



Towards a model of wet deposition of bioaerosols: The raindrop size role

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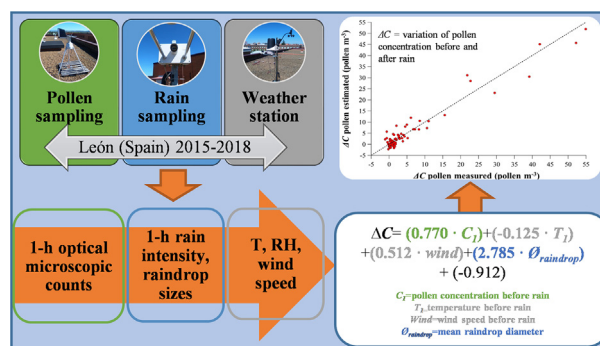
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HIGHLIGHTS

- Hourly variables of rain and 9 types of pollen were measured during 4 years in León.
- An effective scavenging was observed for all types of pollen in 71% of rain events.
- The swept volume was mainly caused by raindrops of sizes between 0.25 and 3 mm.
- Using meteorological and pollen variables, a linear model predicts pollen after rain.

GRAPHICAL ABSTRACT



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ABSTRACT

Bioaerosols play a major role in the plant life of ecosystems. In addition, they have a profound impact on human health, since they may cause lung diseases or allergies. The key objective of this study is to assess the below cloud scavenging effect of rainfall on pollen concentration. The analysis is based on a sampling carried out in León, Spain, between 2015 and 2018. The rainfall variables and the pollen concentrations have been obtained with a disdrometer and a volumetric Hirst type spore-trap, respectively. In order to evaluate the scavenging, three parameters have been calculated: scavenging efficiency (through the concentration-weighted average (% ΔC)), the scavenging coefficient (λ) and the percentage of events with a decrease in pollen concentration (%ES) also called events with effective scavenging. 71% of rain events presented an effective scavenging that affected all types of pollen. The % ΔC mean value of total pollen was $24 \pm 18\%$ (positive values indicate an effective scavenging) and the types of pollen with the highest values were *Castanea* and *Cupressaceae* (71 and 40%, respectively). A linear model ($R^2 = 0.94$) to estimate the pollen concentration after rain was built with variables such as pollen concentration before rain and other variables from a weather station and a disdrometer. Furthermore, we have shown the possibility of knowing in real time the probable *Cupressaceae* pollen concentration, from the initial pollen concentration and the physical parameters of rain (such as raindrop size, rain intensity or volume swept by raindrops in their falling path).

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Abbreviations: %ES, percentage of events with effective scavenging; % ΔC , scavenging efficiency with the concentration-weighted average; % ΔC_{rel} , scavenging efficiency; λ , scavenging coefficient; BCS, Below-Cloud Scavenging; D_p , raindrop diameter; ICS, In Cloud Scavenging; LPM, Laser Precipitation Monitor; PM, Particulate Matter; $S(D_p)$, raindrop section; S_v , swept volume; $U_t(D_p)$, terminal velocity of raindrops; $\Delta C = C_1 - C_2$, variation of the total pollen concentration.

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1. Introduction

Poor air quality causes a negative impact on human health (Apte et al., 2015; HEI Review Panel, 2013; WHO, 2013) and on the environment (Fröhlich-Nowoisky et al., 2016; Menon et al., 2002; Pöschl, 2005). Air pollutants include aerosols or particulate matter (PM), that may be bioaerosols (pollen, fungal spore, viruses or bacteria) and non-biological particles (Oduber et al., 2019). Focusing on bioaerosols, and more concretely on pollen, it has to be emphasized that its concentration causes a clear impact on human health, such as lung diseases or allergies. It should be borne in mind that, in the past four decades an increase in pollen allergies has been registered, and it is one of the main health problems that concerns population in urban and industrialized areas (D'Amato et al., 2002, 2010). In Spain, pollen allergies concern about 15% of the total population and 30% of the young population (Caillaud et al., 2014; Subiza Garrido-Lestache, 2004), being a topic of study in recent years (Aguilera and Ruiz-Valenzuela, 2019; Recio et al., 2018). Furthermore, air pollutants can act as adjuvants and alter the immunogenicity of allergenic proteins of pollen while climate change affects the atmospheric abundance of bioaerosols and aeroallergens (Reinmuth-Selzle et al., 2017) and hence, the human exposure to them.

Wet deposition is the main process to remove bioaerosol from the atmosphere (Fröhlich-Nowoisky et al., 2016; Textor et al., 2006). Depending on the height of aerosol particles in the atmosphere, there are two types of processes: In-Cloud Scavenging (ICS), when aerosol particles serve as nuclei for cloud droplets or ice crystals, and Below-Cloud Scavenging (BCS), when aerosol particles are collected by falling raindrops (it depends on the collection efficiency between a falling raindrop and aerosol particles). In this work, we will focus on BCS suffered by nine types of pollen.

The processes that produce the collection of aerosol particles by raindrops are various (Brownian diffusion, interception, inertial impaction, etc.) and they have been previously discussed (Chate, 2005; Zhao and Zheng, 2006). These processes are similar to fog scavenging (Collett et al., 2001, 2008; Gilardoni et al., 2014), since rain and fog are a set of water drops with different sizes and velocities. Inasmuch as pollen particle diameters range between 10 and 100 μm , the process of inertial impaction (prevailing for particles with size $d_p > 1 \mu\text{m}$) is the most important one (Rasch et al., 2000). It should be noted that BCS of pollen could depend on several factors such as pollen concentration, composition and presence of electric charges (Ladino et al., 2011) or raindrop size distribution and rainfall intensity (Blanco-Alegre et al., 2018; Zikova and Zdimal, 2016).

The influence of rainfall on the pollination process is clear (Lawson and Rands, 2019) and climate change drives alterations in the plant-pollinator interaction (Byers, 2017). Consequently, the analysis of the main bioaerosol particle sinks, including rainfall, has become a key objective in recent research. Several studies have related the rainfall seasonality with pollen concentration (El-Moslimany, 2019; Jantz et al., 2013) and other works have found negative correlations between aeroallergens/pollen and rainfall (Fernández-González et al., 2019; Oduber et al., 2019).

Concerning the impact on human health, Idrose et al. (2019) noted that there is no evidence of interaction between rainfall and total pollen counts in the cases of asthma admissions. Nevertheless D'Amato et al. (2016) reported that during the first 30 min of a thunderstorm, patients suffering from pollen allergies may inhale a high concentration of the allergenic material dispersed into the atmosphere, which in turn can induce severe asthmatic reactions. This fact may be caused by the increase in air humidity and the change of pressure which facilitates the rupturing of pollen grains and the release of allergen bearing respirable particles (Kluska et al., 2020). Other studies have revealed that the interaction between air pollutants and pollen grains may aggravate allergic disorders, since air pollutants adhered to pollen grains facilitate the release of allergens. Thus, the pollen grains are not directly

responsible for allergic disorders inasmuch as their size ($>10 \mu\text{m}$) constrains their penetration into the respiratory tract (Bartra Tomás et al., 2007; D'Amato et al., 2001, 2007, 2010; Sedghy et al., 2018).

