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Biogeographical estimates of allergenic pollen transport over regional scales: Common ragweed and Szeged, Hungary as a test case

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ABSTRACT

Long-distance pollen transport can substantially raise local pollen levels, but their relative contribution has not yet been quantified temporally or spatially in ragweed infested regions. Using common ragweed (*Ambrosia artemisiifolia*) pollen accumulation at a ragweed infested area, Szeged, Hungary as a test case, this study attempted to: (1) identify, using cluster analysis, biogeographical regions that contribute to long-range transport of ragweed pollen to Szeged; (2) quantify the relative

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contribution of ragweed pollen from these regions; (3) determine the relative contribution of "local" and "transported" pollen for Szeged. Using the HYSPLIT model, three-dimensional backward trajectories were produced daily over a 5-year period, 2009–2013 for ragweed pollen accumulation at Szeged. A *k*-means clustering algorithm using the Mahalanobis distance was applied in order to develop trajectory types. Nine back-trajectory clusters were identified. Cluster 1 (direction: from the Channel area south of Great Britain) and cluster 5 (direction: from Northern Mediterranean) were found the most relevant potential long-distance sources for *Ambrosia* pollen transport to Szeged. Potential source contribution function (*PSCF*) and concentration weighted trajectory (*CWT*) values indicated additional potential source areas including the central and eastern part of France, the northern part of Italy and the Carpathian Basin. For Szeged on non-rainy days, medium-range transport is important, while on rainy days the two transport ranges have equal weights. Based on the Granger causality, annual pollen amount transported by the atmospheric circulation is 27.8% of the annual total pollen at Szeged. From this quantity, 7.5% is added to (due to transport), while 20.3% is subtracted from (e.g. because of wash-out by frontal rainfalls going towards Szeged) local sources.

1. Introduction

1.1. Ambrosia *pollen in Europe*

Among the most important invasive plants introduced to Europe, *Ambrosia artemisiifolia*, or common ragweed involves the highest environmental threat through its impact on yield losses, pollen allergy and other direct and indirect social and economic injuries (Makra et al., 2005; Bullock et al., 2010; Vinogradova et al., 2010). The genus of ragweed (*Ambrosia spp.*) comprising 42 species is widely acknowledged as a global source of allergenic pollen and a significant cause of seasonal allergic rhinitis (Béres et al., 2005). Allergic disorders are a widespread recognised chronic disease, those economic and health costs can run into billions of Euros (Bullock et al., 2010).

The spread of *A. artemisiifolia* across Europe from pre 1900 to 2011, regarding four time periods (up to 1900, up to 1960, up to 1990 and up to 2011), shows an accelerated increasing expansion reaching even until the East European Plain in Russia in east and even until the mid-northern areas of Sweden and Finland in north (Bullock et al., 2010; Cunze et al., 2013; Prank et al., 2013; Chapman et al., 2014; Storkey et al., 2014).

The most important habitat areas of common ragweed in Europe, in decreasing order, are as follows. (1) The Ukraine (Rodinkova et al., 2012; Rodinkova, 2014) and the southern part of the European Russia, particularly the latter area, could be a substantial, if not the most important source of ragweed pollen in Europe (Reznik, 2009). Note that ragweed pollen measurements over the most extended habitat areas of *Ambrosia* in the Ukraine (Victoria Rodinkova, personal communication) and the European Russia (Elena Severova, personal communication) have only started at the very end of the 2000s and even the data are rather incomplete. Accordingly, no reliably long data sets are available for quantitatively evaluating *Ambrosia* pollen abundance over these areas compared to well-known pollen sources in Europe. (2) The Pannonian Plain, especially its part, the Hungarian Great Plain including major area of Hungary and Serbia; furthermore, extended regions of Croatia, Slovenia, Slovakia and Romania within the Carpathian Basin (Kiss and Béres, 2006; Makra et al., 2005) are the largest areas of ragweed population in the European Union. (3) The Po River valley with special attention to Western Lombardy in Italy (Bonini et al., 2012), and (4) the Rhône-Alpes region and the central areas in France (Chauvel et al., 2006; Gladieux et al., 2011; Thibaudon et al., 2014) are the most extended habitat areas in Western Europe.

Because of its importance in public health (Cecchi et al., 2006; D'Amato et al., 2007), biogeographical information on the sources and movement of ragweed pollen can be an effective tool in its mitigation and control. Although the establishment of common ragweed has long been recognised in European habitats, current and potential transport of its pollen has not been well quantified. Long-distance pollen transport may substantially raise local pollen levels. Main aspects of this issue are summarised as follows.

1.2. *Long-distance transport of* Ambrosia *pollen*

Since the settling velocity of a ragweed pollen grain is 1 cm s^{-1}, it would take about 1 day for the p EEA Technical ollen to fall 1000 m through the atmosphere. Considering the average wind speed in Europe (EEA Technical Report, 2009), this involves well over 100 km travel for the pollen without falling out, being the lower threshold of the long-distance transport (OECD, 2008; WMO, 2008).

Several authors detected source areas of ragweed pollen arriving at the target area through long-distance transport (Table 1). It was found that ragweed pollen arrived at Poland through long-range transport from Czech, Slovakia and Hungary (Smith et al., 2008; Kasprzyk et al., 2011), as well as from the Ukraine (Kasprzyk et al., 2011). A major long-range source area of *Ambrosia* pollen for Szeged (Hungary) was found the Pannonian Plain in the Carpathian Basin (Makra et al., 2010). Ragweed pollen peaks were recorded in Florence and in Pistoia (both in Italy) when north-north-east winds were observed. Due to back-trajectory analysis, southern Hungary was found as a possible source area of *Ambrosia* pollen over these cities through long-distance transport (Cecchi et al., 2006). Zemmer et al. (2012) detected that transported ragweed pollen arrived at the atmosphere of Istanbul from Bulgaria (a regional source area), furthermore from Moldova, the Ukraine and the Russian coastal region of the Black Sea (long-distance sources). Šikoparija et al. (2009) found the southern part of the Pannonian Plain around Novi Sad and Ruma as a potential source region for *Ambrosia* pollen recorded at Niš and Skopje in the Balkans (Šikoparija et al., 2009). In addition, some authors (Šikoparija et al., 2009; Zink et al., 2012) concluded that long-distance transport should not be neglected when predicting ragweed pollen concentrations over a target area. Peak *Ambrosia* pollen counts in Szczecin (Poland) in 2002 (Puc, 2004) and 2003 (Puc, 2006) are also associated with long-range airflows from south and south-east. In Sweden, occurrences of *Ambrosia* spp. were rare formerly. In recent years, however, long-range pollen transport has been detected in South Sweden (Šikoparija et al., 2013) that is likely to be increasingly more common as ragweed is rapidly spreading in Europe (Dahl et al., 1999; Bullock et al., 2010; Cunze et al., 2013; Prank et al., 2013; Smith et al., 2013; Chapman et al., 2014; Storkey et al., 2014) (Table 1).

Table 1
Ragweed related studies modelling long-range transport of ragweed pollen, comprising model name, information on the study area, sites and the period involved, back-trajectory information and a device for interpreting pollution episodes with the reference of the study.

Pollen transport model and its reference	Study			Back-trajectories		Device for interpreting pollution episodes	Reference
	Area	Sites	Years	[a]Number/site	Duration (day)		
Receptor models							
Air Resources Laboratories (ARL) Atmospheric Transport and Dispersion Model	Albany, NY, USA	1	n/a	9	n/a	n/a	Guercio et al. (1980)
[b]MASS (Koch et al., 1985)	Spain	3	1996	1 at each site	2	Single backward trajectories	Belmonte et al. (2000)
Synoptic back-trajectories method (Saar et al., 2000)	Baltic states	3	1996, 1997	30, 30, 4	4	Single backward trajectories	Saar et al. (2000)
[c]HYSPLIT version 4 (Draxler and Rolph, 2003)	Italy	2	2002	1 at each site	2	Single backward trajectories	Cecchi et al. (2006)
[c]HYSPLIT version 4 (Draxler and Rolph, 2003)	Italy	2	2002, 2004	1 at each site	2	Single backward trajectories	Cecchi et al. (2007)
[c]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Hungary	1	1991, 1992, 1993, 1994, 2000, 2003	3	2, 3.5, 4, 7	Single backward trajectories	Makra and Pálfi (2007)
[b]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Hungary	1	1993, 2000, 2003	3	3, 7	Single backward trajectories	Makra et al. (2007)
[d]ACDEP (Skjøth et al., 2002)	Poland	1	2002, 2005	6	1	Single backward trajectories	Stach et al. (2007)
[c]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Germany	2	2006	1 at each site	1	Single backward trajectories	Gabrio et al. (2008)
[d]ACDEP (Skjøth et al., 2002)	Poland	8	2005	2 or 7 per site	1	Single backward trajectories	Smith et al. (2008)
[b]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Lithuania	3	2004–2009	126 at each site	3	Single backward trajectories	Šaulienė and Veriankaitė (2009)
[d]ACDEP (Skjøth et al., 2002)	Serbia, Macedonia	2	2007	24 and 36	1 and 2	Single backward trajectories	Šikoparija et al. (2009)
[c]**HYSPLIT version 4.8** (Draxler and Hess, 1998)	**Hungary**	**1**	**1998–2002**	**450**	**4**	**Cluster analysis**	Makra et al. (2010)
[c]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Lithuania	3	2004	3 at each site	3	Single backward trajectories	Veriankaitė (2010)
[e]**CMAQ** (Byun and Ching, 1999) **and** [b]**HYSPLIT version 4.8** (Draxler and Hess, 1998)	**USA**	**1**	**2002**	**n/a**	**n/a**	**Cluster analysis**	Efstathiou et al. (2011)
[d]ACDEP (Skjøth et al., 2002)	Poland	3	1999, 2002	48 at each site	2	Single backward trajectories	Kasprzyk et al. (2011)
[c]HYSPLIT version 4 (Draxler and Rolph, 2003)	Lithuania	1	2008	63	3	Single backward trajectories	Saulienė et al. (2011)
[c]HYSPLIT version 4 (Draxler and Rolph, 2003)	Spain	8	1983–2010	3 at each site	5	Single backward trajectories	Fernández-Llamazares et al. (2011)
[b]HYSPLIT version 4 (Draxler and Rolph, 2003)	Spain	1	2004	3	5	Single backward trajectories	Fernández-Llamazares et al. (2012a)
[c]HYSPLIT version 4 (Draxler and Rolph, 2003)	Spain	8	2004	3 at each site	5	Single backward trajectories	Fernández-Llamazares et al. (2012b)
[f]WRF-ARW (Skamarock and Weisman, 2009)	Croatia	2	2002, 2003	9 at each site	2	Single backward trajectories	Prtenjak et al. (2012)
[c]HYSPLIT version 4.9 (ARL, 2011)	Turkey	1	2007	27	3	Single backward trajectories	Zemmer et al. (2012)
[c]HYSPLIT version 4.8 (Draxler and Hess, 1998)	Serbia	1	2011	2 backtr.[g] 2 forwtr.[h]	3 4	Single backward trajectories	Šikoparija et al. (2013)

