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A new method for determining the sources of airborne particles

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ABSTRACT

Air quality is a major issue for humans owing to the fact that the content of particles in the atmosphere has multiple implications for life quality, ecosystem dynamics and environment. Scientists are therefore particularly interested in discovering the origin of airborne particles. A new method has been developed to model the relationship between the emission surface and the total amount of airborne particles at a given distance, employing olive pollen and olive groves as examples. A third-degree polynomial relationship between the air particles at a particular point and the distance from the source was observed, signifying that the nearest area to a point is not that which is most correlated with its air features. This work allows the origin of airborne particles to be discovered and could be implemented in different disciplines related to atmospheric aerosol, thus providing a new approach with which to discover the dynamics of airborne particles.

1. Introduction

Tropospheric air quality is an important issue for humans because it directly affects quality of life. An important aspect of air pollutants are airborne particles of biological origin, such as bacteria, fungal spores and pollen grains. These aerobiological particles have a great effect on human health (Barber et al., 2008; Dales et al., 2010), cultural heritage (Ruga et al., 2008) or the dynamics of terrestrial ecosystems (Belmonte and Vila, 2004; Ellstrand, 1992), among others. This paper has been particularly focused on olive pollen, since it is the main cause of pollen allergies in the Mediterranean area (D'Amato et al., 2007). Olive has a high socio-economic impact in the area (Barranco, 2008) and it is considered to be one of the best biological indicators regarding the effects of weather variations on Mediterranean ecosystems (Orlandi et al., 2005). The olive pollen monitoring is therefore useful to prevent allergy symptoms (Oteros et al., 2013c), to study

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the effects of climate change in the Mediterranean area (Garcia-Mozo et al., 2014) and also to forecast the olive harvest (Oteros et al., 2014).

It is thus particularly interesting to understand the relationship between the emission source and air quality. In Aeropalynology, it is known that the relationship between airborne pollen and vegetation depends on a number of factors, such as the pollen production of individual species, pollen dispersal (dependent on the pollen aerodynamic features and atmospheric conditions) and the spatial distribution of the vegetation around the sampling site (Prentice, 1985). In this respect, the aerobiological properties of an area depend on multiple factors such as vegetation, climate conditions, topography and atmospheric dynamics (Smith et al., 2014).

One of the most frequently posed questions in atmospheric sciences concerns the source of airborne particles, and a comprehensive answer has not yet been found. In fact, many studies have been carried out in order to discover the origin of airborne pollen and to understand the relationship with the focus of emissions. These studies have been carried out by phenology and land use relationship (Estrella et al., 2006; León-Ruiz et al., 2012; Siljamo et al., 2008; Tormo et al., 2011), through theoretical models about back trajectories and particle dispersion, by solving dispersion equations (Alarcón-Jordán et al., 2013; Prentice, 1985; Sadyś et al., 2014; Sofiev et al., 2006; Sugita, 1994) or by combining different approaches (Fernandez-Rodriguez et al., 2014; Hernández-Ceballos et al., 2011, 2014; Hidalgo et al., 2002; Pauling et al., 2012; Skjøth et al., 2010). Although none of these methods have discovered the global relationship that exists between the distance of the emission source and the air particles of an area, they are nevertheless very useful to analyze the origin of particles in specific episodes. Few experimental methods, tested to relate pollen-nearby vegetation, are too tedious to find a global relationship. These are based on pollen monitoring at different distances from a located source, which may be supplemented by genetic studies (Robledo-Arnuncio and Gil, 2005).

The present study is based on the hypothesis that the emission surface around a point is correlated with the total amount of airborne particles. We hypothesize that possible variations in this correlation, at different distances between both elements, are determined by changes in the influence of the emission source on air features. Therefore the main goal has been to develop an experimental method to model the relationship between the surface of the emission source and the total amount of airborne particles at a determined distance, employing olive pollen and olive groves as experimental examples.

2. Methods

2.1. Data source

The present study was performed by using the emission surface data and pollen counts in the air. As study case, the aerobiological particle modeled was olive pollen (*Olea europaea* L.) and the emission modeled was land uses of olive groves.