In recent years the modeling of the elimination of different biological agents has been the subject of many studies through the use of local conditions and the execution of onsite measurements (Fenu and Mallocci, 2020; Mirra et al., 2020; Oduber et al., 2021a, 2021b; Wang et al., 2011; Zamani Beidokhti et al., 2019). Although the studies on wet removal of ultrafine and fine aerosol particles and other pollutants are common (Blanco-Alegre et al., 2018, 2019; Chate and Pranesha, 2004; Laakso et al., 2003; Maria and Russell, 2005), the information related to the scavenging of pollen by rain is scarce. Huffman et al. (2013) and Dong et al. (2019) indicated that bioaerosols have been detected in fog, rain and snow, and they may influence the water cycle (acting as cloud condensation and ice nuclei). The dry deposition of pollen and spores was predominant (87%) in the study carried out by Dong et al. (2019) while wet deposition, including BCS, caused the remaining 13%. That study suggested that rain scavenging may be independent of the taxon studied; however, Kluska et al. (2020) observed differences between taxa in the washing by rain.

Therefore, the main aim of this study is the evaluation of below cloud scavenging caused by rain on nine types of airborne pollen in an urban background station. In addition, we have estimated the efficiency of BCS as a function of several rain parameters, including raindrop size. The novelty of this study lies in the BCS evaluation on airborne pollen with a time resolution of one hour, and the study of the wash-off caused by the different raindrop diameters, a type of information scarcely found in the scientific literature. This innovative approach may allow the improvement of pollen forecast in the Mediterranean area, a region with serious concerns related to pollen allergies.

The paper is structured as follows: i) Section 2 (material and methods) includes a description of the sampling site, instruments and methodology; ii) in Section 3 (results), a study of rain scavenging effect on different types of pollen has been conducted to propose a model to estimate the change of pollen concentration during rainfall events taking into account the different raindrop sizes; iii) Section 4 (conclusions) includes the main outcomes from the research together with the major impacts.

2. Material and methods

2.1. Sampling site

The sampling campaign was conducted between January 2015 and December 2018 in León, Spain, a city located northwest of the Iberian Peninsula (838 m above sea level) with a population of about 200,000 residents in 2018. León has a Mediterranean climate with continental features, but it is tempered by the Cantabrian range next to the area. The instruments were located at about 15 m above the ground, on the terrace of the Faculty of Veterinary Sciences at the University of León, on the north-eastern border of the city (Fig. A1). In the study area, the major sources of aerosol particles are traffic, domestic heating devices in winter, and wildfires and Saharan intrusions in summer (Oduber et al., 2021a, 2021b). With regard to bioaerosols, the main source area is the Cantabrian range (about 30 km from the city northbound). The main vegetation characteristics of this area are: i) forest with different types of vegetation, whose pollination contributes to a high pollen concentration; ii) several deciduous forests, like poplars (*Populus L.*) or ash trees (*Fraxinus Tourn. ex L.*) are present in valleys and as ornamental flora; iii) vegetation like grasses (Poaceae Barnhart) and other herbaceous plants are also found in grasslands and meadows (Del Río González, 2005). The vegetation of green areas near the sampling point (university campus and parks) is mainly characterized by trees of the families Cupressaceae and Pinaceae, *Platanus hispanica*, *Aesculus hippocastanum*, *Fraxinus excelsior* and *Fraxinus angustifolia*, *Betula pubescens*, *Acer negundo*, *Tilia platyphylus*, as well as other trees with

entomophilous pollination. The lawns are basically constituted by grasses and *Plantago* sp., which are often cut before flowering.

2.2. Pollen concentration characterization

The atmospheric pollen concentration was measured using a volumetric Hirst type spore-trap (Lanzoni VPPS 2000) (Hirst, 1952). The hourly counts by optical microscope for the visual detection of pollen and spores were carried out following the recommendations of the Spanish Aerobiological Network, fulfilling the European Standard: Ambient air - Sampling and analysis of airborne pollen grains and fungal spores for networks related to allergy - Volumetric Hirst method (2018 CEN FprEN 16,868:2018 E). The pollen measurement uncertainty was about 10%, due to the determination of flow rate of Hirst type pollen trap (Oteros et al., 2017; UNI 11108:2004, 2004). The analyses were done using a 40× optical microscope (400 magnifications). Four longitudinal bands of a Melinex tape of 48 mm × 12 mm area were analyzed. The analysis according to the microscope model assumes 12% of that surface, higher than the 10% established by the standard.

The pollen identification included 54 types:

- Families: Apiaceae, Asteraceae, Brassicaceae, Campanulaceae, Cupressaceae, Cyperaceae, Chenopodiaceae, Ericaceae, Fabaceae, Juncaceae, Lamiaceae, Myrtaceae, Poaceae, Polygonaceae, Oleaceae, Rosaceae, Scrophulariaceae, Urticaceae.
- Genera: *Acer*, *Aesculus*, *Ailanthus*, *Alnus*, *Artemisia*, *Betula*, *Carpinus*, *Castanea*, *Cedrus*, *Corylus*, *Echium*, *Fagus*, *Fraxinus*, *Galium*, *Helianthus*, *Juglans*, *Ligustrum*, *Mercurialis*, *Morus*, *Olea*, *Papaver*, *Pinus*, *Plantago*, *Platanus*, *Populus*, *Quercus*, *Rumex*, *Salix*, *Sambucus*, *Sedum*, *Taraxacum*, *Tilia*, *Ulmus*, *Typha*.
- Species: *Quercus pyrenaica* and *Quercus rotundifolia*.

We selected the following types of pollen to analyze: *Betula* (18 to 28 μm), *Castanea* (10 to 16 μm), Cupressaceae (20 to 30 μm), Oleaceae (20 to 32 μm), *Pinus* (45 to 100 μm), *Plantago* (19 to 35 μm), Poaceae (20 to 45 μm), *Quercus* (25 to 30 μm) and Urticaceae (12 to 18 μm) (Trigo Pérez et al., 2008). The mean hourly pollen concentrations were expressed as pollen grains per cubic meter of air. These types of pollen, of different allergenic potential, have been selected for several reasons: shape, ornamentation of the exine and dimensions (Trigo Pérez et al., 2008), since these parameters can modify the degree of hydration of the pollen and/or the permanence of water on the surface of the pollen and, consequently, change its weight. The period of pollination of the vegetation was also considered, so that the different seasons are represented.

2.3. Rain characterization and meteorological data

In order to characterize raindrops, a disdrometer Laser Precipitation Monitor (LPM) of Thies Clima has been used to detect drops between 0.125 and 8 mm in 22 channels, on a one-minute basis, during all the campaign. The LPM was placed beside the Hirst type spore-trap. The LPM has been accurately described by Fernández-Raga et al. (2009). Several rainfall variables have been calculated on a one-hour basis (like the pollen concentration data): precipitation intensity, total accumulated precipitation, number of drops by channels, drop concentration, total drop section (cross-sectional area), volume swept by falling drops by channels, and mean and standard deviation of raindrop sizes. The basic meteorological variables (temperature, wind speed and direction and relative humidity) have been measured through a weather station placed next to the disdrometer, on a one-minute basis.