[a] Number/site = the number of back-trajectories per site. If, for example, there is only one site in column 3 (column "Sites"), then the number in column 5 (column "Number/site") shows how many back-trajectories are analysed for the given site. At the same time, if there are two or more sites in column 3, than the numbers in column 5 indicate how many back-trajectories are analysed for these sites, respectively.
[b] MASS = Mesoscale Atmospheric Simulation System.
[c] HYSPLIT = **Hy**brid **S**ingle-**P**article **L**agrangian **I**ntegrated **T**rajectory model.
[d] ACDEP = **A**tmospheric **C**hemistry and **Dep**osition model.
[e] CMAQ = **C**ommunity **M**ultiscale **A**ir **Q**uality mode.
[f] WRF-ARW: WRF = Weather Research and Forecasting; ARW = Advanced Research WRF.
[g] backtr. = backward trajectory.
[h] forwtr. = forward trajectory.

1.3. Viability of the pollen grains in long distance transport

Several studies suggest the allergen existence in the atmosphere separately from the pollen grains. However, there is no general understanding of the underlying processes, and the phenomenon itself is still debated (Sofiev et al., 2012). Another new area with strongly insufficient knowledge is the interactions of airborne allergens and chemical air pollutants (Motta et al., 2006). The study of long-range pollen transport has encouraged a new research area about pollen allergenicity (Cecchi et al., 2010) and pollen viability (Bohrerova et al., 2009; Sofiev et al., 2012) after the long distance dispersion. Main transformation processes occurring during long-range transport are as follows. (1) Loss and gain of water depending on the air humidity (Emberlin, 1999; Bohrerova et al., 2009). Sometimes this situation provokes the pollen grain rupture and allergen release. (2) Loss of viability due to temperature and UV radiation (Bohrerova et al., 2009). (3) Chemical damage of the grains by strong oxidants that can take place in polluted environments, inducing intense respiratory allergy symptoms (Motta et al., 2006). (4) Release of the pollen content due to the grain rupture or (pseudo)germination (Pacini, 1997; Sofiev et al., 2012).

Pollen grains that are partly hydrated at the time of dispersal are usually short-lived and cannot stand dehydration (Pacini and Hesse, 2004). In the context of pollen allergies, it is important to distinguish pollen viability and allergenicity. The pollen allergens which are glycoproteins produced by the pollen, may retain their impact on the human immune system for a long time, after the pollen grain is essentially dead (Yli-Panula and Rantio-Lehtimäki, 1995). At the same time, pollen viability indicates no direct correlation with pollination (Pacini, 1997). It is estimated that, due to abiotic stresses including pollen type dependent temperature and relative humidity (Emberlin, 1999) as well as UV-A and UV-B irradiance, around 50% of the pollen grains would survive 24 h of long distance transport under typical external conditions (maize, Emberlin, 1999; pine, Bohrerova et al., 2009; Scots pine, Varis et al., 2009). The viable range of the pollen is, therefore, shorter than the physical dispersal distance (Bohrerova et al., 2009). Long-range transported pollen grains maintain allergenic properties (Grewling et al., 2013) and therefore are expected to trigger allergic reactions among sensitive persons (birch, Hjelmroos, 1991; ragweed, Bullock et al., 2010).

1.4. Importance of 3D back-trajectory clustering vs single back-trajectory analysis

Numerous authors used single backward trajectories to interpret particular pollution episodes (e.g. Hjelmroos, 1991; Franzen et al., 1994; Rousseau et al., 2004, 2008; Cecchi et al., 2006; Šikoparija et al., 2009; Kasprzyk et al., 2011).

However, trajectories arriving at a given location over a relatively long distance can be analysed by a cluster analysis in order to investigate patterns of data within a dataset. Cluster analysis has been applied by many authors to classify individual atmospheric trajectories into a relatively small number of groups (Dorling et al., 1992; Zemmer et al., 2012). This procedure identifies clusters of data sets without providing information on cause-effect relationships. Nevertheless, these structures can help us to interpret airborne ragweed pollen data, e.g. identify possible transport patterns. Of course, the reliability of the results using cluster analysis increases with increasing number of the backward trajectories analysed (Stohl, 1998; Borge et al., 2007).

1.5. Objectives

The Hungarian Great Plain in the Pannonian Plain, with the city of Szeged at its geographical centre, is noted for its high diurnal and annual ragweed pollen counts (Fig. 1) (Makra et al., 2005) and has among the highest rates of allergic rhinitis in Europe (Harsányi, 2009). At present, no information is available that has quantified the relative contribution of long-distance sources of ragweed pollen to Szeged; or how that contribution varies spatially or temporally. Using Szeged and ragweed pollen as a test case, the purpose of the current study was to: (1) identify, using cluster analysis, biogeographical regions that contribute to long-range transport of ragweed pollen to Szeged; (2) quantify the relative contribution of ragweed pollen from these regions; (3) detect casual relationship between *Ambrosia* pollen at Szeged and other sites and hence (4) determine the relative contribution of “local” and “transported” components of the actual *Ambrosia* pollen levels for Szeged.

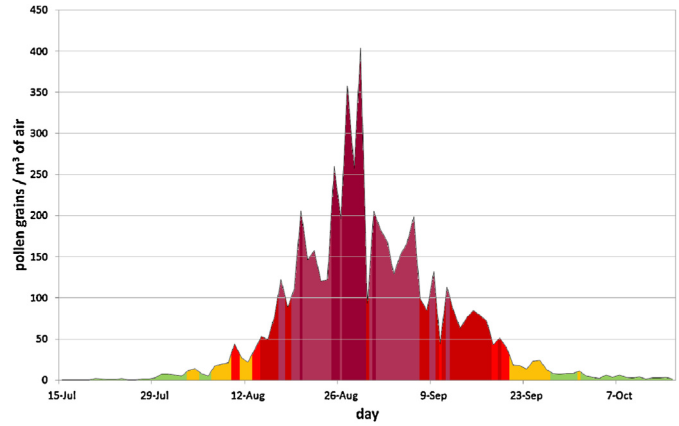

Fig. 1. Daily mean ragweed pollen concentration, Szeged, 2009–2013, July 15–October 15. (The most serious daily mean pollen concentrations occur on August 20, August 25, between August 27–29 and on August 31.) [The colours shown here are introduced by the National Environmental Health Institute in Hungary in order to demonstrate and explain different categories of ragweed pollen load and their health related symptoms (Mányoki et al., 2011).] Colours belonging to the threshold days are as follows: green: July 15–August 2; orange: August 3–4; green: August 5–6; orange: August 7–9; red: August 10; orange: August 11–12; red: August 13; brick: August 14–16; burgundy: August 17; brick: August 18; burgundy: August 19; **dark burgundy: August 20;** burgundy: August 21–24; **dark burgundy: August 25;** burgundy: August 26; **dark burgundy: August 27–29;** brick: August 30; **dark burgundy: August 31;** burgundy: September 1–7; brick: September 8; burgundy: September 9; red: September 10; burgundy: September 11; brick: September 12–17; red: September 18; brick: September 19; red: September 20; orange: September 21–26; green: September 27–30; orange: October 1; green: October 2–15. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2. Materials and methods

2.1. Study area and climate

Szeged (46.25°N; 20.10°E), the largest settlement in Southern Hungary (population: 230,000) is located at the confluence of the Rivers Tisza and Maros within the Carpathian Basin. The area is characterised by an extensive flat landscape, with an elevation of 79 m above sea level. The climate is characterised by relatively mild winters and hot summers, namely **Cfa** in Köppen climate classification (Köppen, 1931). The southern part of Hungary, including Szeged area, shows one of the highest ragweed pollen loads in Europe (Fig. 1) (Mányoki et al., 2011) (Fig. 2; Table 2).