Airborne pollen data were obtained from the Spanish Aerobiology Network (REA), supported by each of the 19 monitoring stations involved in the study (Fig. 1). All monitoring stations are located in standard conditions, following the Aerobiology Minimum Requirements for ensuring data quality (Galán et al., 2014). It was used data about airborne pollen concentrations (pollen grains/m^3) during the period 2005–2007, due to 2006 was the newest data from land use information in Spain. All data were obtained following the standardized procedure of REA (Galán et al., 2007). Daily pollen concentrations were collected using a Hirst-type volumetric spore-trap, a volumetric suction sampler that is based on an impaction process (Hirst, 1952). This type of device is proposed by the European Aeroallergen Network (EAN), which offer the possibility to obtaining reliable daily and even hourly data (Galán et al., 2014). Air is sucked into the trap at a constant rate of 10 L/min through an orifice (2 mm × 14 mm). Behind the orifice, the air impacts over a rotating drum that moves at 2 mm/h and airborne particles are trapped on a transparent Melinex tape with an adhesive coated. After sampling, the tape is then mounted between a glass slide and a cover slip using a mounting media. Each slide is examined with light microscopy, covering more than 10% of the sampled surface. REA ensures the quality of data by performing frequent quality control programs (Oteros et al., 2013a).

Land use data about olive grove surfaces in Spain during the year 2006 were obtained from the CORINE Land Cover database, as the newest available information (CLC, 2006). Fig. 1 shows the geographical distribution of olive groves in Spain, which is the largest area of olive production in the world (IOC, 2014).

2.2. Concentric Ring Method

The present study was performed by using a new methodology termed “Concentric Ring Method (CRM)”. The information employed regarding airborne particles and the surface of emission source in surrounding areas was, in this study case, based on olive pollen and olive groves. CRM is developed in eleven steps:

1. The Pollen Index (*PI*) is the average amount of annual pollen collected during three consecutive years (1):

$$PI = \frac{\sum DP_{n-1} + \sum DP_n + \sum DP_{n+1}}{3} \tag{1}$$

Where $\sum DP_n$ is the summation of Daily Pollen concentrations (pollen grains/m^3) during the year n, n being the year of emission surface information according to CORINE Land Cover database.

2. The calculation of concentric buffers around each sampling point with the same separation distance. It was calculated 300 buffers around each sampling point with a difference of 1 km radius.
3. The calculation of the sum of Actual Emission Surface (*AES*) included in each ring, where ring is the surface located between two consecutive buffers. That means the summation of the emission area contained on each ring.
4. To calculate Pearson correlation between PI and AES in each ring, as quantitative data. I.e. we took the AES of the determined ring of all the 19 sites and correlate them with the PI of all the 19 sites. And so on for all the 300 rings. We hypothesize that the amount of olive grove that exists around each sampling point will be related to the *PI* on each sampling point. And that changes in the correlation between the *PI* and the surface of olive groves on each ring (*AES*), as longer distances are tested, they are related to changes in the influence of the emission surface on the *PI.*
5. The calculation of the mathematical relationship between correlation coefficients and the ring distance from the station (external buffering radius), expressed as x, by means of polynomial interpolation. The polynomial ($f(x)$) is calculated until the cut with x axis or the relative minimum after the inflection point.

