 0.05$.

The *Fraxinus* MPS was positively correlated with relative humidity, minimum temperature and accumulate precipitation during the flowering period (January–April) (0.51, 0.44, 0.59, $p < 0.05$ and $p < 0.01$, respectively,). Thus, the duration of the main pollen season is favored by cold and wet conditions, typical of autumn and winter in León. Other authors have reported the influence of minimum temperatures and rainfall on the *Fraxinus* pollination period (Jato et al., 2004; Peeters, 2000; Tsai et al., 2016), and observed that the main pollen season is longer if there are low temperatures and heavy rain when the first pollen grains are present in the air.

The Spearman correlation coefficients show that the *Populus* MPS has a significant negative correlation with the minimum temperature before flowering (−0.45, $p < 0.05$). Makra et al. (2012) report an opposite behavior for *Populus* in southern Hungary and conclude that the main pollen season is extended in a warm and dry climate, promoting pollen release. In contrast, in León, the extension of the flowering period of *Populus* is favored by low temperatures. As a result, it has been observed that the *Populus* shows a great tolerance to climate change with extensions of the MPS favored in both warm and cold conditions.

The Poaceae MPS correlates with the minimum temperature before the flowering period (0.42, $p < 0.05$) and SPIn shows a significant positive correlation with the rainfall before and during the flowering period (0.52, 0.44, respectively, $p < 0.05$). Authors like Cariñanos et al. (2004), which have observed in different regions of Spain a more intense flowering if there was a period of rain in the 2–4 weeks previous to the flowering season. García-Mozo et al. (2016) and Makra et al. (2012) have reported the same behavior in studies carried out in Córdoba (Spain) and in Szeged (Hungary), respectively. Moreover, Recio et al. (2010) found a significant correlation between Poaceae SPIn and the total annual rainfall and the minimum temperature in Málaga (Spain). These four cities (León, Córdoba, Málaga and Szeged) are located between latitudes of 36° and 46 °N, with a population between 130,000 and 570,000 inhabitants, are characterized by low rainfall, hot summers and cold winters, with the exception of Málaga, which has mild winters due to its location in the southern coast of the Iberian Peninsula. In contrast to León, Córdoba and Málaga have average temperatures during spring that are higher than 10 °C; however, the average annual precipitation is similar to that observed in León. These studies show that, if there is enough water available, pollen concentrations during the late spring can be high, even in cases where the early spring temperatures are higher. If the spring is cold and the precipitation at the beginning of summer is high, pollen grains can be removed from the air and the pollen concentration can be reduced to a minimum, shortening the MPS.

3.3.2. Correlations between pollen concentration and air pollutants

The Spearman coefficients were also computed for SPIn and MPS and the air pollutant concentrations (CO, NO, NO_2, O_3, PM_{10}, SO_2) recorded by the traffic station León1 (Table 3). Significant correlations were only observed for *Fraxinus*.

Spearman correlation shows that the *Fraxinus* SPIn are negatively correlated with the concentration of several contaminants. However, the Poaceae and *Populus* SPIn and MPS do not show a clear correlation with air pollutant concentrations. In studies carried out in Oporto, Portugal, (Sousa et al., 2008) found a non-significant correlation of *Fraxinus* and Poaceae pollen with O_3 and PM_{10} concentrations.

As mentioned above, the flowering period of *Fraxinus* is from January to April. The O_3 concentrations before the flowering period are negatively correlated with the *Fraxinus* pollination season (MPS). Consequently, high levels of O_3 may inhibit the plant development and cause a shortening of the pollination period. In some urban areas, where the O_3 levels are low, an improvement in plant growth has been reported (Ziello et al., 2012).

Table 3
Spearman coefficients between pollen concentration parameters (SPIn and MPS) and atmospheric pollutant concentrations (CO, NO, NO2, PM10, SO2, and O3) for Fraxinus, Poaceae and Populus.