The volume $S_V(D_p)$ (in $\text{mm}^3 \text{m}^{-3}$) swept by a raindrop of diameter D_p is defined in Eq. (1), where $S(D_p)$ is the section of the raindrop, $U_t(D_p)$ is the terminal velocity of raindrops, calculated by Fernández-Raga et al. (2009), based on studies of Gunn and Kinzer (1949) and t is the sampling time (one minute in this campaign).

$$S_V(D_p) = S(D_p) \cdot U_t(D_p) \cdot t \quad (1)$$

2.4. Rain events selection

The rain event selection has been carried out in four steps:

- 1) Data elimination when the event data are not complete, due to temporary errors in the data collection of rain or pollen.
- 2) Rain criteria: events were only considered when
 - i. an accumulated precipitation higher than 0.1 mm was registered in one hour, so the precipitation intensity in the event (total rain h^{-1}) exceeded 0.1 mm h^{-1} . This criterion is more restrictive than the ones found in other studies about scavenging of aerosol particles (Laakso et al., 2003);
 - ii. there was a minimum of 2 rain-free hours between events (Blanco-Alegre et al., 2019). This interval was selected to eliminate the errors due to overlapping of samples to be examined under the optical microscope.
- 3) Meteorological conditions could affect the scavenging process (Hussein et al., 2006). Thus, to elude changes in pollen concentration caused by meteorological factors beyond precipitation, we only consider those events with variations in temperature and wind speed below $\pm 3^\circ\text{C}$ and $\pm 2 \text{ m s}^{-1}$, respectively, between 2 h before and after rain. Similar meteorological conditions have been used by other authors to avoid the influence of meteorology on rain scavenging (Blanco-Alegre et al., 2019; Kyrö et al., 2009; Zhao et al., 2015).
- 4) The selected events must present a pollen concentration higher than 1 pollen m^{-3} in the air for 5 days in a row before rain. This indicates that pollination has begun and the presence of pollen is not due to distant transport (Jato et al., 2006).

The selected events were classified by rain intensity: low ($<1 \text{ mm h}^{-1}$), medium ($1\text{--}5 \text{ mm h}^{-1}$) and high ($>5 \text{ mm h}^{-1}$). Besides, we classified the selected events by total rainfall recorded: lower than 1 mm, between 1 and 5 mm, between 5 and 20 mm and higher than 20 mm.

In keeping with these criteria, a total of 122 events have been retained to analyze during the sampling campaign.

2.5. Scavenging efficiency and coefficient

Once the rain events had been selected, the pollen concentration was analyzed 2 h before rain (t_1), during rain, and 2 h after rain (t_2). To evaluate the BCS we have calculated 2 parameters for each event: the scavenging efficiency ($\%\Delta C_{rel}$) and the scavenging coefficient (λ). $\%\Delta C_{rel}$ was determined by Eq. (2), to evaluate the change in pollen concentration in the interval between t_1 and t_2 with concentrations C_1 and C_2 . It should be noted that the minus sign has been introduced in order to get a positive value of $\%\Delta C_{rel}$ with effective scavenging (when pollen concentration decreases). To eliminate the influence of daily pattern, the concentration of each type of pollen was corrected, using the approach previously proposed by Blanco-Alegre et al. (2018).

$$\%\Delta C_{rel} = -\left(\frac{C_2 - C_1}{C_1}\right) \cdot 100 \quad (2)$$

The λ coefficient, defined as a rate of aerosols washout by precipitation, was calculated following the criteria proposed by Seinfeld and Pandis (2016) through Eq. (3), the same used for the concentration change C of aerosol particles (Chate et al., 2003; Laakso et al., 2003).

$$\lambda = -\frac{1}{C} \cdot \frac{dC}{dt} \quad (3)$$

From the semi-empirical approach described by other authors (Laakso et al., 2003; Mircea and Stefan, 1998), Eq. (3) was integrated

between the instants t_1 and t_2 with concentrations C_1 and C_2 , obtaining Eq. (4). As with $\% \Delta C_{rel}$, positive values of λ are indicative of effective scavenging.

$$\lambda = -\frac{1}{t_2 - t_1} \cdot \ln\left(\frac{C_2}{C_1}\right) \quad (4)$$

Besides, for each type of pollen, we calculated two additional global parameters: the percentage of events with effective scavenging (%ES) and the scavenging efficiency with the concentration-weighted average (% ΔC), calculated from Eq. (5) for all events of each type of pollen.

$$\% \Delta C = \frac{\sum (\% \Delta C_{rel} \cdot C_1)}{\sum C_1} \quad (5)$$

Subsequently, a statistical analysis with the Kruskal-Wallis (Kruskal and Wallis, 1952) one-way analysis of variance test (at a significance level of 5%) was conducted as a non-parametric method to test $\% \Delta C$ values between pollen types and rainfall groups, and a post-hoc analysis was also conducted to observe between which pairs of rainfall groups there were statistically significant differences.

2.6. Linear model

The variation of pollen concentration during rain will be model through the application of an automatic stepwise linear model (IBM SPSS) with an entry probability of 0.05 and a removal probability of 0.10. The methodology used involved the construction of a model from a random sample, which consisted of 75% of total data. The model was finally applied to the remaining 25% of data. Subsequently, the Kolmogorov-Smirnov goodness of fit test was conducted to check how well the model fits the measured data.

3. Results and discussion

3.1. Meteorological and precipitation analysis

Based on the data obtained from the four years of the sampling campaign, between January 2015 and December 2018 a total of 472 precipitation-days were registered, with 236 possible events to analyze, of which only 122 complied with the selection criteria described in Section 2.4 for total pollen concentration. In order to discard extreme events, we only have evaluated the events included between percentiles P_5 and P_{95} .

The rain events presented a mean duration of 214 min (median: 120 min), a mean rain accumulated of 3.58 mm (median: 0.73 mm) and a mean rainfall intensity of 0.87 mm h⁻¹ (median: 0.37 mm h⁻¹). It is noteworthy that the types of pollen had different numbers of events analyzed, since there were events with and without concentration of some types of pollen. A summary of rain characteristics for each type of pollen is shown in Table 1.

During these events, the mean temperature was 14.0 °C, with a maximum of 34.2 °C and a minimum of -3.2 °C. The wind speed ranged between 0.0 and 5.6 m s⁻¹ (mean value of 1.6 m s⁻¹) but without variations higher than 2 m s⁻¹ between before and after rain. Thus, the meteorological influence in the scavenging process during rain has been purged with the restrictive selection criteria carried out.

For *Castanea*, whose pollen is typical in summer (the driest season), only five rain events were considered. For Cupressaceae and Poaceae, with typical pollens of autumn-winter and spring-early summer (periods with high precipitation), the highest numbers of events (47 and 49, respectively) were registered. The mean raindrop sizes in the low rain intensity group ranged between 0.38 ± 0.20 mm (Cupressaceae) and 0.48 ± 0.30 mm (Urticaceae), in the medium rain intensity between 0.38 ± 0.23 mm (Oleaceae) and 0.62 ± 0.38 mm (*Plantago*) and in the high rain intensity between 0.30 ± 0.26 mm (Cupressaceae)

Table 1

Mean values of: number of drops, drop concentration, accumulated rain, rain intensity, swept volume (S_v) and mean and standard deviation of raindrop size for each type of pollen and for total pollen concentration classified according to rain intensity groups.

