APPLICATION OF A SOURCE-RECEPTOR MODEL TO DETERMINE SOURCE AREAS OF POLLEN IN CATALONIA (NE SPAIN)

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Abstract: In this study we characterize the long-range transport of allergenic pollen to the North East of Spain. To this end, cluster analysis of back-trajectories and a source-receptor model was applied to a dataset of 10 years of airborne monitoring data corresponding to the period 2000-2009. The aim of the study is to examine and interpret the main transport routes and source areas affecting levels in Catalonia for a total of 20 pollen types. The cluster analysis of trajectories revealed that trajectories entering to the region by East France represent well the long distant transport of some taxa which are scarce in the Catalan territory or present only in limited areas (*Ambrosia*, *Betula*, *Corylus* and *Fagus*) together with other common in Catalonia (*Fraxinus*, *Quercus* deciduous type and *Pistacia*) showing agreement with the source areas obtained from the source-receptor methodology: Whereas, for taxa with local/regional sources (*Artemisia*, *Castanea*, total Poaceae, *Plantago*, total Polygonaceae, *Olea*, total *Quercus* and *Quercus* evergreen type) with high concentration levels associated to situations of recirculation over the Iberian Peninsula, the source-receptor model did not show reliable source regions.

Key words: *Long-range transport, back-trajectories, cluster analysis, source-receptor model, airborne pollen*

INTRODUCTION

The atmosphere contains biological material such as microorganisms, mould spores, diaspora of plants (pollen and seeds of small size), and small animals like insects and arachnids that spend part of their life cycle in the atmosphere. The pollen grains incorporated into the atmosphere by anemophilous plants can produce allergic symptoms in a part of the population sensitive to it and the number of the derived health problems can be induced by long-range transport episodes. Indeed, the airborne pollen patterns in Catalonia show abrupt high concentrations in areas with usually low local influence. The use of modeling is a good tool to study and understand the atmospheric mechanisms that cause these peaks. Cluster analysis of back-trajectories has been widely used to characterize the synoptic situations associated to long-range transport (Dorling and Davies, 1995; Jorba et al., 2004) that affect air pollution.

On the other hand, the Seibert's methodology, based on concentration fields, (Seibert et al. 1994) as well as other source-receptor models, have been profusely used for the interpretation of air pollutant transport, mainly of chemical compounds such as ozone, acidifying components, mineral dust, and other pollutants (Stohl, 1996; Charron et al., 1998; Polissar et al., 2001; Hoh and Hites, 2004; Salvador et al., 2004) and show also valid results when applied to pollen and spore particles (Izquierdo et al., 2011; Fernández-Llamazares et al., 2012).

The aim of this study is to examine and interpret the main transport routes and source areas affecting levels for a total of 20 pollen types in Catalonia. A 10-year flow climatology has been made by means of cluster analysis of back-trajectories in order to characterize synoptic weather regimes affecting long-range transport of airborne pollen to NE Spain. To determine the geographical location of the potential source regions, a source-receptor model has been applied to a dataset of airborne monitoring and back-trajectory data for the same 10-year period from 2000 to 2009.

MATERIAL AND METHODS

Pollen data

Airborne pollen data were recorded for the period 2000 – 2009 by the Aerobiological Network of Catalonia (Xarxa Aerobiològica de Catalunya, XAC) at the Bellaterra monitoring station (Figure 1). Samples were obtained daily from Hirst samplers (Hirst, 1952), the standardized method in European aerobiological networks, and analyzed following the standardized Spanish method (Galán et al. 2007). Twenty relevant pollen taxa from the landscape point of view were tested: *Ambrosia*, *Artemisia*, *Betula*, *Castanea*, *Corylus*, *Cupressaceae*, *Fagus*, *Fraxinus*, total Poaceae, *Olea*, *Pinus*, *Pistacia*, *Plantago*, *Platanus*, total Polygonaceae, Chenopodiaceae-Amaranthaceae, *Quercus* deciduous type, *Quercus* evergreen type, total *Quercus* and *Ulmus*.