	Fraxinus		Poaceae		*Populus*	
	SPIn	MPS	SPIn	MPS	SPIn	MPS
Annual						
CO	**−0.484***	−0.096	0.262	−0.080	−0.186	−0.337
NO	**−0.468***	−0.223	0.194	−0.121	−0.056	−0.338
NO_2	−0.436	−0.233	0.134	−0.105	0.023	−0.181
O_3	−0.533	−0.209	0.231	0.061	0.258	−0.069
PM_{10}	**−0.553***	−0.084	0.292	−0.111	−0.313	−0.243
SO_2	**−0.531***	−0.105	0.248	−0.028	−0.287	−0.361
January-April						
CO	**−0.598****	−0.138	0.313	−0.177	−0.215	−0.377
NO	−0.379	-0.210	0.182	−0.056	0.086	−0.321
NO_2	−0.238	−0.175	0.230	−0.147	0.229	−0.156
O_3	−0.302	−0.302	0.401	0.019	0.071	0.127
PM_{10}	−0.376	−0.155	0.189	0.110	−0.241	−0.399
SO_2	−0.383	−0.133	0.179	0.031	−0.075	−0.438
May- August						
CO	**−0.450***	−0.041	0.338	−0.088	−0.144	−0.272
NO	**−0.600****	−0.154	0.239	−0.096	−0.274	−0.319
NO_2	**−0.532***	−0.318	0.029	−0.041	0.027	−0.274
O_3	−0.291	−0.390	−0.104	0.283	0.258	−0.228
PM_{10}	**−0.555***	−0.090	0.286	−0.098	−0.245	−0.279
SO_2	**−0.641****	−0.005	0.317	−0.120	−0.418	−0.252
September- December						
CO	**−0.484***	−0.086	0.111	0.042	−0.214	−0.418
NO	**−0.502***	−0.138	0.135	−0.175	−0.107	−0.132
NO_2	−0.438	−0.151	0.141	−0.126	0.042	−0.114
O_3	−0.533	**−0.637***	−0.170	0.217	0.159	−0.228
PM_{10}	**−0.576****	0.067	0.323	−0.184	−0.409	−0.205
SO_2	**−0.644****	0.128	0.340	−0.087	−0.430	−0.196

** *$p < 0.01$.*
* *$p < 0.05$.*

The *Fraxinus* SPIn also shows a significant negative correlation with SO_2 concentrations before the pollination season. Air pollutants such as SO_2 and NO_2 can negatively affect the development of the plant and the flowering phenology and, consequently, reduce the pollen production. Moreover, in vitro studies have demonstrated that the exposition of pollen to SO_2 and NO_2, even under safe values for human health, cause a general drop of the soluble protein content released by pollen including allergen (Bist et al., 2004; Cuinica et al., 2013, 2014; Sousa et al., 2012).

The significant correlation observed for both the *Fraxinus* pollen and the air pollutant concentrations coincides with the long-term trends described in sections 3.2 and 3.3. Even though the correlation may simply indicate unrelated trends in time, we speculate that the statistically significant increase on pollen concentrations of *Fraxinus* in León, may have been influenced by the decrease in the air pollutant concentrations in León since 1997.

Table 3 shows a negative correlation between the particulate matter and the pollen concentrations. Several authors have demonstrated that the particulate matter in the atmosphere adheres to the pollen surface of different species (Bellanger et al., 2012; Chehregani et al., 2004; Majd et al., 2004; Okuyama et al., 2007; Rezanejad, 2007; Rezanejad et al., 2003), causing morphological modifications and anomalies in the pollen structure (Majd et al., 2004; Majd and Mohamadi, 1992; Molfino, 1992; Rezanejad, 2007; Ruffin et al., 1983). In this study we have not found an influence of the particulate matter onto the pollen MPS.

3.4. Location of the main sources

For the identification of the pollen and pollutant sources, Spearman correlation between pollutant concentrations, correlation between SO_2 with T_{Min} and polar plots were carried out. The Spearman correlation coefficients were computed for the pollutants measured in the traffic station León1. There is a significant correlation between PM_{10} and NO_x (0.6, $p < 0.01$) which points towards road traffic emissions as a common origin. The high correlation between PM_{10} and CO (0.74, $p < 0.01$) shows the relationship with the primary emissions from combustion processes. The PM_{10} also correlates with SO_2 (0.75, $p < 0.01$) due the contribution of fossil fuel combustion (Calvo et al., 2013). This behavior was also reported by Karanasiou et al. (2014) in Spanish cities like Madrid, Barcelona and Huelva.