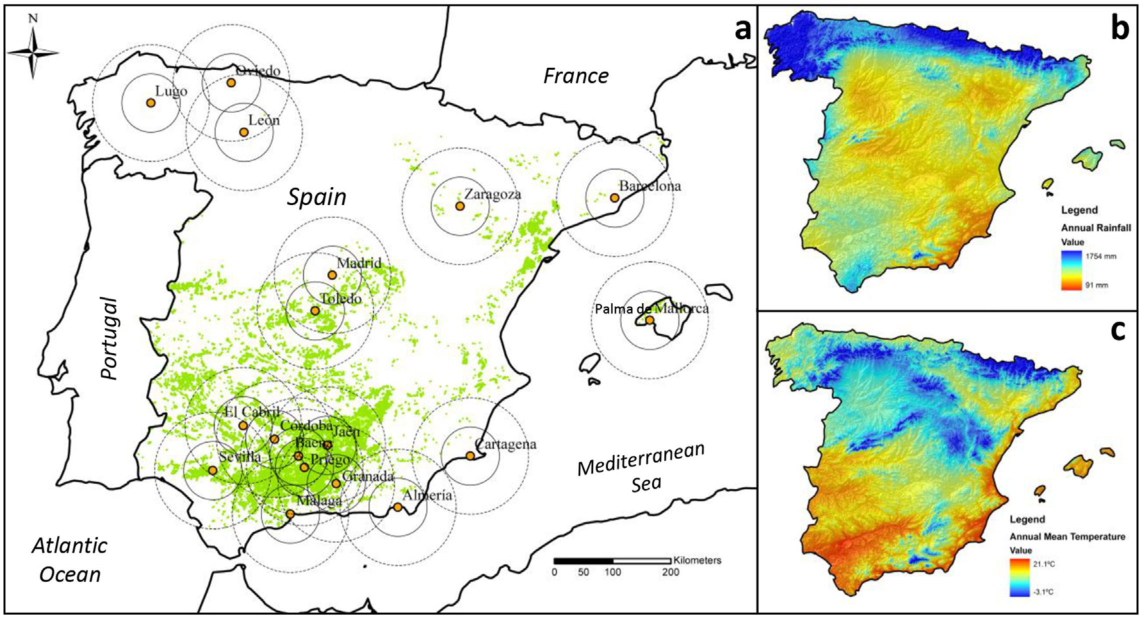

Fig. 1. Study area. (a) shows pollen sampling points location and olive groves surface, smooth circles show buffers of 50 km radius around each sampling point and dashed circle shows buffers of 100 km radius, as reference distances. (b) shows averaged yearly rainfall in study area (1950–2000). (c) shows annual mean temperature in study area (1950–2000) (Hijmans et al., 2004).

6. In order to discover the Theoretical Influence (*TI*) that the emission surface has on *PI*, it is necessary calculating the area under the curve of the polynomial (*f(x)*). *TI* summarizes the influence that the emission surface of each ring has on *PI*; but under a theoretical conditions of continuous emission surface in the surrounding area. This is accomplished by performing the mathematical integrals of the polynomial bounded at different distance ranges (α and β) (2):

$$TI_p = \int_{\beta}^{\alpha} f(x)dx \tag{2}$$

Where TI_p is the influence of the surface of the ring *p* on *PI*, in which α is the farthest distance from the sampling point (the radius of external buffer) and β is the nearest distance to the sampling point (the radius of internal buffer), under theoretical continuous emission surface in surrounding area of sampling point. The ring *p* number is the same as the ring α value, which defines the ring number.

7. The TI_p of a ring *p* is divided between the total surface of the ring (S_p), and the Influence Index (II_p) is thus obtained. II_p summarizes the influence of one surface unit of the ring *p* on *PI*, under a theoretical continuous emission surface in surrounding area of sampling point:

$$II_p = \frac{TI_p}{S_p} \tag{3}$$

where S_p is the total surface of the ring *p*, defined as:

$$S_p = \left(\pi\alpha^2\right) - \left(\pi\beta^2\right) \tag{4}$$

where α is the radius of external buffer and β is the radius of internal buffer.

8. The calculation of the Specific Influence Index (*SII*) (5):

$$SII = \sum_{p=1}^{z} (II_p * AES_p) \tag{5}$$

where *p* is the ring number and *z* is the ring number of the last ring with observed influence (with $TI \geq 0$).

9. The calculation of the Emission Index (*EI*) by linear regression of least squares between *PI* and *SII*, also calculated as (6). *EI* is a constant of emission which depend on external conditions during the study period.

$$EI = \frac{\overline{PI}}{\overline{SII}} \tag{6}$$

where $\overline{PI}$ is the average of *PI* of studied cases and $\overline{SII}$ is the average of *SII* of study cases.

10. The Actual Influence (*AAI*) of AES_p on *PI*, in a determined location, is obtained using the following function (7):

$$AAI_p = II_p * AES_p * EI \tag{7}$$

11. In order to forecast expected values of *PI* in a determined location, the entire *AES* in the surrounding area must be tested by using function (8):

$$PI = \sum_{p=1}^{z} AAI_p \tag{8}$$

2.3. Validation

The method was validated by two ways: by internal validation, by forecasting the PI of 19 sampling units involved in the general model, and by external cross validation, by comparing two completely different models. For external validation, the monitoring stations were ordered alphabetically by the names of the cities in which the sampler are located in order to develop a total random selection. A first model (model A) was calculated using the first ten stations and a second model (model B) was calculated using the other nine stations. Model parameters calculated were *II* and *EI*. Finally, the airborne pollen from the first ten stations was forecasted by using model B parameters and *AES* of forecasted stations. The airborne pollen from the last nine stations was forecasted by using model A parameters and *AES* of forecasted stations. Observed and expected values were compared in order to calculate the determination coefficient (R^2) and the Root Mean Square Error (RMSE). The RMSE was obtained by using the expression (9), where Y = observed data and F = expected data. In this way, the error in cross PI predictions allow us to know fitness and robustness of CRM method.