Air mass trajectories

The provenance of the air-masses transporting pollen was examined using backward atmospheric trajectories. Daily isentropic 96-h back-trajectories at 1500 m.a.s.l., starting at 12 UTC from the coordinates of the monitoring site were computed using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT-4) of the National Oceanic and Atmospheric Administration (NOAA) [available at http://www.arl.noaa.gov/ready/hysplit4.html, Draxler & Rolph, 2003] from the gridded meteorological fields of the FNL and GDAS archive data. The interpretation of the backward trajectories was complemented with meteorological synoptic maps from the UK Meteorological Service [available at http://www.weathercharts.org/ukmo-analysis/].

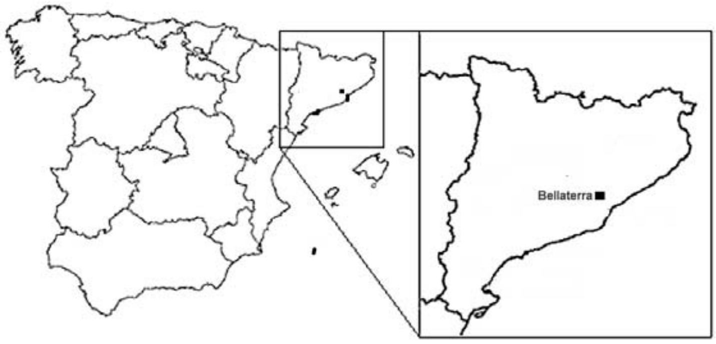

Figure 1. Location of the Bellaterra pollen monitoring station of the Aerobiological Network of Catalonia (Xarxa Aerobiològica de Catalunya, XAC).

Cluster analysis of back-trajectories was done in order to group trajectories by using the HYSPLIT cluster module based on maximizing the Total Spatial Variance (TSV) between clusters and minimizing the spatial variance (SPVAR) within them (Draxler *et al*. 2009). The final number of clusters is determined by a change in TSV as clusters are iteratively paired. Anova and Tukey test were applied to the pollen counts in the clusters in order to determine whether the groups significantly differed between them.

Source-receptor model

A statistical approach combining pollen concentration data at the monitoring station with the backward trajectories ending at the location was applied to infer the source areas for the pollen reaching Catalonia in the study period. Such source-receptor methodologies establish relationships between a receptor point and the probable source areas by associating each value of pollen abundance with its corresponding backward trajectory.
In this study, two daily 72-hour backward trajectories (at 00 and 12 UTC) were considered at 1500 m height during the flowering period of each taxon, for the ten years from 2000 to 2009. A grid, in this case composed of 2601 cells of 1ºx1º latitude and longitude, was then superimposed on the integration region of the trajectories in order to map the contributing areas.
The Seibert methodology [Seibert *et al*., 1994], in which a logarithmic mean pollen concentration is computed for each grid cell based on the residence time of the trajectories in the cells, was applied:

$$\log C_{ij} = \frac{\sum_l n_{ijl} \log C_l}{\sum_l n_{ijl}} \qquad (1)$$

where C_{ij} is the pollen concentration in the (i,j) cell, l is the index of the trajectory, n_{ijl} is the number of time steps of the trajectory l in the cell (i,j), and C_l is the pollen concentration measured at the receptor point corresponding to the trajectory l. To minimize the uncertainty of the trajectories, a smoothing was applied and the value of each cell was replaced by the average between the cell and the eight neighboring cells. A final filter excluded cells with less than 30 end points. The abundance field map obtained in this manner reflects the contribution of each cell to the pollen abundance at the receptor point.