Type of pollen (N)	Rain intensity group (mm h ⁻¹)	Number of drops (# m ⁻²)	Drop concentration (# m ⁻³)	Accumulated rain (mm)	Rain intensity (mm h ⁻¹)	S_v (mm ³ m ⁻³)	Raindrop size (mm)
Total pollen (122)	< 1	4.13 10 ⁶	9.41 10 ⁶	1.85	0.38	2.99 10 ⁹	0.42 ± 0.25
	1–5	7.19 10 ⁶	1.22 10 ⁷	5.85	1.55	6.70 10 ⁹	0.45 ± 0.28
	> 5	3.50 10 ⁶	3.88 10 ⁷	34.10	9.09	2.35 10 ¹⁰	0.36 ± 0.28
<i>Betula</i> (11)	< 1	6.21 10 ⁵	9.77 10 ⁵	0.49	0.29	6.03 10 ⁸	0.41 ± 0.19
	1–5	3.13 10 ⁵	3.14 10 ⁷	6.84	1.37	9.92 10 ⁹	0.37 ± 0.22
	> 5	1.55 10 ⁷	1.96 10 ⁵	5.81	5.81	4.73 10 ⁹	0.34 ± 0.23
<i>Castanea</i> (5)	< 1	1.51 10 ³	1.65 10 ⁵	0.36	0.26	1.47 10 ⁸	0.50 ± 0.38
	1–5	–	–	–	–	–	–
	> 5	–	–	–	–	–	–
Cupressaceae (47)	< 1	6.85 10 ⁶	1.20 10 ⁷	2.26	0.37	3.91 10 ⁹	0.38 ± 0.20
	1–5	8.01 10 ⁶	8.55 10 ⁶	4.79	1.84	5.86 10 ⁹	0.41 ± 0.25
	> 5	4.63 10 ⁵	3.76 10 ⁷	16.12	5.38	1.38 10 ¹⁰	0.30 ± 0.26
Oleaceae (24)	< 1	1.11 10 ⁷	5.75 10 ⁶	1.52	0.33	2.73 10 ⁹	0.40 ± 0.23
	1–5	3.00 10 ⁷	7.76 10 ⁶	10.51	1.26	1.24 10 ¹⁰	0.38 ± 0.23
	> 5	–	–	–	–	–	–
<i>Pinus</i> (31)	< 1	1.25 10 ⁶	3.66 10 ⁶	1.16	0.37	1.54 10 ⁹	0.40 ± 0.21
	1–5	5.62 10 ⁶	7.98 10 ⁶	5.37	1.08	5.39 10 ⁹	0.39 ± 0.22
	> 5	1.55 10 ⁷	1.96 10 ⁵	5.81	5.81	4.73 10 ⁹	0.34 ± 0.23
<i>Plantago</i> (39)	< 1	9.55 10 ⁵	8.11 10 ⁶	1.26	0.36	1.86 10 ⁹	0.48 ± 0.32
	1–5	1.78 10 ⁵	1.01 10 ⁷	4.72	1.83	3.83 10 ⁹	0.62 ± 0.38
	> 5	4.18 10 ⁶	2.47 10 ⁷	11.26	6.89	9.10 10 ⁹	0.33 ± 0.25
Poaceae (49)	< 1	7.19 10 ⁶	4.03 10 ⁶	1.36	0.36	2.14 10 ⁹	0.48 ± 0.31
	1–5	8.09 10 ⁴	7.06 10 ⁶	2.88	1.15	2.67 10 ⁹	0.53 ± 0.29
	> 5	4.18 10 ⁶	2.47 10 ⁷	11.26	6.89	9.10 10 ⁹	0.33 ± 0.25
<i>Quercus</i> (33)	< 1	1.27 10 ⁶	1.22 10 ⁷	1.96	0.40	3.03 10 ⁹	0.47 ± 0.29
	1–5	8.09 10 ⁴	7.21 10 ⁶	2.95	1.23	2.84 10 ⁹	0.49 ± 0.28
	> 5	7.90 10 ⁶	1.20 10 ⁷	7.71	7.71	6.06 10 ⁹	0.33 ± 0.24
Urticaceae (24)	< 1	1.21 10 ⁶	1.08 10 ⁷	1.39	0.37	2.28 10 ⁹	0.48 ± 0.30
	1–5	2.35 10 ⁵	2.02 10 ⁷	6.74	2.34	6.82 10 ⁹	0.40 ± 0.26
	> 5	–	–	–	–	–	–

N is the number of events for each type of pollen.

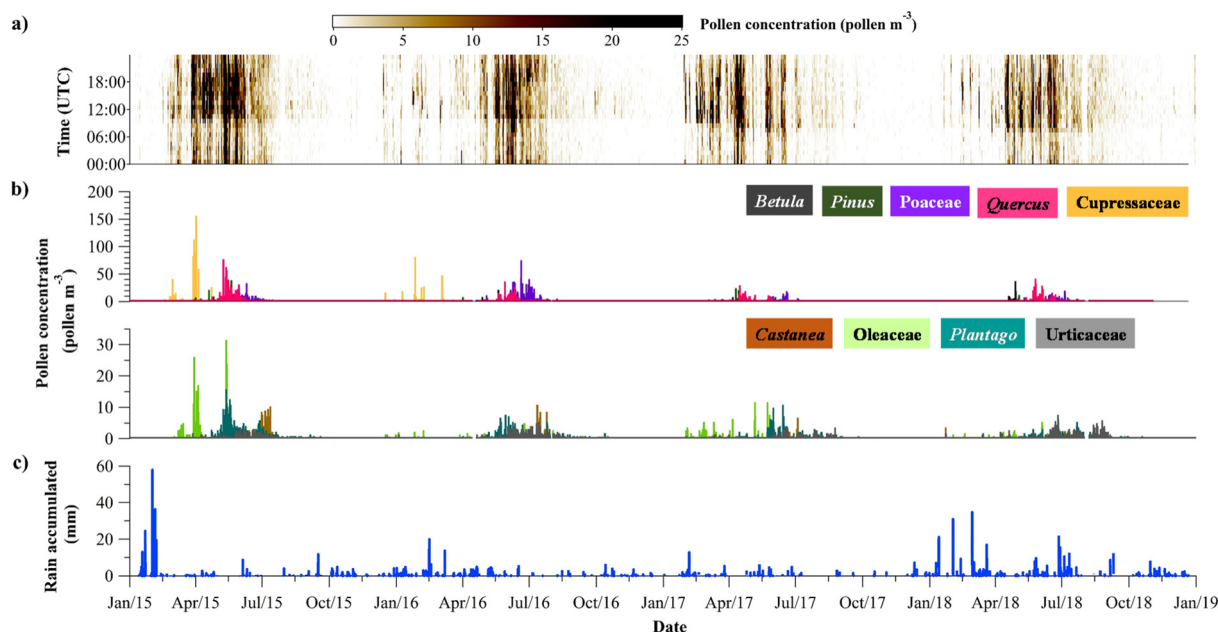


Fig. 1. a) Total pollen concentration registered each hour; b) daily concentration of different types of pollen and c) distribution of rain accumulated during the sampling campaign.

and 0.34 ± 0.23 mm (*Betula*). There were no statistical differences in raindrop size between the events registered for the different types of pollen ($p = 0.170$), so the results between types of pollen are comparable. With respect to the accumulated rain and rainfall intensity, the averages were similar between the events corresponding to the different types of pollen and rain intensity groups.

3.2. Pollen concentration analysis

The evolution of pollen concentration of the different types analyzed is depicted in Fig. 1 along with the precipitation. During all the sampling campaign, the highest pollen concentrations were reached between March and August. A detailed description about the main pollen seasons by types can be found in Oduber et al. (2019). The maximum pollen concentration was observed on 7 March 2017, with 211.5 pollen m^{-3} . By types, the highest daily concentrations were reached by Cupressaceae (7 March 2017 with 205.7 pollen m^{-3} and 31 March 2015 with 156.2 pollen m^{-3}), *Quercus* (7 May 2017 with 77.4 pollen m^{-3}) and Poaceae (18 June 2016 with 75.6 pollen m^{-3}). The highest pollen concentrations occurred on days without rain, with a statistically significant negative correlation ($r = -0.068$; $p = 0.01$) among pollen concentration and daily precipitation.