Lyon (45.77°N; 4.83°E) has the second largest metropolitan area in France, with a population of 1.8 million. The city is located in the Rhône valley with an elevation of 175 m AMSL. In the Köppen system its climate is of the **Cfb** type. That is, it has a temperate oceanic climate with mild winters and cool-to-warm summers, as well as a uniform annual precipitation distribution (Köppen, 1931) (Fig. 2; Table 2).

Legnano (45.60°N; 8.92°E), is 33 km from the centre of Milan and 35 km north of the River Po in Western Lombardy, Italy with an elevation of 122 m AMSL (population: 59,000). In the Köppen climate classification system, the city has a warm-temperate climate with a uniform annual precipitation distribution (**Caf**) (Köppen, 1931) (Fig. 2; Table 2).

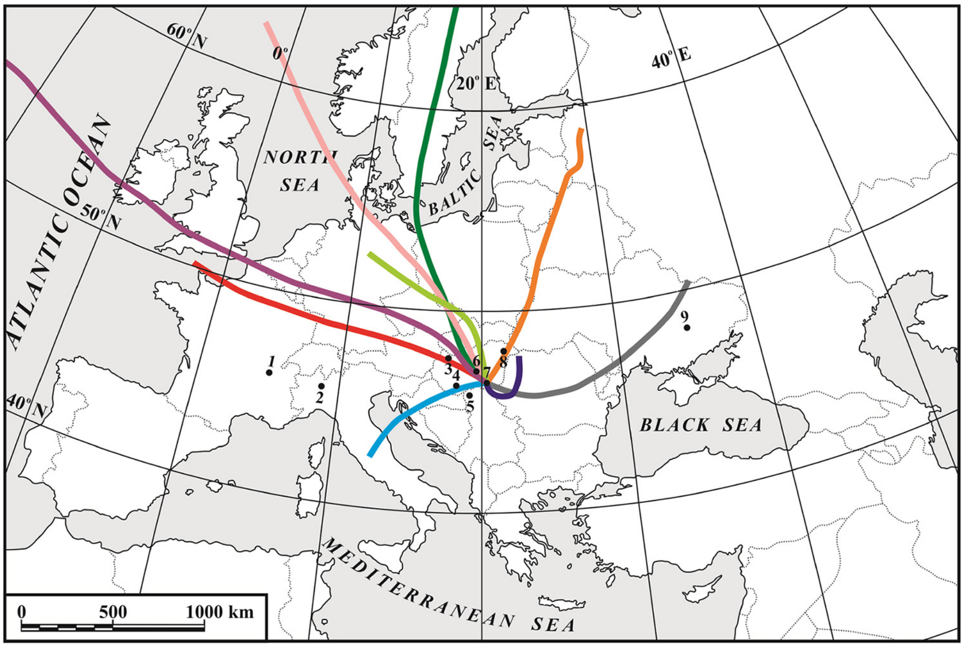

Fig. 2. Geographical location of the aerobiological stations with the centroid 3D backward trajectories of the 9 clusters, top view (see Figs. 3-4). 1: Lyon (France); 2: Legnano (Italy); 3: Győr (Hungary); 4: Pécs (Hungary); 5: Sombor (Serbia); 6: Kecskemét (Hungary); 7: Szeged (Hungary); 8: Nyíregyháza (Hungary); 9: Zaporizhia (Ukraine). The above-mentioned centroid 3D backward trajectories belong to the following clusters: red: cluster 1; **light-green:** cluster 2; **dark-blue:** cluster 3; **dark-pink:** cluster 4; **light-blue:** cluster 5; **brown:** cluster 6; **dark-gray:** cluster 7; **light-pink:** cluster 8; **dark-green:** cluster 9. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Sombor (45.77°N; 19.12°E) with an elevation of 82 m AMSL (population (48,000) is characterised by an extensive flat landscape at the North-western part of Serbia, around 20 km to the border of Hungary. Its climate is similar to that of Szeged, namely it is **Cfa** in Köppen climate classification (Köppen, 1931) (Fig. 2; Table 2).

Zaporizhia (47.85°N; 35.15°E), the sixth largest city in Ukraine (population: 771,000) is located in south-eastern Ukraine with an elevation of 50 m AMSL and lies along the Dnieper River. Its climate is **Bsk** in the Köppen system, namely a cold, dry climate with a dry summer (Köppen, 1931) (Fig. 2; Table 2).

Kecskemét (46.92°N; 19.67°E) (population: 112,000), Nyíregyháza (47.95°N; 21.72°E) (population: 118,000), Pécs (46.07°N; 18.25°E) (population: 149,000) and Győr (47.67°N; 17.60°E) (population: 128,000) on the Pannonian Plain are in Hungary. Climate of Nyíregyháza is **Dfb**, while the remaining three cites belong to **Cfb** climate in the Köppen system (Köppen, 1931) (Fig. 2; Table 2).

Rainfall deficit is characteristic for all of *Ambrosia* occurrences in Europe with high pollen load; namely, Rhone valley in France, Western Lombardy in Italy, Pannonian Plain in Central Europe including areas of Hungary, Slovenia, Croatia, Serbia, Romania and Slovakia, furthermore, the Ukraine and the south-western part of the European Russia (Ziska et al., 2007) (Fig. 2; Table 2).

2.2. Data

The pollen season of *Ambrosia* lasts form mid-July till mid-October. Seasonality of *Ambrosia* pollen concentrations is the strongest and their peak values are the highest compared to those of all taxa. They show their maximum values in the late summer – early autumn period. Ragweed favours temperate climate and prefers dry, sunny grassy plains, sandy soils, river banks, roadsides, and disturbed soils such as vacant lots and abandoned fields (Ziska et al., 2007).

For each station considered, ragweed pollen grains were collected using 7-day recording volumetric pollen traps of the Hirst design (Hirst, 1952). For the start (end) of the season for every station we used the first (last) date on which 1 pollen grain m^{-3} of air is recorded and at least 5 consecutive (preceding) days also show 1 or more pollen grains m^{-3} (Galán et al., 2001). Because the length of the pollen season varies from year to year, the longest pollen season during the study period was used for each year even if the remaining years had either later start or earlier end.

Besides Szeged, only the above-mentioned 8 ragweed pollen stations (https://ean.polleninfo.eu/ean) within Europe, representing potential source areas, were considered for the *Ambrosia* pollen season of Szeged for the period 2009–2013. Concerning long-distance ragweed pollen transport, the main habitat centres, characterised by the highest ragweed pollen levels in Europe were highlighted in the manuscript. Partly only typical stations of these habitat centres with their ragweed pollen data were selected for the analysis; namely, Lyon in the Rhône valley (France), Legnano near Milan in Western Lombardy (Italy) and Zaporizhia (South-eastern Ukraine). At the same time, another reason of involving relatively few stations in the source area analysis is the extended missing data in the individual databases (https://ean.polleninfo.eu/ean). Geographical coordinates and *Ambrosia* pollen related characteristics of the aerobiological stations involved in the study are reported in Table 2 and Fig. 2.

The meteorological data used for the computation of the back-trajectories were obtained from NOAA NCEP/NCAR Global Reanalysis database (http://rda.ucar.edu/datasets/ds090.0/#metadata/grib.html?_do=y) and were downloaded by HYSPLIT (4 parameters from NCEP Parameter Code Table 1 and 93 parameters from NCEP Parameter Code Table 2). The meteorological data have $1^\circ \times 1^\circ$ of spatial resolution as it is used in several studies (e.g. Hernández-Ceballos et al., 2014, 2015; Rojo and Pérez-Badia, 2015).

Daily meteorological data for Szeged (Environmental and Natural Protection and Water Conservancy Inspectorate of Lower-Tisza Region, Szeged, Hungary) were also used in the study for the period 2009–2013 in order to separate regional pollen transport

Table 2
Geographical coordinates, height of the pollen trap and ragweed related pollen characteristics of the aerobiological stations, involved in the study.