RESULTS AND DISCUSION

Climatology of back-trajectories

The cluster analysis of back-trajectories at 1500 m.a.s.l. showed a flow climatology associated to the six following clusters (Figure 2): C1 corresponded to a West zonal flow from the Atlantic Ocean at medium distance (ATMD_W) containing 19% of trajectories; C5 corresponded to a North West long-distance flux (ATLD_NW), with 7% of frequency. Both, ATMD_W and ATLM_NW, were characterized by fast-moving trajectories from the Atlantic Ocean, common during the winter months. C2 were associated to an Atlantic short distance flow (ATSD_NW) with 19% of presence, with slow trajectories from W and NW present all year, but mainly in summer (Figure 3), entering the Iberian Peninsula by the Cantabric Sea. A regional flow (PEN, C3), the most frequent, which contributed 24%, corresponded to slow air-masses, including recirculations over the Iberian Peninsula and southern advective flows, typical of the summer months during situations with weak baric gradient. The cluster C4 corresponded to continental European advection entering the Iberian Peninsula from eastern France (FR_E, 20%) with fluxes from central Europe and the Mediterranean Sea, frequent in winter and spring, with cold, cloudy and dry weather. Cluster C6 (France West, FR_W) with 11% of trajectories, represented the northern advections from France and United Kingdom, reaching Catalonia by the Pyrenees, which are frequent in winter and spring, from November to April (Figure 3). The centroids of the clusters are represented in Figure 2.

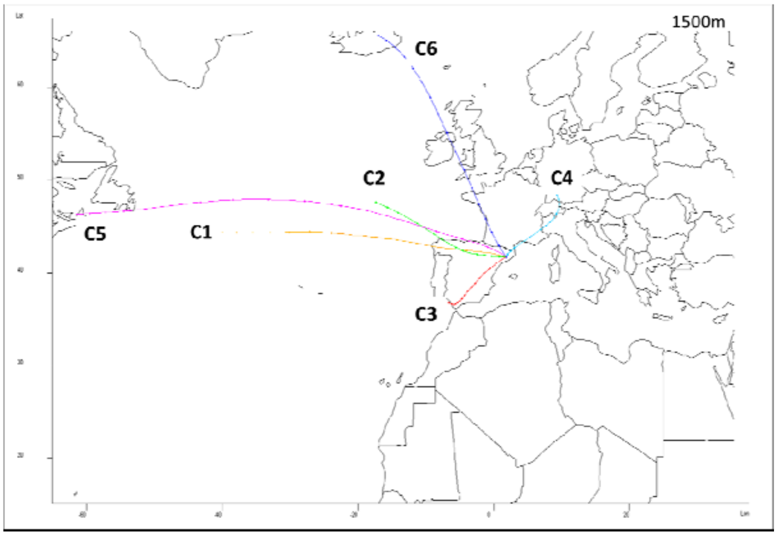

Figure 2. Trajectories representing the centroids of the six groups obtained from the cluster analysis for the 10-year period 2000-2009 at 1500 m.a.s.l.

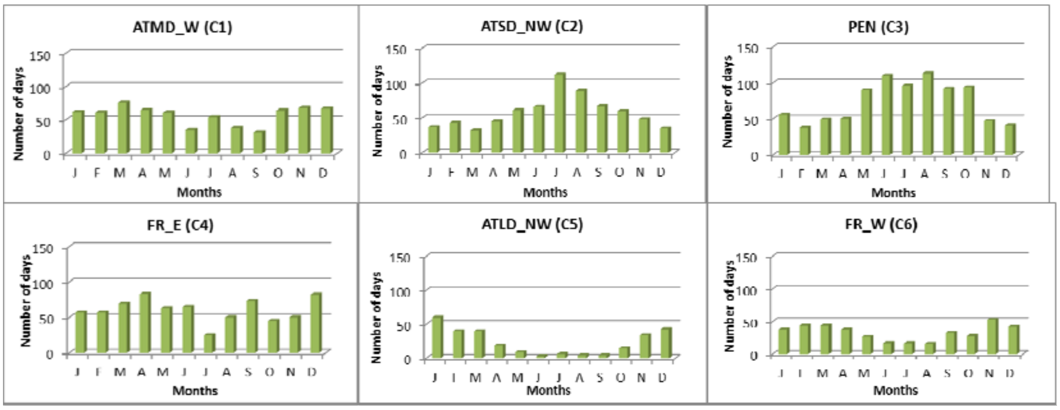

Figure 3. Monthly distribution (number of days during the 10-year period) of each cluster