3.3. Study of the scavenging effect

Airborne pollen scavenging has been analyzed by means of different parameters, as indicated in Section 2.5.

3.3.1. Scavenging efficiency

The $\% \Delta C$ values have been calculated for each pollen type (Table 2). Globally, there was a clear effective BCS for all pollen types. The $\% \Delta C$ mean value for total pollen was $24 \pm 18\%$ and the types of pollen with the highest values were Cupressaceae and *Castanea* (71 and 40%, respectively). On the other hand, the types with lower scavenging were *Betula*, Oleaceae, Poaceae, *Quercus* and Urticaceae (ranging between 14 and 16%). Dong et al. (2019), in a study conducted in Seoul (South Korea), examined plant assemblages collected in atmospheric deposits to evaluate dry and wet deposition. They reported that a small fraction of bioaerosols (~15%) precipitated via ICS or BCS. Only a small number of plant genera presented a higher wet deposition (e.g. *Quercus*), probably due to their role in precipitation, acting as nucleation species in the atmosphere. Also, they said that the different plant taxa did not vary significantly between dry and wet deposition. Thus, contrary to suggested by Dong et al. (2019), our results show clearly differences in the washing parameters depending on the type of pollen, a behaviour previously found by Kluska et al. (2020). The $\% \Delta C$ values obtained were similar to those estimated for aerosols in the coarse mode (19%) in León from July 2015 to January 2016 (Blanco-Alegre et al., 2018) and lower than values (70%) based on models in July 1997 (Loosmore and Cederwall, 2004). It should be borne in mind that in the coarse mode (pollen sizes included), inertial impaction is the most important process (Rasch et al., 2000), while in ultrafine and fine modes, Brownian diffusion and interception mechanisms prevail (Chate, 2005).

Table 2

Mean, P_5 and P_{95} percentiles and quartile 2 values of $\% \Delta C_{rel}$ (scavenging efficiency), effective scavenging ($\% ES$) and mean and standard deviation of $\% \Delta C$ (scavenging efficiency using the concentration-weighted average) obtained for each type of pollen and for total pollen concentration.

	Total pollen	<i>Betula</i>	<i>Castanea</i>	Cupressaceae	Oleaceae	<i>Pinus</i>	<i>Plantago</i>	Poaceae	<i>Quercus</i>	Urticaceae
N	122	11	5	47	24	31	39	49	33	24
$\% ES$	71	82	60	66	75	55	77	78	73	63
P_5	-400	-73	0	-486	-50	-400	-105	-93	-100	-134
Q_2	50	100	100	86	100	-67	100	100	75	42
P_{95}	100	100	100	100	100	100	100	100	100	100
$\% \Delta C$	24.3	15.1	40.3	70.9	16.7	20.1	24.6	15.3	14.8	14.2
SD	17.9	5.5	4.5	39.0	5.5	15.1	6.3	7.7	7.5	9.4

The inverse relationship between pollen concentration and total rainfall have also been reported by several authors in various climatic regions. Alan et al. (2018), in a study conducted between 2015 and 2016 in Ankara and Zonguldak (Turkey), two cities with different climatic properties, studied the correlations between Poaceae pollen and Phl p 5 allergen. They found different results according to the climatic characteristics and topography of each city. Also, Bruffaerts et al. (2018) examined the evolution of pollen concentrations in Brussels (Belgium) during 40 years and the effect of meteorological changes on pollen concentration. They found that changes in pollen annual cycles are related with changes in the annual cycles of temperature, radiation, humidity and rainfall. It should be noted that Kluska et al. (2020) reported this inverse relationship but only with rainfall intensities higher than 5 mm h^{-1} in Poland, but this threshold may vary by climatic zones.

Another important parameter to analyze the pollen scavenging is %ES, in order to quantify the sensitivity of each pollen type to rain scavenging. The %ES mean value of total pollen was 71%, so 71% of events presented an effective scavenging during rain, with *Betula* being the type of pollen with the highest %ES value. Huffman et al. (2013), in a study carried out in a montane pine zone in Colorado (USA) between July and August of 2011, found that rain can cause intense bursts of bioaerosol emission during or after rain events, causing an increase of bioaerosol concentration by an order of magnitude. They detected the presence of bioaerosols in rain samples, concluding that atmospheric bioaerosols and rainfall are tightly coupled, at least at midlatitude semi-arid forest ecosystems. The presence of bioaerosols in rain samples may be explained by the effective scavenging on pollen particles. It is noteworthy that pollen types with the highest values of % ΔC (*Castanea* and Cupressaceae) presented the lowest %ES values, together with *Pinus* and Urticaceae. There were statistical differences in % ΔC between the types of pollen ($p = 0.043$). After a post hoc analysis, there were statistical differences between Oleaceae and: Cupressaceae ($p = 0.003$), *Pinus* ($p = 0.007$), *Quercus* ($p = 0.008$) and Urticaceae ($p = 0.008$). The *Castanea* results must be carefully tackled due to the low number of events.

For the study of scavenging depending on rain characteristics, the % ΔC values have been obtained per pollen type and rain intensity groups, as depicted in Fig. 2. The medium ($1\text{--}5 \text{ mm h}^{-1}$) and high rain intensity ($>5 \text{ mm h}^{-1}$) presented the highest % ΔC values (69.7 and 69.2%, respectively) for total pollen concentration. The low rain intensity ($<1 \text{ mm h}^{-1}$) also presented a low value of 40.9%. In addition, %

ES values were higher in medium and high intensities (92.9 and 100%, respectively) than in low intensity (71.4%). Thus, with rain intensities higher than 5 mm h^{-1} , the total pollen concentration always decreased.

In regards to accumulated rain, all groups presented effective scavenging: lower than 1 mm (40.3%), between 1 and 5 mm (58.6%) and between 5 and 20 mm (39.2%). Particularly striking is the high value obtained for events with an accumulated rain higher than 20 mm (99.8%), with a practically total scavenging of airborne pollen. There were no statistical differences in % ΔC between the three groups of rain intensity ($p = 0.243$). On the contrary, there were statistical differences in % ΔC between the groups of accumulated rain lower than 1 mm and higher than 20 mm ($p = 0.029$). Therefore, contrary to reported by Kluska et al. (2020), there is an effective washing of pollen with precipitation intensities below 5 mm h^{-1} . This fact may be related with different spatial characteristics among regions (Rzeszów in Poland and León in Spain), such as altitude (210 vs 838 m) or climate (polar maritime vs Mediterranean climate with continental features) and the different number of pollen types analyzed (four vs nine).

By pollen types, it is worth noting that with low rain intensities, all types presented an effective scavenging, with values ranging between 22.4% (*Betula*) and 79.5% (Cupressaceae). Besides, all %ES values were higher than 70%, showing effective scavenging with rain intensities lower than 1 mm h^{-1} on pollen. However, in medium rain intensity there was divergence according to the pollen type. Only *Betula*, Cupressaceae, *Pinus*, *Quercus* and Urticaceae presented an effective scavenging. Oleaceae, *Plantago* and Poaceae presented negative % ΔC values and low values of %ES. Regarding high rain intensity, all types presented high values of % ΔC and %ES, except *Pinus*, the pollen type with the highest sizes. The *Pinus* pollen behaviour could be explained by its physics characteristics; although it is the largest type due to its two lateral air sacs (like floaters), these do not present cytoplasmic content and do not participate in the hydration of the pollen; they only serve to float better (Trigo Pérez et al., 2008). Therefore, it would not be heavier despite its size and this fact can explain the lower scavenging suffered.