City (country)	Geographical coordinates		Height above sea level, m	Height of the pollen trap above ground level, m	[a]Mean start of the pollen season, day of the year	[a]Mean end of the pollen season, day of the year	[a]Mean maximum daily pollen counts, pollen grain m^{-3} of air	[a]Mean maximum daily pollen counts, day of the year	[a]Mean annual total pollen counts, pollen grain m^{-3} of air
	Longitude, °N	Latitude, °E							
Lyon (France)	4.86	45.76	173	25	210	293	282	245	1486
Legnano (Italy)	8.92	45.60	199	17	209	292	360	243	3430
Győr (Hungary)	17.60	47.67	116	20	212	283	346	242	4580
Pécs (Hungary)	18.25	46.07	128	22	203	291	582	242	6344
Kecskemét (Hungary)	19.67	46.92	130	15	203	293	702	241	8686
[b]**Szeged (Hungary)**	**20.10**	**46.25**	**80**	**18**	**196**	**288**	**685**	**243**	**5574**
Nyíregyháza (Hungary)	21.72	47.95	115	15	207	290	803	245	10,845
[c]Sombor (Serbia)	19.12	45.77	82	15	205	302	603	242	7901
Zaporizhia (Ukraine)	35.15	47.85	70	20	182	262	872	239	8430

[a] For the period 2009–2013.
[b] **bold**: target station.
[c] There were 29 missing days in the period Augus–September, 2011.

including local pollen release and long-distance pollen transport in the measured local pollen concentration. These meteorological parameters are as follows: daily mean temperature (°C), daily mean global solar flux (W m^{-2}), daily mean relative humidity (%) and daily rainfall total (mm). The distance between the aerobiological and the meteorological stations in Szeged is 2 km.

2.3. Cluster analysis and backward trajectories

Cluster analysis is a statistical technique to objectively group elements such as atmospheric trajectories using a similarity measure. The aim is to maximise the homogeneity of elements within the clusters and to maximise the heterogeneity among the clusters. Borge et al. (2007) used a two-stage clustering procedure as they observed that the original one-stage cluster analysis including all trajectories was strongly influenced by the trajectory length. Long trajectories representing fast-moving air masses were highly disaggregated, even though they often came from the same geographical region. Many short trajectories representing slow-moving air masses, however, were grouped together, although they came from very heterogeneous regions. Here, a one-step k-means clustering algorithm was used with the Mahalanobis distance (Mahalanobis, 1936), as this metric vanishes the need for two stages.

Note that Mahalanobis metric is better than the generally used Euclidean one. One can get this metric if principal component analysis is performed on the vectors, then each of the *n*-dimensional principal component vectors are standardised and cluster analysis is performed with these new vectors using Euclidean metric. In this way, Mahalanobis metric takes into account different standard deviations of the components of the vectors as well as the correlations between the components (Mahalanobis, 1936). (In other words, using Mahalanobis metric, we can avoid that major number of variables correlating with each other, namely involving less information compared to their number, are considered with unduly high weight when classifying, than lower number of variables being non-correlated and, in this way, having higher amount of information.) In order to demonstrate the role of different standard deviations take a difference of 200 km in the position of a given trajectory point. Such a difference around 1500 km from us almost goes for nothing, while the same difference counts very large near the arriving point of the trajectory. Nevertheless, several papers use Euclidean metric (e.g. Borge et al., 2007; Markou and Kassomenos, 2010; Dimitriou and Kassomenos, 2013a,b, 2014a,b).

Separation of the back-trajectory clusters and preparation of figures for clusters were performed with the help of a function called “convhull”. The algorithm (qhull procedure; http://www.qhull.org) gathers the extreme trajectory positions (positions farthermost from the centre) belonging to a cluster, which are then enclosed. More precisely, the procedure creates the smallest convex hull with minimum volume covering the backward trajectories of the clusters (Preparata and Hong, 1977; Cormen et al., 2001). Note that, to our knowledge, no one has applied 3D convex hulls besides us for inclosing groups of backward trajectories (Fig. 3).

Hegarty et al. (2013) evaluated three widely used Lagrangian particle dispersion models (LPDMs) – the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT), Stochastic Time-Inverted Lagrangian Transport (STILT), and Flexible Particle (FLEXPART) models – with measurements from the controlled tracer-release experiments Cross-Appalachian Tracer Experiment (CAPTEX) and Across North America Tracer Experiment (ANATEX). The LPDMs were run forward in time driven by identical meteorological inputs from the North American Regional Reanalysis (NARR) and several configurations of the Weather Research and Forecasting (WRF) model, and the simulations of tracer concentrations

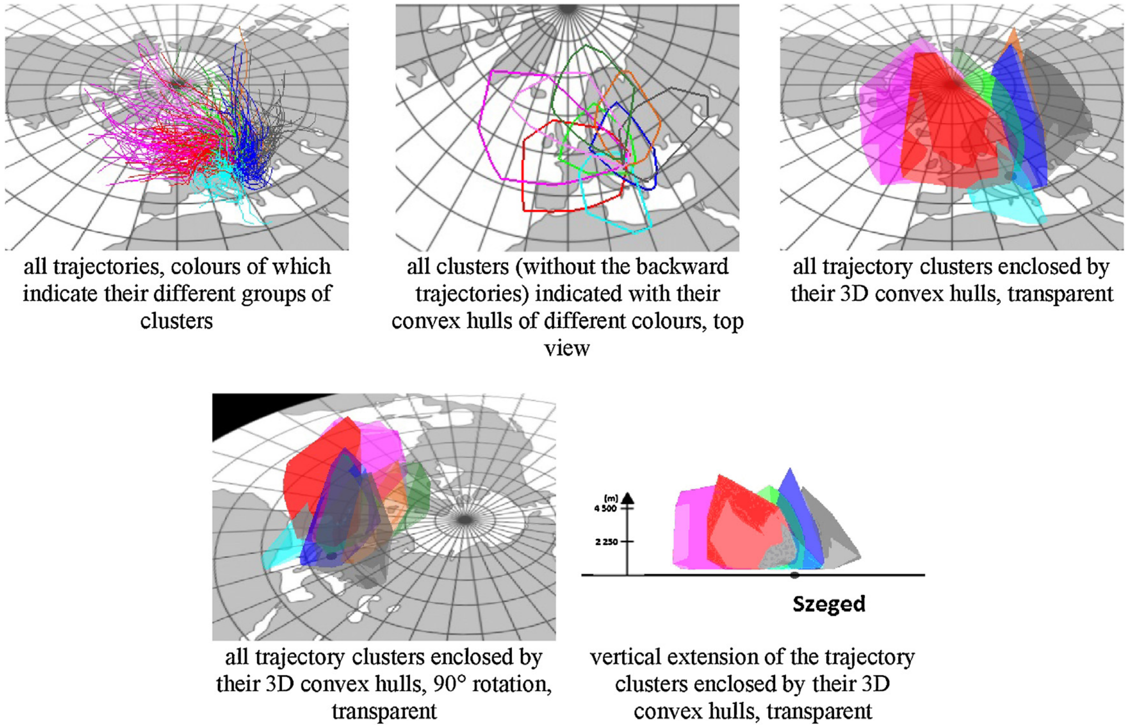

Fig. 3. 3D clusters of the backward trajectories retained, Szeged, $h = 500$ m above ground level.

were evaluated against the measurements with a ranking procedure derived from the combination of four statistical parameters (Figure of Merit in Space, Probability of Detection, False Alarm Rate and Threat Score/Critical Success Index). The statistical evaluation revealed that all three LPDMs had comparable skill in simulating the tracer plumes when driven by the same meteorological inputs, indicating that the differences in their formulations play a secondary role. The most frequently applied HYSPLIT model (according to the scientific search site "Web of Science") was decided to use in our study (Draxler and Hess, 1998).

The atmospheric backward trajectories arriving at Szeged were simulated with the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT), version 4.8 model (Draxler and Hess, 1998) of the National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory (ARL) (http://www.arl.noaa.gov/ready/hysplit4.html). This methodology allows obtaining back-trajectories hourly in 3D (latitude, longitude and elevation) from meteorological data.

Trajectory position error is typically considered to be about 20% of the traversed distance (Stohl, 1998), but the statistical uncertainty will be substantially reduced when using a large number of back-trajectories. Therefore, backward trajectories arriving at Szeged at $h = 500$ m above ground level over a 5-year period from 2009 to 2013 were taken into account. This is an altitude that is suitable to identify long-range transport impacts ensuring that the trajectory starts in the near ground atmospheric boundary layer (Karaca et al., 2009; Makra et al., 2011; Dimitriou and Kassomenos, 2014a,b). 4-day, 6-hourly three-dimensional (3D) backward trajectories (3 coordinates: φ – latitude, λ – longitude, h – height, above ground level) arriving at 1200 GMT were used. It is noted that three-dimensional trajectories were selected as they are known to be more accurate than any other type of trajectories, including isentropic ones (Stohl and Seibert, 1998).

The importance of slow moving short range back-trajectories in long-range transport of ragweed pollen (Makra et al., 2010) or PM (Salvador et al., 2010; Borge et al., 2007; Makra et al., 2011, 2013; Fleming et al., 2012; Kassomenos et al., 2012) has been reported in several studies. Namely, in short-range transport, the airflow is more influenced by emission source areas than in long range transport, where different exchange and mixing processes (e.g. deposition and advection), physical losses and chemistry have more influence on the composition at the receptor location (Fleming et al., 2012).

Clustering with the k-means algorithm was performed by using MATLAB 7.5.0 software. Trajectory clusters are projected on a stereographic projection supported by HYSPLIT (Taylor, 1997). A great advantage of the stereographic projection is that it keeps angle and, accordingly, direction as accurate as possible. In other words the projection reflects exactly from and where the back-trajectories go.