It was observed that there is a negative correlation between SO_2 concentrations and minimum temperatures in León (−0.17, $p < 0.01$). This behavior may be associated to the noteworthy contribution of coal combustion devices (emitting high amounts of SO_2) still in use in León during cold months, as evidenced by the increase of SO_2 (19 ± 20 and 24 ± 23 $\mu g\ m^{-3}$ in summer and winter, respectively).

The polar plots in Fig. 5 show the variation of the air pollutant concentrations with wind direction and speed, using the concentration data for the entire study period.

In general, a decrease in the air pollutant concentrations was observed with the increase of wind speed (except for ozone). The main emission sources of NO, NO_2 and SO_2 come from the residential areas around the sampling point, which may include heating systems, industrial areas and road traffic emissions.

Daily variation of the pollen concentration with the wind direction and speed during the pollination seasons over the entire study period is represented in Fig. 6. Polar plots show that the main source of pollen is located close to the sampling point and towards NE. In general, a decrease in the pollen concentration is observed when wind speed increases.

The origin of the pollen from the NE and its high concentration in the case of *Fraxinus* is a consequence of the fact that the largest plant

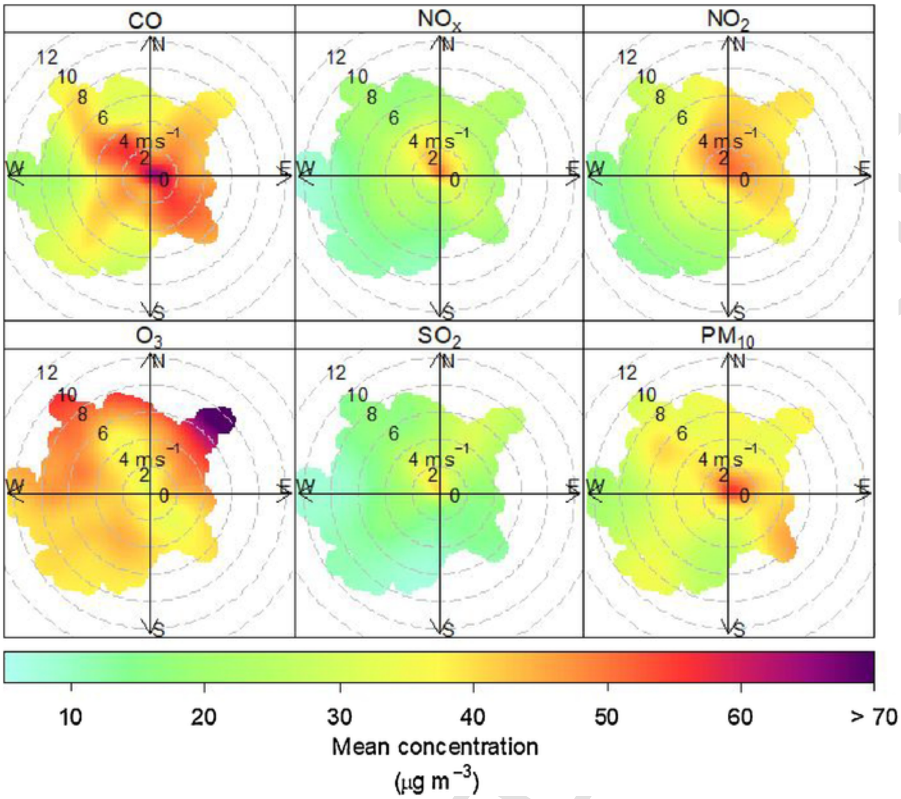

Fig. 5. . Polar plots (daily variation of the pollutant concentrations as a function of wind speed and direction) for CO (concentration × 5·10^4 µg m^{-3}), NO$_x$, NO$_2$, O$_3$, SO$_2$ and PM$_{10}$ concentrations of the entire study period.