Also, the minor scavenging effect of rain in *Betula* and *Pinus* may be related to their biology of pollination, since these types shed a large amount of pollen during their short pollen season (around one month) while other types like Poaceae, Cupressaceae or Urticaceae present a longer pollen season. Previous studies report that the high pollen concentration during precipitation may be favored by the

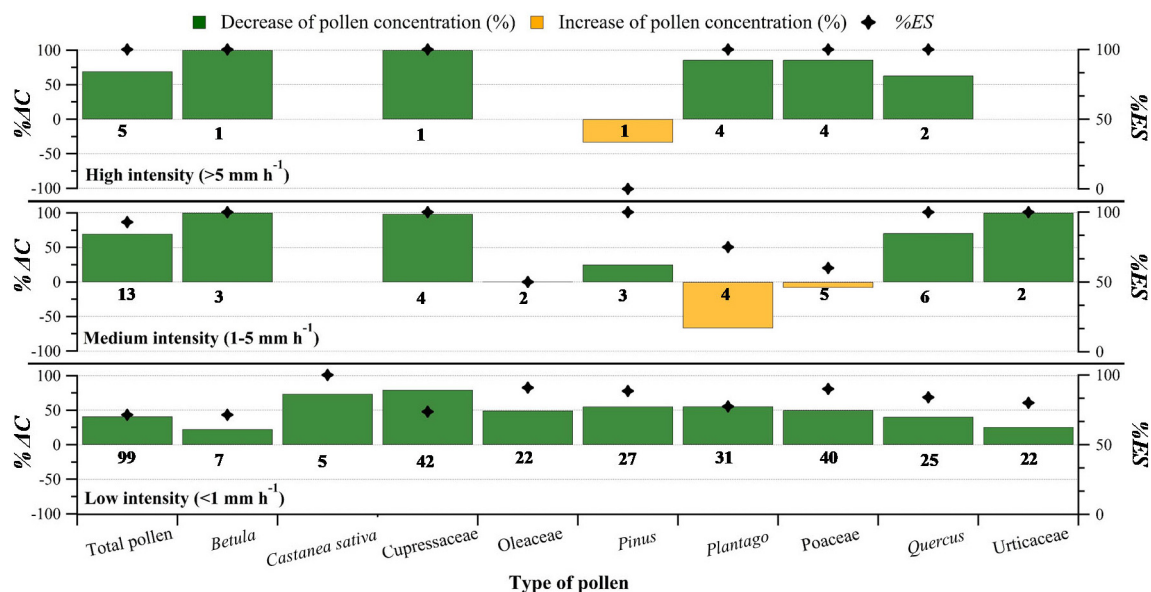


Fig. 2. Scavenging efficiency using the concentration-weighted average (% ΔC) and effective scavenging (%ES) averages obtained for each type of pollen by rain intensity groups. The number indicates the number of events.

Table 3

Mean, standard deviation, median, P_5 and P_{95} values and of scavenging coefficients (λ), for each type of pollen and for total pollen concentration.

λ (s^{-1})	$\bar{x}$	SD	Median	P_5	P_{95}
Total pollen	$2.50 \cdot 10^{-2}$	$5.95 \cdot 10^{-2}$	$5.30 \cdot 10^{-3}$	$-1.46 \cdot 10^{-2}$	$1.76 \cdot 10^{-1}$
<i>Betula</i>	$1.15 \cdot 10^{-1}$	$1.29 \cdot 10^{-1}$	$5.60 \cdot 10^{-2}$	$-8.94 \cdot 10^{-3}$	$3.35 \cdot 10^{-1}$
<i>Castanea</i>	$2.01 \cdot 10^{-1}$	$1.84 \cdot 10^{-1}$	$3.26 \cdot 10^{-1}$	0	$3.44 \cdot 10^{-1}$
Cupressaceae	$5.82 \cdot 10^{-2}$	$9.74 \cdot 10^{-2}$	$1.16 \cdot 10^{-2}$	$-2.34 \cdot 10^{-2}$	$3.21 \cdot 10^{-1}$
Oleaceae	$1.17 \cdot 10^{-1}$	$1.25 \cdot 10^{-1}$	$8.01 \cdot 10^{-2}$	$-3.04 \cdot 10^{-3}$	$3.30 \cdot 10^{-1}$
<i>Pinus</i>	$7.80 \cdot 10^{-2}$	$1.30 \cdot 10^{-1}$	$6.10 \cdot 10^{-3}$	$-1.86 \cdot 10^{-2}$	$3.21 \cdot 10^{-1}$
<i>Plantago</i>	$1.45 \cdot 10^{-1}$	$1.47 \cdot 10^{-1}$	$1.07 \cdot 10^{-1}$	$-1.16 \cdot 10^{-2}$	$3.45 \cdot 10^{-1}$
Poaceae	$1.23 \cdot 10^{-1}$	$1.39 \cdot 10^{-1}$	$5.53 \cdot 10^{-2}$	$-9.38 \cdot 10^{-3}$	$3.27 \cdot 10^{-1}$
<i>Quercus</i>	$9.77 \cdot 10^{-2}$	$1.39 \cdot 10^{-1}$	$2.31 \cdot 10^{-2}$	$-8.66 \cdot 10^{-3}$	$3.34 \cdot 10^{-1}$
Urticaceae	$9.25 \cdot 10^{-2}$	$1.42 \cdot 10^{-1}$	$4.51 \cdot 10^{-3}$	$-1.16 \cdot 10^{-2}$	$3.39 \cdot 10^{-1}$

transport of pollen by thermal currents (Zhao et al., 2014) and by the impaction and condensation effects (Kasprzyk et al., 2001).

3.3.2. Scavenging coefficient

For total pollen concentration, a mean scavenging coefficient (λ) of $2.50 \cdot 10^{-2} s^{-1}$ and a median of $5.30 \cdot 10^{-3} s^{-1}$ have been obtained (Table 3). The mean value is three orders of magnitude higher than λ obtained for fine ($2.6 \cdot 10^{-5} s^{-1}$) and coarse aerosol modes ($5.8 \cdot 10^{-5} s^{-1}$) (Blanco-Alegre et al., 2018) in a sampling campaign carried out at the same place as this study, showing a high scavenging effect on pollen concentration. One of the pollen types with higher values of λ and ΔC was *Castanea*. Conversely, Cupressaceae presented a higher ΔC but a low value of λ and ΔES . This may be due to the characteristics of Cupressaceae rain events: they were long or with high rain intensity when the scavenging was effective. Another important fact is that Cupressaceae has a tiny opening and a very fine exine, with much cytoplasmic content (Trigo Pérez et al., 2008). It takes a long time to activate and its mechanisms are not well known at present. Therefore, it takes time to fill up with water and it takes time to sediment. The rest of pollen types with higher values of λ were *Betula*, Oleaceae, Poaceae and *Plantago*, with values ranged between $1.15 \cdot 10^{-1}$ and $1.45 \cdot 10^{-1} s^{-1}$. There were no statistically significant differences in λ between types of pollen ($p = 0.194$).