Pollen in clusters

Relevant clusters for airborne pollen transport were ATMD_W, ATLD_NW, FR_E and PEN. The main cluster for each taxon and the percentage of pollen in the cluster are shown in Table 1. The main flow routes of incoming pollen in Catalonia were: Central Europe (FR_E) for *Ambrosia*, *Betula*, *Corylus*, *Fagus*, *Fraxinus*, *Quercus* deciduous type and *Pistacia*; the Peninsular recirculations and southern advections, represented by cluster PEN, had a major influence for *Artemisia*, *Castanea*, total Poaceae, *Plantago*, total Polygonaceae, *Olea*, total *Quercus* and *Quercus* evergreen type. The western provenances at long and medium distance (ATLD_NW and ATMD_W) were important for *Cupressaceae*, *Olea*, *Pinus*, *Platanus*, *Quercus* deciduous type, total *Quercus* and *Ulmus*. And the NW provenance at short distance (ATSD_NW) was the cluster with major load of Chenopodiaceae-Amaranthaceae. The FR_W cluster was not representative for the transport of any taxon, probably because it is associated to the presence of a deep depression over the North Sea which produces intense cloudiness and frequent rainfall in NE Spain, cleaning the atmosphere of any kind of pollution. Clusters only presented significant differences in their pollen content, according to Anova and Tukey's test, for six of the taxa: *Cupressaceae* (ATMD_W), *Fagus* (FR_E), total Poaceae (PEN), *Plantago* (PEN), Chenopodiaceae-Amaranthaceae (ATSD_NW) and *Ulmus* (ATLD_NW) (Table 1).

In order to establish the pollen load associated to the different clusters, the percentage of presence of the main cluster during the pollination period was computed for each taxon. Then, the quotient between the percentage of pollen in this main cluster and its percentage of presence provides insight into the load carried by the air-mass represented by the cluster. A high value of this ratio for a given taxon indicates that the meteorological situation associated to its main cluster is not very frequent during the pollination period but, when it is done, the air-mass transports high load of this pollen. For instance, in the case of *Fagus*, the main cluster was FR_E. This cluster accounted for 63% of the total *Fagus* pollen collected, although it was present only in 21% of the days of the *Fagus* pollination period. The ratio between these two percentages is about 3, reflecting the high pollen load associated to this pathway. FR_E gave also a high load ratio (≈ 2) for *Betula* and *Ambrosia*, and was >1 in all the taxa. In contrast, the regional and Peninsular recirculations (PEN) corresponded to the cleanest air-masses, giving load ratios lower or slightly higher to 1, for all the taxa in which this cluster was the main one.

Table 1. Main cluster for each taxon, the percentage of pollen in each cluster and the ratio between pollen load (%) and the presence of each main cluster (%). The * indicates taxa with significant difference between groups in their pollen content.

Taxon	**Cluster**	**Pollen in cluster (%)**	**Ratio**	**Taxon**	**Cluster**	**Pollen in cluster (%)**	**Ratio**
Ambrosia	FR_E	32	2.0	*Pinus*	ATMD_W	24	1.3
Artemisia	PEN	24	0.8	*Pistacia*	FR_E	28	1.2
Betula	FR_E	49	2.1	*Plantago*	**PEN ***	29	1.0
Castanea	PEN	34	1.0	*Platanus*	ATMD_W	27	1.2
Corylus	FR_E	35	1.6	*Total Polygonaceae total*	PEN	25	0.8
Cupressaceae	**ATMD_W ***	28	1.5	Chenop.-Amaranthac.	**ATSD_NW ***	24	1.0
Fagus	**FR_E ***	63	3.0	*Quercus* deciduous t.	ATMD_W / FR_E	26 / 26	1.5/ 1.3
Fraxinus	FR_E	25	1.1	*Quercus* evergreen t.	PEN	27	1,1
Total Poaceae	**PEN***	26	1.0	*Quercus* total	ATMD_W / PEN	24 / 24	1.3/ 0.9
Olea	ATMD_W / PEN	26 / 26	1.6/ 0.8	*Ulmus*	**ATLD_NW ***	22	1,6