$$RMSE = \sqrt{\frac{\sum_{t=1}^{N}(Y_t - F_t)^2}{N}} \tag{9}$$

3. Results

Applying the Concentric Ring Method (*CRM*), the observed emission surface on each ring was correlated with the *PI* from each monitoring station and the relationship between correlation coefficients and rings distances were plotted (Fig. 2). CRM is developed with the information from 300 correlation coefficients. Each correlation coefficient is developed from 19 cases in our general model (we only have developed two different models for validation, but all figures are for a general model). Fig. 2 shows the relationship between the outcome of each correlation analysis and the number of analyzed ring. This figure shows that the relationship between both correlation coefficients and the distance of the ring is well fixed to a third-degree polynomial relationship. Results show a high spatial autocorrelation between correlation coefficients, in spite of the lack of spatial overlap between rings. However, the correlation coefficients of nearest rings show less spatial autocorrelation than the following coefficients owing to the "City effect", which is produced for the absence of olive groves near the suction trap, usually located in urban centers. The relative maximum in the polynomial provides information about the distance between a point and the pollen emission source in which the emission focus contributes with the highest quantity of particles in the aerobiological "load" of the point. In this study, the maximum influence is observed at 37 km. Furthermore, the cut with the x axis or the relative minimum, after the inflection point, provide information about the theoretical end of influence as regard with the emission source and the aerobiological features. In this case, there is a lack of correlation at 209 km.

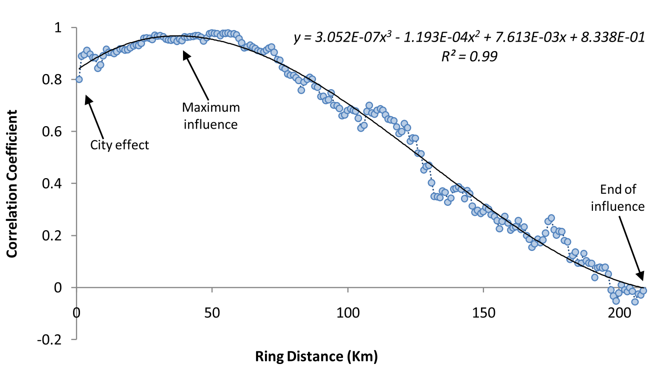

Fig. 2. Polynomial relationship of the correlation coefficient and Pollen Index and Actual Emission Surface on each ring and the ring distance to the sampling point.

The mathematical integration of the polynomial allows us to discover the influence that the emission source has at different distances, thus allowing us to represent the spatial model of olive pollen dynamics, the Theoretical Index (*TI*). Under the continuous emission surface in surrounding area, the emission surface of the first 34 km (3632 km^2 of emission surface) contribute to 25% of airborne pollen, the first 66 km (13,685 km^2 of emission surface) contribute to 50% and the first 104 km (33,979 km^2 of emission surface) contribute to 75% of *Olea* pollen, which is the origin of more than 99% of pollen from the surrounding 208 km (135,918 km^2 of emission surface). In order to discover the actual contribution of each olive grove in each location, it is necessary to calculate the Influence Index (*II*) and multiply by the Actual Emission Surface (*AES*) at each distance from the studied location.

Fig. 3a shows the percentage of influence that the emission surface of each ring has on the PI at different distances under a theoretical continuous emission surface in the surrounding area (from *TI*). The olive pollen-vegetation relationship does not have a spatial "bell-shaped" relationship, but it is rather similar to a spatial "volcano-shaped" relationship. This is because the ring surface increases dramatically with the distance from the monitoring station. Fig. 3b shows the percentage of influence that a km^2 of emission surface has on the Pollen Index at different distances under a theoretical continuous emission surface in the surrounding area (from *II*). It shows a potential mathematical relationship between the percentage of influence of each Km^2 on each ring and the ring distance from the station, in which the contribution of the emission surface to aerobiological features in an increasing ring distance decreases sharply.