2.4. Modelling ragweed pollen transport

2.4.1. INDEX1 and INDEX2

The role of long-range transport was studied by analysing cluster occurrence on days when 24-h pollen concentrations exceeded a threshold value. This threshold was taken to be the upper quartile ($x_{0.75}$) for *Ambrosia* pollen. Based on the five-year standardised data sets, this threshold value for *Ambrosia* pollen in Szeged is 0.63.

Two statistical indices related to the probability (*INDEX1:* relative frequency of exceedance within a cluster) and frequency (*INDEX2:* ratio of exceedance in a cluster to total number of exceedances) of daily pollen exceedance episodes for every trajectory cluster were calculated as in Borge et al. (2007). For cluster i, *INDEX1* is defined as

$$INDEX1_i(\%) = \frac{D_{(x>x_{0.75})i} \cdot 100}{D_i}, \qquad (1)$$

where D_i is the number of occurrences of cluster i, and $D_{(>x)i}$ is the number of 24-h pollen exceedances in cluster i. *INDEX1* tells us the likelihood of an exceedance under a given cluster. *INDEX2* is defined as

$$INDEX2_i(\%) = \frac{D_{(x>x_{0.75})i} \cdot 100}{E}, \tag{2}$$

where *E* is the total number of pollen exceedance days. *INDEX2* may be interpreted as the likelihood of certain trajectory being present in a pollen exceedance day.

2.4.2. Potential source contribution function (PSCF)

The *PSCF* is a conditional probability function to calculate and describe possible source locations using back-trajectories. The structure of *PSCF* is based on air mass residence time allocation over specific regions in order to localise potential remote sources of transported *Ambrosia* pollen affecting local *Ambrosia* pollen level at the target area. The *PSCF* value for the *ij*th cell of the study area is defined, as follows:

$$PSCF_{i,j} = \frac{m_{i,j}}{n_{i,j}}, \tag{3}$$

where n_{ij} is the total number of back-trajectory endpoints (more precisely: given-hourly points of the back-trajectories) falling into the *ij*th cell, and m_{ij} is the number of back-trajectory endpoints in the same *ij*th cell on the days that had an *Ambrosia* pollen source contribution greater than the threshold value (Wang et al., 2006; Kocak et al., 2011; Fleming et al., 2012; Kavouras et al., 2013; Dimitriou and Kassomenos, 2014a,b). Here the threshold value may differ (Sirois and Bottenheim, 1995; Polissar et al., 2001; Hsu et al., 2003; Wang et al., 2006; Kaiser et al., 2007; Kong et al., 2013). We used the upper quartile of the daily *Ambrosia* pollen concentrations.

The study area extends from 12°W to 46°E and from 36°N to 66°N, composing thus 1740 cells of a 1° × 1° resolution in latitude and longitude. The total number of back-trajectory endpoints (more precisely: given-hourly points of the back-trajectories) located in the grid are 42,799 so that on average there are about 25 endpoints per cell.

Sparse trajectory coverage of the more distant grid cells may result in highly uncertain extreme values of *PSCF* (Polissar et al., 2001; Dimitriou and Kassomenos, 2014a). At the same time, for large values of $n_{i,j}$ there is higher statistical stability in the calculated value. In order to take into account the number of back-trajectory endpoints (more precisely: given-hourly points of the back-trajectories) that fall in a grid cell and minimise uncertainties [to reduce the effect of small values of $n_{i,j}$], *PSCF* was multiplied with a weight function *W*(*i,j*) (Polissar et al., 2001; Wang et al., 2006; Karaca et al., 2009; Kocak et al., 2011; Kong et al., 2013; Dimitriou and Kassomenos, 2014a). Namely,

$$W(i,j) = \begin{cases} 1.00 & 80 < n_{i,j} \\ 0.70 & 20 < n_{i,j} \le 80 \\ 0.42 & 10 < n_{i,j} \le 20 \\ 0.05 & n_{i,j} \le 10 \end{cases}. \tag{4}$$

W(*i,j*) reduced the *PSCF* values when the total number of the endpoints in a particular cell was less than about three times the average value of the endpoints per each cell (Polissar et al., 2001).

Note that a limitation of the *PSCF* method is that grid cells can have the same *PSCF* value when the sample concentrations are either only slightly higher or much higher than the criterion. As a result, it can be difficult to distinguish moderate sources from strong ones. PSCF and CWT calculations were performed using TrajStat software (Wang et al., 2009).

2.4.3. Concentration weighted trajectory (CWT)

In the *CWT* method, each grid cell is assigned by a weighted concentration by averaging the sample concentrations that have associated back-trajectories that crossed the grid cell as follows:

$$C_{ij} = \frac{1}{\sum_{k=1}^{N} n_{ijk}} \cdot \sum_{k=1}^{N} C_k \cdot n_{ijk}, \tag{5}$$

where C_{ij} is mean weighted *Ambrosia* pollen concentration in *ij*th grid cell, *k* is the index of the trajectory, *N* is the total number of trajectories, C_k is observed pollen concentration of *k*th back-trajectory in the receptor point on the arrival day of back-trajectory *k*, and n_{ijk} is the number of *k*th back-trajectory endpoints (more precisely: number of given-hourly points of *k*th back-trajectory) in *ij*th grid cell (Hsu et al., 2003; Wang et al., 2006; Žabkar et al., 2008). Higher n_{ijk} value indicates the longer time spent in the *ij*th cell by trajectory *k* due to the fixed hourly trajectory endpoints interval. A high value of C_{ij} implies that air parcels travelling over *ij*th cell would be, on average, associated with high *Ambrosia* pollen concentrations at the receptor.

2.4.4. Causal relationship (Granger causality) between Ambrosia pollen counts at a target site and other sites

Let $\underline{X}_t$ be an *m*-dimensional random vector at time *t* representing standardised logarithmically transformed daily *Ambrosia* pollen concentrations at *m* sites. For simplicity, the target station Szeged is assigned to the first element of the vector $\underline{X}_t$.

Take a *p*th order vector autoregressive (VAR(p)) model as

$$\underline{X}_t = \underline{\underline{A}}_1 \underline{X}_{t-1} + \cdots + \underline{\underline{A}}_p \underline{X}_{t-p} + \underline{e}_t, \tag{6}$$

where $\underline{e}_t$ is a white noise vector process. Our null-hypothesis is that $a_{k,1j} = 0$, $j = 2, ..., m$, $k = 1, ..., p$, where $a_{k,ij}$ is (*i,j*)th element of matrix $\underline{\underline{A}}_k$. This means that pollen concentration at Szeged on a day is not influenced by concentrations at other sites on previous days. Otherwise there exist integers *K* ($1 \le K \le p$) and *L* ($2 \le L \le m$) such that $a_{k,1j} \ne 0$, $j = j_1 \ne 1, ..., j_L \ne 1$, $k = k_1, ..., k_K$. In this latter case, concentrations at j_1th, ..., j_Lth sites are Granger cause of concentration at Szeged. It is interpreted as that phenomena related to atmospheric circulation, such as transport or wash-out by frontal rainfalls going towards Szeged, have influence on the concentration at Szeged. For instance, as the simplest case, let the order of autoregression *p* is 1, and take only one site apart from Szeged (*m* = 2). Then componentwise writing of Eq. (7) becomes to

$$\begin{aligned} x_{t,1} &= a_{1,11} x_{t-1,1} + a_{1,12} x_{t-1,2} + e_{t,1}, \\ x_{t,2} &= a_{1,21} x_{t-1,1} + a_{1,22} x_{t-1,2} + e_{t,2}. \end{aligned} \tag{7}$$

When the null-hypothesis $a_{1,12} = 0$ is true, then pollen concentration at Szeged on a day is not influenced by concentration at the other site on the previous day. In contrast, when $a_{1,12} \ne 0$, the situation is said that concentration at the other site is Granger cause of concentration at Szeged.

After fitting the VAR model to the available data set with the least squares technique, the task is to check whether Granger causality (Granger, 1969) holds, namely to test whether the null-hypothesis is rejected at a reasonable significance level. To save space we omit the discussion of such tests, just refer to Sfetsos and Vlachogiannis (2013). The first step is, however, to determine the order of autoregression *p*. This can be done using the Akaike Information Criterion (Akaike, 1974).

2.4.5. Statistical treatment

Factor analysis identifies linear relationships among examined variables and thus helps to reduce the dimensionality of a high-dimensional data set of correlated variables expressing them in terms of fewer uncorrelated variables (factors) (Sindosi et al., 2003) with a specific accuracy. Several methods are available for determining the number of clusters to be retained (Jolliffe, 1990, 1993; McGregor and Bamzelis, 1995). We used the method of retaining the number of factors with the largest explained cumulative variance that account for at least 80% of the total variance of the original variables (Jolliffe, 1990).

Factor analysis was applied to the initial standardised data set consisting of 12 variables (3 climatic and 9 trajectory variables introduced in Section 3.4) in order to reduce the original set of variables to fewer variables. These factors can be viewed as the main climate/trajectory features that potentially influence daily *Ambrosia* pollen concentration. After performing factor analysis, a special transformation of the retained factors was performed to find out to what degree the above-mentioned explanatory variables affect the resultant variable (daily ragweed pollen concentration) and to give a rank of their influence (Fischer and Roppert, 1965; Jahn and Vahle, 1968).