Source receptor model

The results of the source-receptor model were consistent with the pathways obtained by back-trajectory cluster only for those taxa not present or scarce in the Catalan territory. Indeed, *Ambrosia*, *Betula*, *Corylus*, *Fagus*, and *Pistacia* showed the main source-regions in central Europe, suggesting a major and quite common mechanism of incoming Catalonia by the SE France, in agreement with the results of the back-trajectory cluster analysis mentioned above. Figure 4 shows the source regions for *Betula*, *Fagus*, *Corylus* and *Pistacia*.

In contrast, for *Artemisia*, *Castanea*, total Poaceae, *Plantago*, total Polygonaceae, *Olea*, total *Quercus* and *Quercus* evergreen type, corresponding to the PEN cluster, which are abundant in the Iberian Peninsula and Catalonia, the source-receptor model did not show coherent source regions. Something similar occurred for *Cupressaceae*, *Pinus*, *Platanus*, Chenopodiaceae-Amaranthaceae and *Ulmus*, associated to W and NW Atlantic fluxes reaching Catalonia after crossing the Iberian Peninsula; for these taxa, also with a high influence of the local and regional transport, the source-receptor model did not show reliable source regions.

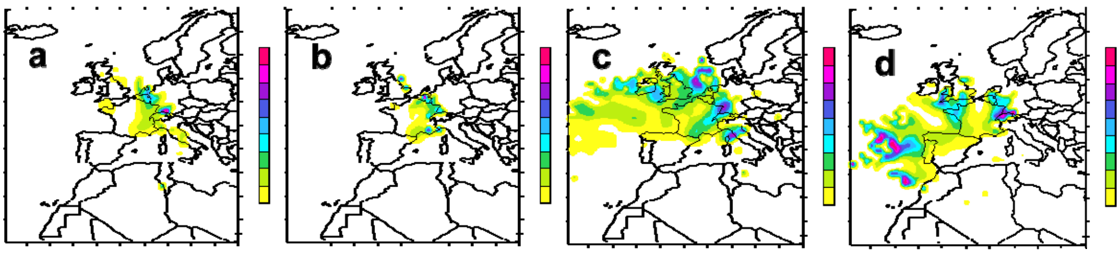

Figure 4. Source regions computed from the source-receptor model for a) *Betula*, b) *Fagus*, c) *Corylus* and d) *Pistacia*

CONCLUSIONS

A 10-year back-trajectory cluster analysis at 1500 m.a.s.l. was applied to characterize the flow climatology affecting synoptic scale transport in order to obtain the main routes of incoming airborne pollen to Catalonia. This methodology revealed that trajectories entering to the region by East France represent well the long distant transport of some taxa which scarce in the Catalan territory or present only in limited areas (*Ambrosia*, *Betula*,

Corylus and *Fagus*) together with other common in Catalonia (*Fraxinus*, *Quercus* deciduous type and *Pistacia*). These meteorological situations included Northern and North-East slow fluxes with large residence time in the atmosphere, reaching the destination region with high pollen loads. On the other hand, the application of a source receptor-model to the same period of time, situated the main source regions for these taxa in central Europe, being this consistent with the results of the cluster analysis.
Trajectory cluster analysis showed also that the situations of recirculation over the Iberian Peninsula are associated with high pollen levels of some taxa (*Artemisia*, *Castanea*, total Poaceae, *Plantago*, total Polygonaceae, *Olea*, total *Quercus* and *Quercus* evergreen type) which are abundant in the Spanish territory and Catalonia. These situations are characterized by western slow fluxes and southern warm air-masses from North Africa. For these taxa of regional and local character, the source-receptor model did not well represent the source regions.

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