Fig. 4 shows the linear relationship between the 19 *PI* and the 19 *SII*, with *EI* equal to 960.2. This represents the average emission rate of particles that have one unit of surface in the study area and during the study period.

Thus, at this point, we have calculated the general forecasting model defined as formula (10). With this general model it is possible to know Olive *PI* on a blind point knowing the emissions surface in surrounding 209 km (under similar external conditions to the period 2005–2007).

$$PI = \sum_{p=1}^{z}(II_p * AES_p * 960.2) \tag{10}$$

where II_p summarizes the influence of one surface unit (one km^2 on our case) of a ring p on *PI*, depending on the distance from the emission source; AES_p is the actual emission surface inside a ring p; 960.2 is the constant of the emission rate (*EI*) of a km^2 during the study period; z is the ring number of the last ring with observed influence (with $TI \geq 0$).

Fig. 5 shows the validation of the general model by means of external cross validation. Linear R^2 was 0.98 and the RMSE was 3071. The internal validation of the total Model shows an RMSE of 2317. Heterogeneity in atmospheric conditions, topography or Emission Indexes ($EI_{\text{model A}} = 934.7$; $EI_{\text{model B}} = 1005$) produce less fitness when using external validation than when using internal validation. However, both summary parameters show the excellent performance of the model for *Olea* pollen.

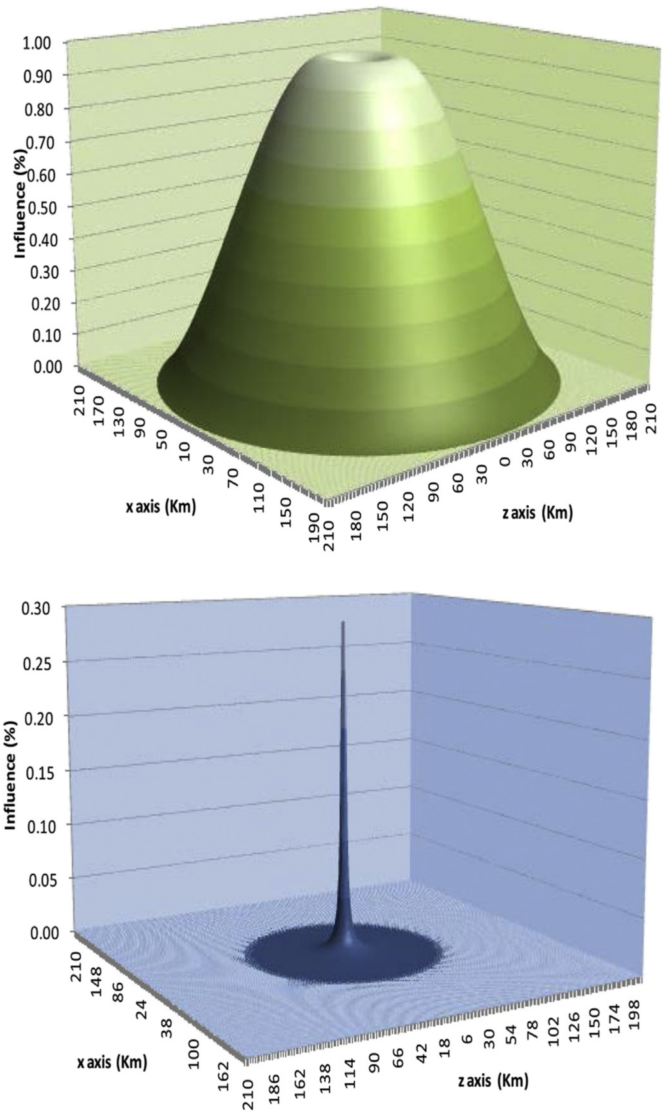

Fig. 3. Fig. 2a shows the 3D-model of influence that has the emission surface of each ring on Pollen Index of sampling point (in center), under theoretical conditions of continuous emission surface. Fig. 2b shows the 3D-model of influence that has each Km^2 of emission surface on Pollen Index of sampling point (in center), under a theoretical conditions of continuous emission surface (following CRM, actual influence of emission source on Pollen Index depend on actual emission surface).