By rain intensity groups, λ average was $2.41 \cdot 10^{-2} s^{-1}$ (low intensity), $2.13 \cdot 10^{-2} s^{-1}$ (medium intensity) and $5.12 \cdot 10^{-2} s^{-1}$ (high intensity), so the rain intensity higher than $5 mm h^{-1}$ was the most efficient. Furthermore, the highest values of λ were registered for an accumulated precipitation higher than 20 mm ($2.94 \cdot 10^{-2} s^{-1}$). The rest of accumulated precipitation groups presented these values: lower than 1 mm, $2.32 \cdot 10^{-2} s^{-1}$; between 1 and 5 mm, $2.65 \cdot 10^{-2} s^{-1}$; between 5 and 20 mm, $2.87 \cdot 10^{-2} s^{-1}$ and higher than 20 mm, $2.94 \cdot 10^{-2} s^{-1}$. There were no statistically significant differences in λ between groups of rain intensity ($p = 0.235$) or between groups of accumulated rain ($p = 0.338$).

Table 4

Variables used to build the linear model to estimate the total pollen variation during rain events.

Symbol	Variable	Units
C_1	Pollen concentration before rain	pollen m^{-3}
ws	Wind speed	$m s^{-1}$
T	Temperature	$^{\circ}C$
P	Pressure	hPa
RH	Relative humidity	%
t	Event duration	h
R	Accumulated precipitation	mm
I	Mean rainfall intensity	$mm h^{-1}$
$\emptyset$	Mean raindrop diameter	mm
$S_{v_{swept}}$	Sum of volume swept by falling drops	$mm^3 m^{-3}$
$Ndrops_x$	Number of drops per m^2 in the size channel of x mm	$\# m^{-2}$
$Ndrops_{total}$	Total drops per m^3	$\# m^{-3}$

3.4. Raindrop size effect

Kluska et al. (2020) proposed that the raindrop size parameter need to be considered. Several decades ago, Gatz and Nelson Dingle (1971) suggested that larger raindrops during high rain intensities were less effective to remove pollen than smaller drops during lower intensity rainfall.

In our study, the scavenging effect of the different raindrop sizes has been evaluated through the swept volume (S_v) caused by each range of raindrop sizes on total pollen concentration (Fig. 3). In low and medium rain intensities, the S_v on the total pollen concentration was caused by raindrops of sizes between 0.25 and 3 mm. However, for high rain intensity events, S_v was higher in the raindrop sizes between 0.5 and 4 mm. Besides, raindrops larger than 4 mm caused scavenging only in high intensity events ($>5 mm h^{-1}$). The total S_v in high rain intensity events ($2.4 \cdot 10^{10} mm^3 m^{-3}$) was eight and three times higher than in low ($3.0 \cdot 10^9 mm^3 m^{-3}$) and medium ($6.7 \cdot 10^{10} mm^3 m^{-3}$) rain intensity events. There were statistically significant differences in S_v between the rain intensity groups ($p < 0.001$). Thus, these results could explain the highest values of ΔC and λ in events with a high rain intensity, caused by a higher swept volume.

3.5. Modeling the variation of pollen concentration

In order to estimate the variation of the total pollen concentration ($\Delta C = C_1 - C_2$) during rain events, a linear model has been built. The variables used are shown in Table 4. Although many other studies have related pollen concentration to rainfall (Buters et al., 2015; Ge et al., 2017; Shen et al., 2006; Zhang et al., 2015), here we will focus on the analysis of the likely impact of rain on pollen concentration using the combination of weather and pollen parameters. As indicated in the paragraph 2.6, the methodology process has been repeated ten times and the

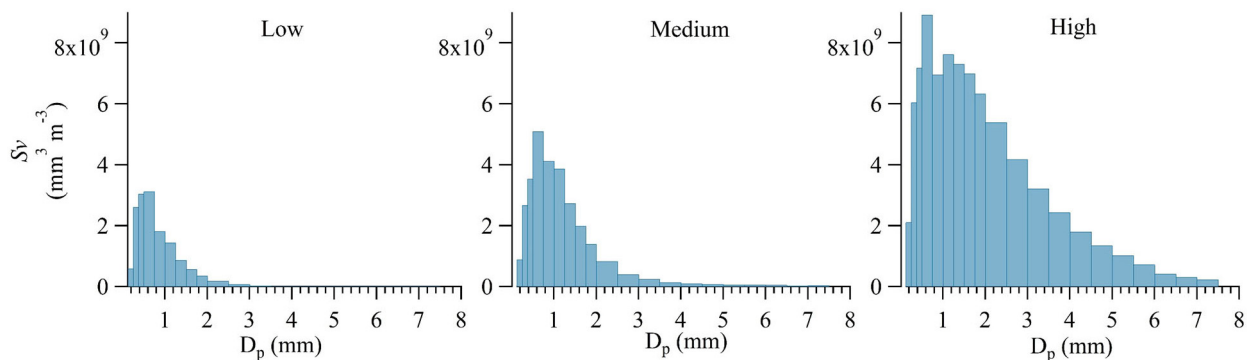


Fig. 3. Mean swept volume (S_v) caused by different raindrop sizes classified by rain intensities (low ($<1 mm h^{-1}$), medium ($1-5 mm h^{-1}$) and high ($>5 mm h^{-1}$)) of rain events considered for total pollen concentration.

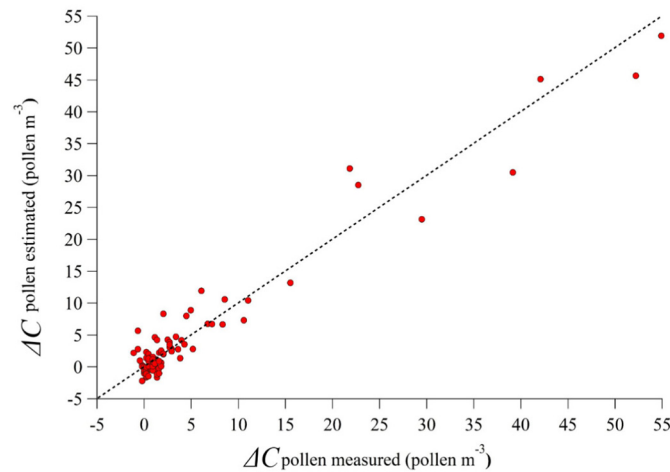


Fig. 4. Variation ΔC of pollen concentration estimated by the model vs variation ΔC of pollen concentration measured after each rain event.

results are shown in Table A1. In all cases significant values ($\alpha > 0.05$) have been obtained, so measured and predicted data are related and, consequently, the model created is consistent. It is noted that in all cases pollen concentration before rain (C_1) and temperature before rain (T_1) were included. Other variables as wind speed (ws) and mean raindrop diameter ($\emptyset$) were not continuous. The inclusion of raindrop diameter confirm the importance of this parameter in the removal of atmospheric pollen, as other authors suggest (Gatz and Nelson Dingle, 1971; Kluska et al., 2020).

Therefore, using the whole data set, a multi-linear regression model has been established ($R^2 = 0.94$), which includes the aforementioned variables. The coefficient values are shown under each term.

$$\Delta C = \underbrace{(k_1 \cdot C_1)}_{0.770 (\pm 0.022)C_1} + \underbrace{(k_2 \cdot T_1)}_{-0.125 (\pm 0.038)T_1} + \underbrace{(k_3 \cdot ws)}_{0.512 (\pm 0.255)ws} + \underbrace{(k_4 \cdot \emptyset)}_{2.785 (\pm 1.169)\emptyset} + \underbrace{k_5}_{-0.912 (\pm 0.333)}$$

The variation of the measured concentration and the results obtained applying the model are similar (Fig. 4); thus, the use of the model is a valuable tool to predict the total pollen concentration after rain.