One-way analysis of variance (ANOVA) was used to determine whether the inter cluster variance is significantly higher than the intra cluster variance. An *F*-test was used to check whether the difference among the cluster-averaged pollen concentrations was significant. If a significant difference among cluster-averaged pollen concentrations was found, the Tukey test (Tukey, 1985) was carried out to identify those cluster pairs that were associated with significantly different pollen grain averages. The test assumes the statistical independence of data. Consecutive pollen data, however, may be correlated and produce higher variances of the estimated means compared to uncorrelated data. The classical Tukey test was therefore modified using the variances of estimated means obtained with the help of a first-order autoregressive (AR(1)) model fitted to data for each cluster separately.

Pollen concentration exhibits a strong annual course. Therefore, before applying ANOVA, the annual course of pollen data was removed and standardised datasets were used thereafter. Standardised datasets are free of annual cycles, guaranteeing that distinguishing between average pollen levels corresponding to trajectory types is due to the types themselves and is not related to periods of the year. A standardised data is defined as the difference between original data and expected value of data divided by the standard deviation of data. As both the expected value and standard deviation have annual course they were approximated by linear combinations of cosine and sine functions with periods of one year and one half year. Unknown coefficients in these linear combinations were estimated via the least squares technique. The procedure was applied to logarithmically transformed pollen data in order to ensure the normal distribution of transformed data (Limpert et al., 2008) which is necessary for Tukey test (Tukey, 1985).

3. Results

3.1. Cluster analysis, ANOVA and Tukey-test

For Szeged, nine clusters were retained in a 3D cluster analysis (Fig. 3). The air masses associated with clusters 1 and 4 are from the Channel area south of Great Britain and the middle-eastern Atlantic region, respectively. These back-trajectories move through Northern France and the middle part of Germany and they finally turn north-westerly to Szeged. Cluster 8 air masses are originated from the north-eastern part of the Atlantic Ocean and arrive straight ahead to the target station. The air masses of cluster 9 are from Northern Europe, over the Arctic Circle and arrive nearly parallel to a longitude before arriving at Szeged. Furthermore, clusters 6 and 7 comprise purely eastern-European air masses that come to Szeged from the north-eastern and south-eastern part of the East-European Plain, respectively. Air masses associated with clusters 5, 2 and 3 include the shortest, hence slowest back-trajectories coming from the Northern Mediterranean, Northern Germany and the North-eastern Carpathians, respectively (Fig. 3, middle left panel; Fig. 4; Table 3).

Table 3
The individual clusters with the name of the source regions and their standardised mean *Ambrosia* pollen concentrations for Szeged, $h = 500$ m above ground level (**bold:** maximum; *italic:* minimum).

Cluster no.	Name of the source region[a]	Standardised mean *Ambrosia* pollen level[b]
1	The Channel area south of Great Britain	0.26
2	Northern Germany	0.09
3	North-eastern Carpathians	−0.13
4	Middle-eastern Atlantic	0.08
5	Northern Mediterranean	**0.35**
6	North-eastern Europe	*−0.37*
7	South-eastern Europe	−0.11
8	North-eastern Atlantic	−0.25
9	Northern Europe	0.17

[a] The source regions are determined on the centroid back-trajectories of the clusters.
[b] The standardised *Ambrosia* pollen values are dimensionless.

For representing Figs. 3 and 4, we wrote an own computer programme, which was encoded in Java programming language using JOGL.

Our concept with Figs. 3 and 4 was **first** to represent both the back-trajectories and their groups on (1) *different spatial ways* (stereographic projection with a view from a given angle), and (2) *a planar way* (stereographic projection, top view); and **second** to represent each cluster of the back-trajectories in top view (stereographic projection) (Fig. 4).

Daily *Ambrosia* pollen concentration exceeded its limit value (0.63) altogether on 101 days (Table 4). The highest *INDEX1* values are associated with cluster 1 (the Channel area south of Great Britain) (36.9%) and cluster 5 (Northern Mediterranean) (30.8%). Furthermore, cluster 1 and 5 show the highest (23.8%) and second highest (11.9%) *INDEX2* values, respectively. This result is consistent with these clusters having the highest mean pollen levels (Table 4). Conversely, the smallest *INDEX1* and *INDEX2* values occur in cluster 6 (North-eastern Europe).

Applying the Tukey test, 20 significant differences were detected among the possible 36 cluster pairs (55.6%) (Table 5). Only clusters accompanied with significantly different means were then analysed (Table 5).

Cluster 1 (orientation: from the Channel area south of Great Britain) is characterised by low pollen levels because the temperature sum here only slightly exceeds the threshold necessary for blooming of ragweed (1400 °C, Cunze et al., 2013). Similarly, pollen dispersion over cluster 5 (orientation: from Northern Mediterranean) is also limited as the lack of sufficient precipitation during the long-lasting and warm summers ragweed pollen production may substantially decline. However, clusters 1 and 5 play the major role in long-range ragweed pollen transport to Szeged because when entering the Pannonian Plain they uptake substantial amount of pollen along their path (Figs. 3–5; Tables 3–5).

As a result, the highest mean *Ambrosia* pollen levels, very high maximum values, slow-moving back-trajectories (for cluster 5), high portions of back-trajectories (especially for cluster 1), high number of exceedances, as well as the highest *INDEX1* and very high *INDEX2* values and high standard deviation (for cluster 1) indicating higher variability and, hence, a higher chance for daily extreme pollen episodes highlight the importance of clusters 1 and 5 in long-range pollen transport in regard to Szeged.

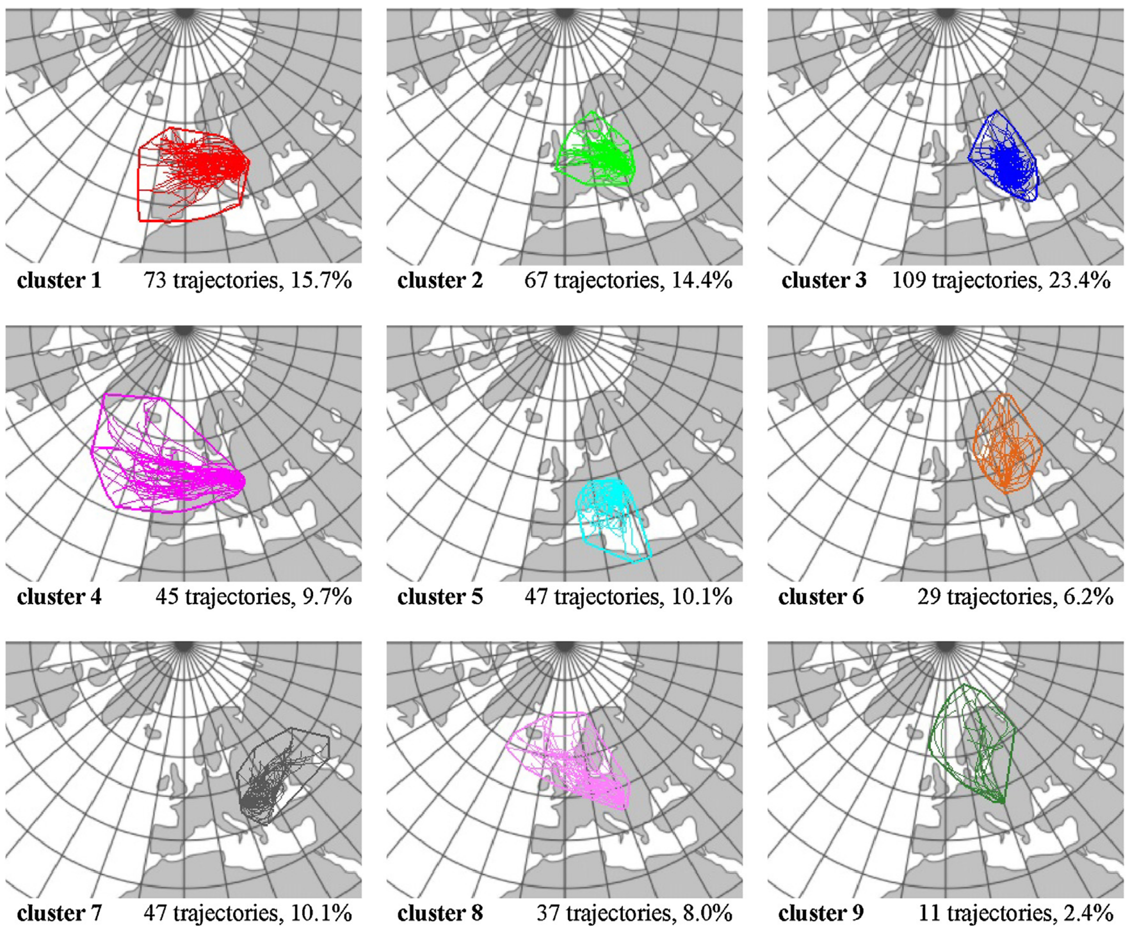

Fig. 4. The individual clusters of the backward trajectories retained, enclosed by their convex hulls, Szeged, top view, $h = 500$ m above ground level.

Table 4
Back-trajectory statistics and parameters of standardised *Ambrosia* pollen concentrations, corresponding to the individual backward trajectory clusters at 500 m above ground level arrival height (**bold:** maximum; *italic:* minimum).