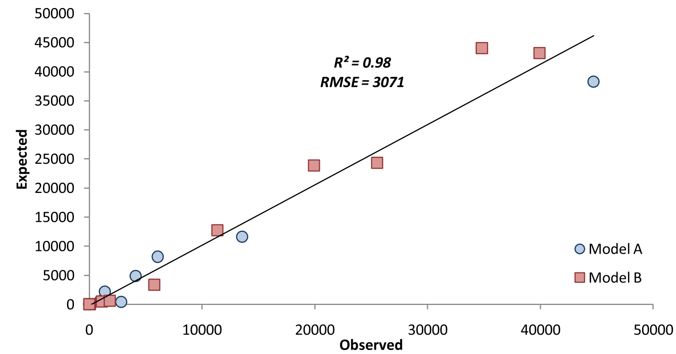

Fig. 5. CRM external cross validation. Pollen Index observed and expected by model A and model B.

4. Discussion

It has been proposed a model for the dynamics of airborne *Olea* pollen in Spain, which allows us to study the distance of the pollen source, and also forecasting the airborne pollen of unmonitored places. The Concentric Ring Method (CRM) can be useful for modeling other kinds of airborne particles such as anthropogenic contaminants or earth emissions, although the resulting model will depend on the features of each particle. In the case of pollen, the suitability for the atmospheric dispersion of airborne particles depends on its aerodynamic features such as size, shape or density (Okubo and Levin, 1989; Prentice, 1985). Sugita suggests, based on theoretical dispersion properties, that the source radius of dispersion of light pollen types could be 100 times larger than that of heavy types (Sugita, 1993). On the other hand, the wind is the main driving force, and the windy conditions can have an impact on the amount of pollen recorded at a site (Damialis et al., 2005). On the other hand, other meteorological features, like rainfall patterns or cloudiness, may also modify the concentrations of airborne particles (García-Mozo et al., 2010). It is even possible that microclimatic features or technical conditions of sampler location could determine the record (Velasco-Jiménez et al., 2012). CRM must therefore be considered as a method for modeling the general dynamics of a particle in a determined area, by extracting the average pattern of the same particle in several sampling points.

Some considerations must be taken into account depending on the aerodynamic features and the emission dynamics of the particle being modeled. In aerobiological studies, when characterizing a geographical point by building the Pollen Index (PI), the pollen

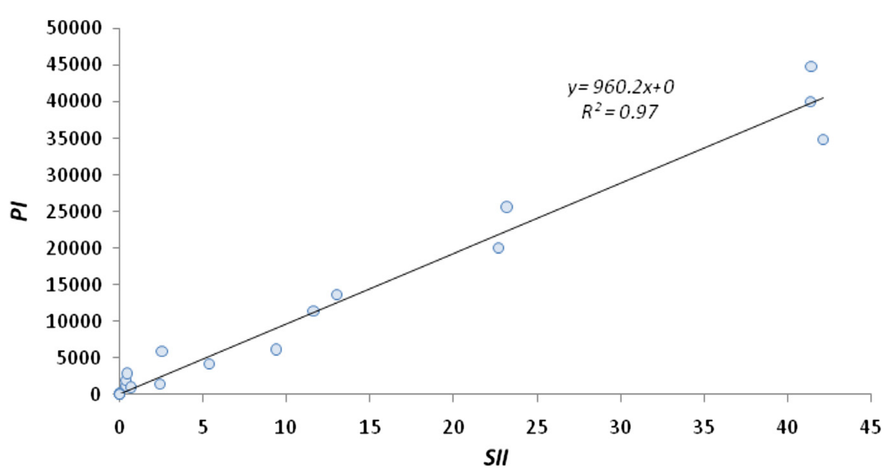

Fig. 4. Linear relationship between Pollen Index and Specific Influence Index.