For each pollen type, the same analysis was performed, in order to obtain individual model variations of each pollen concentration. Nevertheless, the low number of rain events recorded for several types of pollen caused that only one type of pollen presented high values of R^2 : Cupressaceae (0.98). Hereafter, we show the model obtained with 100% of data. The coefficient values for Cupressaceae variation are presented under each term:

$$\Delta C_{\text{Cupressaceae}} = \underbrace{(k_1 \cdot C_1)}_{0.918 (\pm 0.022)C_1} + \underbrace{(k_2 \cdot Ndrops_{0.25})}_{-2.135 \cdot 10^{-5} (\pm 0.038)Ndrops_{0.25}} + \underbrace{(k_3 \cdot Ndrops_{0.5})}_{2.895 \cdot 10^{-5} (\pm 6.0 \cdot 10^{-6})Ndrops_{0.5}} + \underbrace{k_4}_{-0.517 (\pm 0.392)}$$

where C_1 is the Cupressaceae concentration before rain, $Ndrops_{0.25}$ is the number of raindrops between 0.25 and 0.375 mm and $Ndrops_{0.5}$ is the number of raindrops between 0.5 and 0.75 mm. It is worth highlighting that the main scavenging effect was caused by 0.5–0.75 mm size raindrops, the channel with the highest values of swept volume.

A detailed analysis of each type of pollen requires more years of sampling and is one of the next challenges.

4. Conclusions

The evaluation of wet deposition of bioaerosols have been carried out through the study of below cloud scavenging on nine types of pollen under different rainfall conditions during a sampling period of four years in an urban background environment. As a result, a clear effective scavenging in all types of pollen analyzed was observed with a concentration-weighted average ($\% \Delta C$) of $24 \pm 18\%$. The types of pollen with the highest values were *Castanea* and *Cupressaceae* (70.9 and 40.3%, respectively). The rain scavenging for pollen was very different from that of coarse aerosol particles: the mean λ coefficient for pollen scavenging was $2.50 \cdot 10^{-2} \text{ s}^{-1}$ (median of $5.30 \cdot 10^{-3} \text{ s}^{-1}$), three orders of magnitude higher than the value obtained for coarse aerosol in a sampling campaign conducted at the same place. Rain intensity over 5 mm h^{-1} was the most efficient. All pollen types presented high values of $\% \Delta C$ and $\% ES$, except *Pinus*, the largest pollen, probably related with its physics characteristics. This scavenging occurred in the swept volume (S_v) mainly produced by raindrop sizes between 0.25 and 3 mm. The swept volume could explain the highest scavenging with high rain intensities, inasmuch as the average value of S_v in high rain intensity events ($2.4 \cdot 10^{10} \text{ mm}^3 \text{ m}^{-3}$) was eight and three times higher than in low and medium rain intensity events, respectively. Finally, a linear model ($R^2 = 0.94$) was built to estimate the pollen concentration after rain, with input variables from a weather station, a disdrometer and the pollen concentration before rain. Thus, this kind of studies constitutes a valuable tool for the bioaerosol forecast in the Mediterranean area, a region with severe problems with pollen allergies. Future studies will focus on the research of the root causes of the different scavenging efficiency values obtained for the different types of pollen and the mechanism of scavenging on pollen.

CRediT authorship contribution statement

Carlos Blanco-Alegre: Methodology, Formal analysis, Investigation, Writing – original draft, Visualization. **Amaya Castro:** Conceptualization, Investigation, Writing – review & editing, Project administration, Funding acquisition. **Ana I. Calvo:** Conceptualization, Methodology, Writing – review & editing, Project administration, Funding acquisition. **Fernanda Oduber:** Formal analysis, Methodology, Investigation, Writing – review & editing. **Delia Fernández-González:** Conceptualization, Resources, Writing – review & editing. **Rosa María Valencia-Barrera:** Investigation, Writing – review & editing. **Ana María Vega-Maray:** Investigation, Writing – review & editing. **Tibor Molnár:** Investigation, Writing – review & editing. **Roberto Fraile:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision, Project administration, Funding acquisition.

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

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Appendix A: Supplemental file

Towards a model of wet deposition of bioaerosols: the raindrop size role

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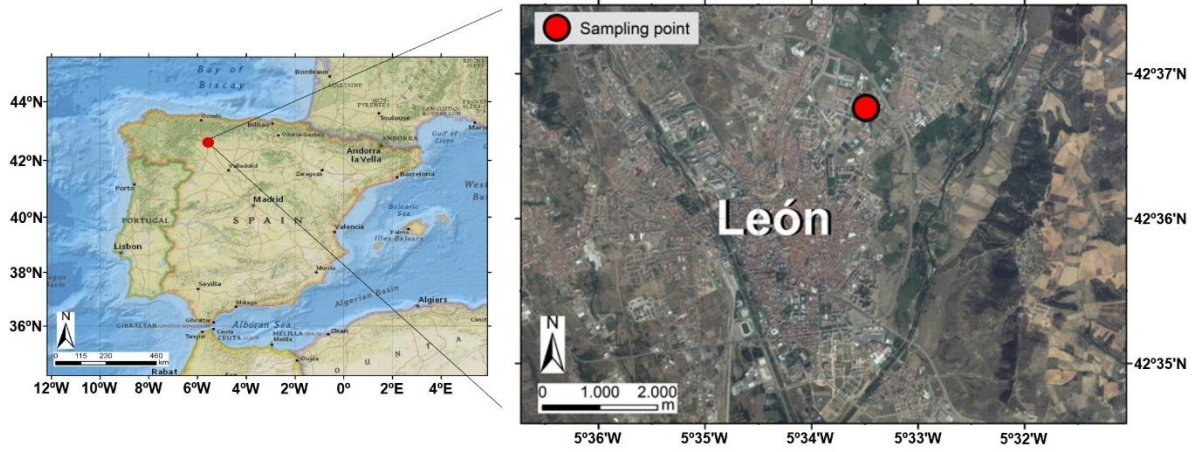


Figure A1. Sampling point and surroundings in the NW Iberian Peninsula. Source: Earthstar Graphics, ESRI.

Table A1. Coefficients obtained to verify the model performed for each of the 10 models, and Kolmogorov-Smirnov (K-S) statistical test carried out between the predicted and the measured values. The last row ($N=0$) includes the model obtained with the whole data set was represented.

N	k_1	k_2 ($\text{m}^{-3} \text{ } ^\circ\text{C}^{-1}$)	k_3 (s m^{-4})	k_4 ($\text{m}^{-3} \text{ mm}^{-1}$)	k_5 (m^{-3})	K-S (statistic)	R^2
1	0.774	-0.101	0.488	-	-0.126	0.429	0.978
2	0.736	-0.121	-	3.019	-0.316	0.353	0.979
3	0.792	-0.139	-	-	0.983	0.360	0.738
4	0.738	-0.123	-	2.988	-0.333	0.261	0.754
5	0.783	-0.121	-	-	1.193	0.545	0.602
6	0.743	-0.138	-	2.985	-0.013	0.259	0.940
7	0.795	-0.127	0.512	-	0.184	0.250	0.926
8	0.790	-0.107	0.599	-	-0.084	0.214	0.968
9	0.736	-0.121	-	3.019	-0.316	0.421	0.962
10	0.776	-0.150	0.556	3.239	-1.084	0.186	0.798
0	0.770	-0.125	0.512	2.785	-0.912	0.165	0.938