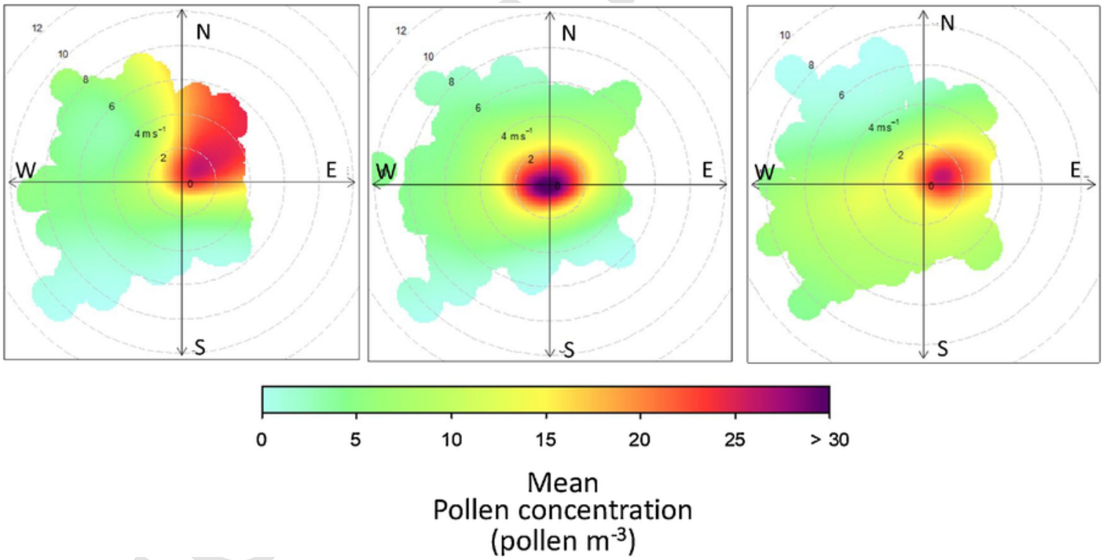

Fig. 6. . Polar Plots (variation of the pollen concentration during the pollination period as a function of wind speed and direction) for a) *Fraxinus* (concentration × 5), b) Poaceae and c) *Populus*, with concentrations of the entire study period.

mass is found on the Torío riverside, less than 3 km from the sampling station. The prevailing pollen types come from native species characteristic of these habitats and in very small numbers from species grown for ornamental purposes next to the sampling collector. Something similar happens with *Populus*, but the scarce concentration (in spite of the large pollen production of this species) shows that most of the trees of this species are further away from the sampling station, although they are also part of the riverside. A smaller amount comes from the SE, where there are also plantations of this tree for logging, about 12–20 km far from the sampling point. The ways of dispersion for arboreal species in relation to the wind were analyzed in some areas of Spain (Maya-Manzano et al., 2017) and Europe (Skjøth et al., 2013). Regarding grasses, the distribution observed in Fig. 6 clearly shows that the pollen comes from the surroundings of the sampling station, as expected from these herbaceous plants widely distributed in meadows and gardens (Peel et al., 2014).

4. Conclusions

The evolution of two decades of data on air pollutants, meteorological parameters and pollen concentration of Fraxinus, Poaceae and Populus, was analyzed in this study. In general, a significant decreasing trend in the atmospheric pollutant concentrations in the city of León was observed, probably due to the efficiency of international and national policies, and to the local measures taken for reducing emissions from road traffic, industrial activities and public electricity. However, no trend was detected in any of the meteorological parameters studied. Likewise, no trends in the advance or in the delay of the main pollen season, as well as in the number of days with presence of pollen were recorded in any of the three taxa analyzed. The observed correlations of the air pollutant concentrations with both the seasonal pollen integral and the main pollen season for Fraxinus, mainly in the months before the pollination period, could suggest a relationship between the increasing trend in Fraxinus pollen concentrations and the decreasing trend in air pollutant concentrations. Moreover, the evolution of the concentrations of the three studied pollen taxa also depends on the changes in the urbanization of the city of León. The localization of the main sources of the three studied taxa is close to the sampling point and in the NE sector, where the main plant mass is located. Another minor contribution comes from the ornamental plants next to the sampling collector. The Spearman correlation shows that the flowering and pollination periods depend largely on the weather conditions before these periods.

This type of interdisciplinary analysis is essential for a better understanding of the intricate interactions between biosphere, atmosphere and lithosphere. This study is a first step towards a better characterization of the multifaceted nature of the topic, which will help policy makers develop measures to further reduce the impact of pollen on human health.

Declarations of interest

None.

Uncited reference

Adhikari et al. (2006).

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.agrformet.2018.09.023.

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