recorded during more than one year should be taken into account because pollen production can undergo interannual variations related to weather or to genetic features (Hicks, 2001). This fact could distort the relationship between airborne pollen and vegetation. In this sense, it was employed three years when creating the aerobiological characterization of each station because the olive tree has large interannual variability as regards its pollen emission (Oteros et al., 2013b). However, with a high number of years characterizing a point, changes in land uses during the study period could also distort the relationship between the airborne particles and the emission source. Olive grove surfaces may undergo important interannual variations because there is a strong anthropogenic influence (Garcia-Mozo et al., 2014). Another consideration according to the emission source inventories is that the canopy surface information will be more accurate than land use surface, or even, where possible, the emission volume. Olive pollen is a particle whose origin is only one species, which facilitates the availability of its source inventories and homogeneity in its emission rates (Trigo et al., 2008). In Spain, olives trees are in olive crops (Barranco, 2008), as natural vegetation (Skjøth et al., 2008), or as ornamental plants (Velasco-Jiménez et al., 2013). However, most *Olea* pollen originates from olive groves (Fernandez-Rodriguez et al., 2014). It can be assumed that the land use data concerning olive groves provides accurate information about the olive tree canopy or the emission surface since, although it does not take into account the density of the olive trees within olive groves which can range from less than 100 trees/ha to more than 300 trees/ha (De Melo-Abreu et al., 2004). Other interesting information are the productivity of different areas, which can show high reproductive variability owing to topographical factors or management (Oteros et al., 2013d), and the expected lack of spatial autocorrelation in the variability of tree densities homogenizes emission surfaces at large scale.

Although the innovative nature of this methodology does not allow making comparisons with previous results, other approaches for determining the airborne particle source have been applied. Most of them are theoretical methods based on the aerodynamic features of particles by applying dispersion equations, which allow us to study the origin of the airborne particles in specific episodes. Olive pollen has specific aerodynamic features that make it suitable for atmospheric dispersion to long distances (Hernández-Ceballos et al., 2014). This is corroborated by the fact that our experimental method shows that 50% of airborne pollen originate from the surrounding 66 km and more that 99% from the surrounding 208 km. Previous works based on particle dispersion theory have shown the potential source of isolated episodes of olive pollen; e.g. in Fernandez-Rodriguez et al. (2014) showed that certain pollen episodes in Badajoz (Spain) can be explained by pollen sourcing at between 30 km and 300 km; Hernández-Ceballos et al. (2011, 2014) showed that most analyzed episodes of *Olea* pollen in Córdoba (Spain) could originate from the surrounding olive groves located between 25 km (Alcolea) and 200 km (Priego de Córdoba). Despite the fact, in punctual events, olive pollen could be transported from longer distances (Galán et al., 2013; Izquierdo et al., 2011). Moreover, recently Maya-Manzano et al. have compared olive airborne pollen in several points with olive groves, land use in surrounding area, making a simple correlation between circle surface and airborne pollen (Maya-Manzano et al., under review). They have obtained similar results to our work, which suggesting a good relationship between airborne olive pollen and land uses (with maximum correlation at 50 km), except for test with total circle surface and not with ring surfaces. Our results agree with these backgrounds and the validation results certify the robustness of the olive model and the CRM's reliability.

5. Conclusions

As conclusion, the CRM is a new method to model the dynamics of airborne particles, thus providing a new approach that can be used to evaluate the capture range of air traps and allow understanding where is the distance of the origin of the records. The CRM will be very useful to design the location of air samplers and the correct interpretation of historical databases concerning airborne particles. The CRM might also be valuable as an experimental approach to validate theoretical assumptions in certain disciplines, such as Paleopalinology, which involves long distance particle dispersion. The present method will improve the usefulness of monitoring data of some kind of airborne particles, thus increasing the quality of their interpretations and optimizing their actual utilities. In the case of Aerobiology applications, as the olive pollen modeling, CRM will be very useful, because improving forecasting models for aeroallergens, in particular for allergenic pollen, and other harmful particles. Furthermore, improve the crop yield forecasting models, the knowledge of the climate change and its impacts on the phenology and pollen production, the past and present vegetation maps, the capacity to examine the gene flow of anemophilous vegetation, or forecasting the airborne characteristics of an unexplored location, and other potential applications.

Author contribution

J.O. data analysis, experimental design and redaction; H.G.M., P.A., E.D.V. and C.G. experimental design, supervision, data collection, redaction and management; P.C., J.B., M.B., D.F.G, A.M.G.B., R.P.B. and M.M.T. data collection, supervision, redaction and testing; D.B., C.D.G., F.G.M., S.M.G., F.J.R.R., L.R.V. and J.S.P. data collection, redaction and supervision.

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