Clusters, 500 m AGL	1	2	3	4	5	6	7	8	[c]9
Centroid length (km)	1131	912	*411*	2475	731	1632	978	2185	**2785**
Mean wind speed (km h^{-1})	11.78	9.50	*4.28*	25.78	7.61	17.00	10.19	22.76	**29.01**
[1]*Amb standardised average	0.26	0.09	−0.13	0.08	**0.35**	*−0.37*	−0.11	−0.25	0.17
[1]*Amb standardised maximum	2.37	1.76	1.70	1.99	1.94	*1.39*	1.70	**2.43**	2.28
[1]*Amb standard deviation	1.04	*0.83*	1.08	0.96	0.91	0.92	0.91	1.08	**1.11**
Number of back-trajectories	73	67	**109**	45	47	29	47	37	*11*
Percentage of back-trajectories (%)	15.7	14.4	**23.4**	9.7	10.1	6.2	10.1	8.0	*2.4*
Number of [a]Amb [b]exceedances	**24**	11	22	12	12	2	10	5	*3*
INDEX1 (%)	**36.9**	20.4	22.7	29.3	30.8	*9.1*	22.2	17.2	30.0
INDEX2 (%)	**23.8**	10.9	21.8	11.9	11.9	2.0	9.9	5.0	*3.0*

[a] *Amb: *Ambrosia* pollen concentration; *the standardised *Ambrosia* pollen counts are dimensionless; standardisation of daily ragweed pollen counts is the guarantee that distinguishing between average pollen levels corresponding to different clusters is due to the clusters themselves and is not related to periods of the year.
[b] Exceedances: It denotes the upper quartile ($x_{0.75}$) for *Ambrosia* pollen; i.e. the relative frequency of pollen concentrations above this threshold is 25%.
[c] Cluster 9, including less than 3.0% of the total back-trajectories is excluded from further consideration (McGregor, 1993).

Table 5
Significant differences between the standardised cluster averages of *Ambrosia* pollen concentrations, based on the Tukey test for Szeged, $h = 500$ m (in *X:* significant at $p < 0.05$, in **X:** significant at $p < 0.01$).

	1							
2	**X**	2						
3			3					
4	**X**	**X**	*X*	4				
5	**X**				5			
6			*X*			6		
7		*X*	**X**	**X**		*X*	7	
8			**X**			**X**	**X**	8
9	**X**	**X**	**X**	**X**	**X**		**X**	*X*

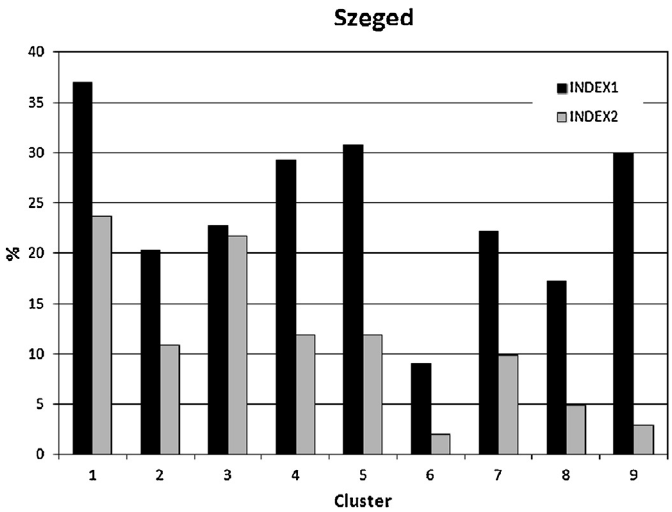

Fig. 5. Indices 1 and 2 for three-dimensional clusters of the backward trajectories for *Ambrosia* in Szeged, $h = 500$ m above ground level.

3.2. *Possible source areas detected by* PSCF *and* CWT *methods*

Based on both the *PSCF* and *CWT* values for back-trajectories the most likely sources of ragweed pollen accumulation at Szeged are located in (1) the southern and south-western part of the Carpathian Basin, including Serbia, Bosnia-Herzegovina, Croatia and Slovenia, (2) the central and eastern part of France and (3) the central and northern part of Italy only for all back-trajectories and cluster 5 (Fig. 6) (Šikoparija et al., 2009; Bullock et al., 2010; Prank et al., 2013; Šikoparija et al., 2013; Smith et al., 2013; Chapman et al., 2014; Storkey et al., 2014; Thibaudon et al., 2014).

According to these analyses, the highest ragweed pollen load over Szeged occurs when back-trajectories pass over the north-eastern part of France, Northern Italy and the Pannonian Basin within the Carpathians (Figs. 2 and 4, cluster 1; Tables 3 and 4), as well as over the south-western part of the Pannonian Basin, in addition from Northern Italy (Figs. 2 and 4, cluster 5; Tables 3 and 4). They are well-recognised as established source areas of ragweed pollen in Europe (Makra et al., 2004, 2005; Prank et al., 2013; Smith et al., 2013; Storkey et al., 2014; Thibaudon et al., 2014). Transportation performance of back-trajectories in cluster 1 is weak due to the remote source area of north-eastern France. However, they represent an important cluster in long-range *Ambrosia* pollen transport since entering the Carpathian Basin they uptake a great amount pollen beyond the 100 km threshold distance of the target area (Figs. 2–4; Tables 3 and 4). Cluster 5 comprising short back-trajectories (Dimitriou and Kassomenos, 2014a,b) and passing over an area of high ragweed pollen load (Northern Italy and the southern and south-western part of the Carpathian Basin, is also a relevant cluster in long-range pollen transport (Figs. 2–4, 6; Tables 3 and 4).

CWT values for the central and eastern part of France, as well as the central and northern part of Italy only for all back-trajectories and cluster 5 (Fig. 6) and the Carpathian Basin, are high – indicating these areas as main source regions of transported *Ambrosia* pollen. However, the *PSCF* values are not high here. This is due to the fact that besides cluster 1 the relatively clean trajectories of cluster 4 also pass over the above regions (excluding Northern Italy) thus lowering the *PSCF* scores (Figs. 2–4; Tables 3 and 4). Hence, the *CWT* approach provides a more comprehensive picture of the *Ambrosia* pollen source regions than *PSCF* (Fig. 6).

Note that individual cells with high *PSCF* and *CWT* values over the North Sea, Northern Europe and the British Isles (*PSCF* and *CWT*, all back-trajectories; Fig. 6), as well as over the Adriatic, the Mediterranean and Central Italy (*PSCF* and *CWT*, all back-trajectories; *PSCF* and *CWT*, cluster 5; Fig. 6), cannot be interpreted. Uncertainties in *PSCF* and CWT values can be explained by the (1) great day-by-day variability of the pollen concentration and (2) relatively short 5-year daily data set that is further shortened by the limited pollen season.

3.3. *Contribution of biogeographic pollen sources to pollen accumulation at Szeged*

Standardised logarithmically transformed data of the nine stations in Fig. 2 were used for the Granger causality test. The optimal order of autoregression used in the VAR model is $p = 2$. Using this test, the daily pollen concentration at Szeged is significantly (at 5% significance level) related to concentrations reported for Győr, Nyíregyháza, Kecskemét and Sombor. It is concluded, therefore, that pollen concentration over these areas significantly contributes to *Ambrosia* pollen levels at Szeged.

Returning from standardised logarithmically transformed data to original concentration data we have two estimates for the observed daily concentrations. The first is obtained from the VAR model including Szeged and the above mentioned stations being in a cause-consequence relationship with Szeged. The second estimate comes from a scalar autoregressive (AR) model fitted merely to Szeged data. It is reasonable that the difference between these two estimates should have information on transported pollen. Daily differences between these two estimates indicate that regional factors contribute solely to pollen loads 40% of the time; whereas atmospheric circulation has a significant role during 60% of the pollen season. Summarising differences between estimated daily concentrations obtained with VAR and AR models over days when VAR estimates are more accurate than AR estimates (60% of days) shows that contribution of the atmospheric circulation to the annual total pollen at Szeged is 27.8%. When VAR estimate is higher than AR estimate, the difference between them can be interpreted as transported amount. In contrast, when VAR estimate is lower than AR estimate, atmospheric circulation probably brings air of reduced pollen levels towards Szeged. To sum up, contribution of transported pollen to the annual total pollen amount at Szeged is thus 7.5%, while pollen reduction due to the atmospheric circulation counts for 20.3% of the annual total pollen.

3.4. *Identification of biogeographical pollen sources by distance*

Short-range transport is only affected by local meteorological parameters. Pollen measuring networks minimise local effects by standard sampling at a height of 10–30 m above ground level. (If the trap were at ground level, it would mainly collect pollen from the immediate vicinity of the pollen trap and the results between sites would not be comparable.) In general, volumetric pollen traps reflect the aerobiological status of the environment located within a 30 km radius of the measuring station. This validity range is the same, established by Skjøth et al. (2010), partly based on the average distance taken by a pollen grain during a day (Katelaris et al., 2004; Avolio et al., 2008). At the same time, according to Strak et al. (2012), representativity range of the pollen measurement is an area with a radius of 50 km centred by the pollen trap. If wind speed is not higher than $0.6\,\mathrm{m\,s^{-1}}$ during a 24-h period, then the pollen concentration measured by the trap is representative for this latter area.

According to the OECD Glossary of Statistical Terms (2008) the threshold of long-range transport of air pollutants is defined as follows: "Long-range transport of air pollutants (LRTAP) refers to the atmospheric transport of air pollutants within a moving air mass for a distance greater than 100 km." This is in accordance with a definition according to which the lower threshold of large scale

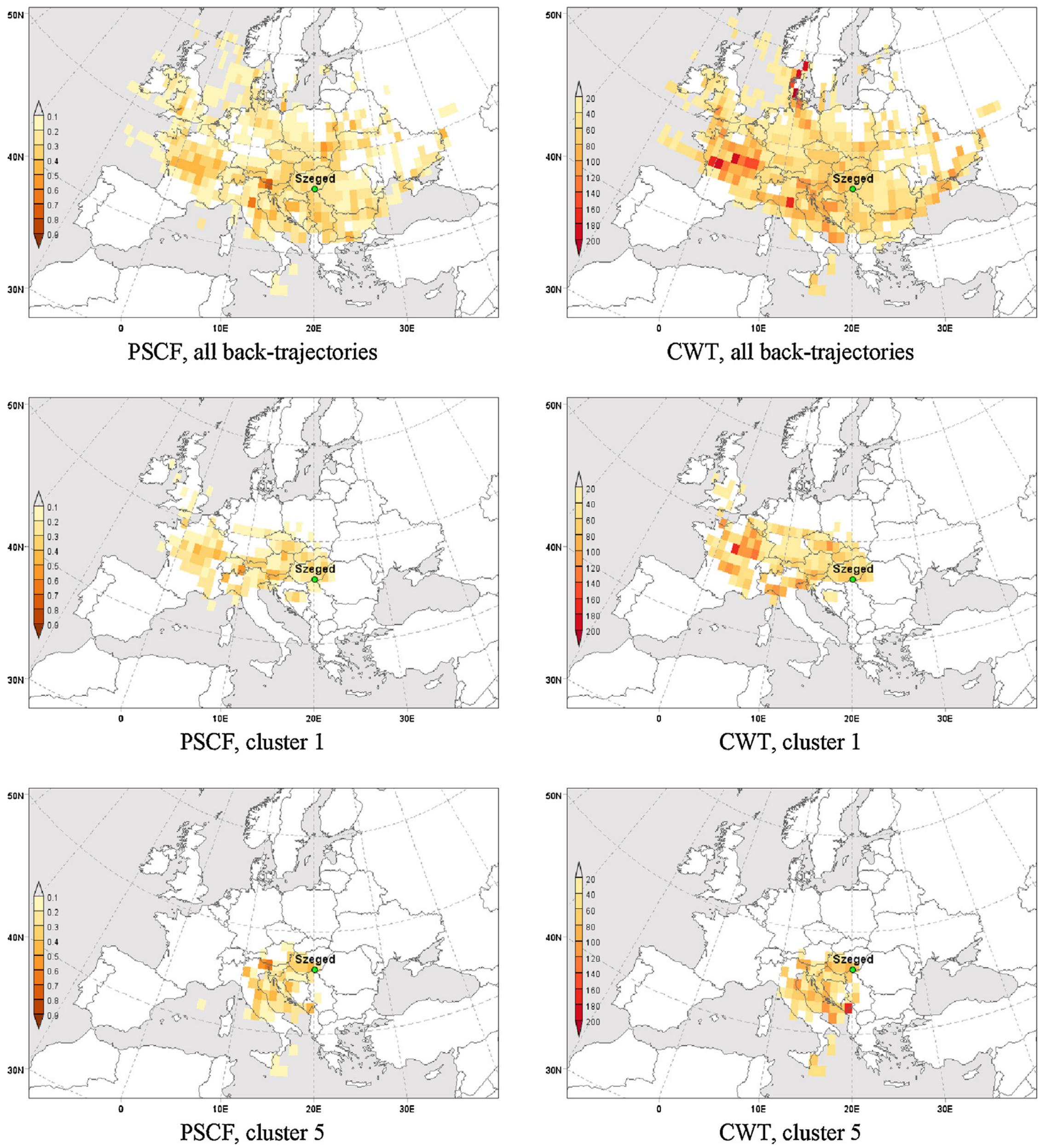

Fig. 6. Potential source contribution function (*PSCF*) and concentration weighted trajectory (*CWT*) maps of *Ambrosia* pollen for all back-trajectory and the most influential back-trajectory clusters, Szeged, 2009–2013. Darker colours indicate greater source potential.

meteorological phenomena is 100 km (WMO, 2008). This is why we selected 100 km as the lower threshold of long-range pollen transport. Under steady wind speed of 1.2 m s^{-1} and steady wind direction, pollen sampling during a 24-h period collects pollen from an area of 100 km radius (this is the radius of the medium-range pollen transport including local pollen release). Under these circumstances, pollen sampling is representative for an area of 100 km radius. Of course, stronger winds can transport pollen grains from much grater distances to the pollen trap. Namely, mean annual observed surface wind speed in Europe is 3–5 m s^{-1} (10 m height above ground level, EEA Technical Report Series, No. 6). Hence, the average travelling speed of an air mass in Europe is 11–18 km h^{-1}. Accordingly, in this latter case the measured pollen concentrations comprise both the medium-range and the long-range pollen transport, as well (Gassmann and Pérez, 2006; Makra et al., 2010).

Another classification is also known in aerobiology for investigating spatial scales. Rigid scale definitions described by Orlanski (1975) is adapted and has been used for air quality modelling (e.g. Kallos, 1998). This classification defines four scales: (1) Microscale (0–2 km): for *Ambrosia*, only Chamecki et al. (2009); (2) Meso-gamma (2–20 km): e.g. (Prtenjak et al., 2012); (3) Meso-beta (20–200 km): Appropriate model tools for ragweed simulations can be both Lagrangian (e.g. Šikoparija et al., 2013) and Eulerian type models such as COSMO-ART (Zink et al., 2012), SILAM (Prank et al., 2012) and CMAQ (Efstathiou et al., 2011); (4) Meso-alfa (200–2000 km): Almost all European studies examine the long-distance transport episodes of *Ambrosia* pollen at this scale e.g. for the Pannonian Plain (Smith et al., 2008; Šikoparija et al., 2009) or Ukraine (Kasprzyk et al., 2011; Zemmer et al., 2012) using trajectory models such as HYSPLIT (Smith et al., 2013).

Taking into account the here-mentioned classifications, we investigate in the paper the (1) long-distance *Ambrosia* pollen transport arriving at the target station beyond 100 km distance (OECD Glossary of Statistical Terms, 2008; WMO, 2008) that

Table 6
Special transformation. Effect of the explanatory variables on *Ambrosia* pollen concentration as resultant variable and the rank of importance of the explanatory variables on their factor loadings transformed to Factor 1 for determining the resultant variable, Szeged (thresholds of significance: *italic:* $x_{0.05} = 0.091$; **bold:** $x_{0.01} = 0.120$).

Variables	500 m arrival height			
	Non-rainy days		Rainy days	
	Weight	Rank	Weight	Rank
Ambrosia pollen concentration (pollen grains m^{-3} of air)	0.961	–	0.932	–
Temperature (°C)	−0.003	12	**−0.188**	4
Global solar flux (W m^{-2})	**0.391**	1	**0.178**	5
Relative humidity (%)	0.044	10	0.009	11
Mean sub-total weight	0.146	–	0.125	–
Real 3D length of the back-trajectories	**0.139**	2	−0.011	10
Length of the 3D back-trajectories as the crow flies	**0.125**	4	−0.021	9
Ratio of the real length and the "crow fly" length	0.044	9	**−0.216**	2
Average daily highest positions of the back-trajectories	−0.059	7	**−0.198**	3
Average daily lowest positions of the back-trajectories	0.035	11	**−0.171**	6
Easternmost points of the back-trajectories	0.056	8	**0.287**	1
Westernmost points of the back-trajectories	**−0.122**	5	*0.104*	8
Northernmost points of the back-trajectories	−0.075	6	*−0.118*	7
Southernmost points of the back-trajectories	**−0.132**	3	−0.008	12
Mean sub-total weight	0.087	–	0.126	–
Mean total weight	0.117	–	0.126	–

involves meso-alfa and also meso-beta scales (Orlanski, 1975) and (2) medium range transport including local pollen release, as for pollen transport from within 100 km distance to the target station (OECD Glossary of Statistical Terms, 2008; WMO, 2008) that comprises micro-sacle, meso-gamma and meso-beta scales, respectively (Orlanski, 1975).

Medium-range pollen transport is characterised next via daily mean temperature, daily relative humidity and daily global solar flux (Environmental and Natural Protection and Water Conservancy Inspectorate of Lower-Tisza Region, Szeged, Hungary). This is because ragweed pollen concentration displays a significant positive correlation with the daily mean temperature (Bartková-Ščevková, 2003; Štefanič et al., 2005; Peternel et al., 2006; Puc, 2006; Kasprzyk, 2008), and as expected it displays a negative correlation with the daily relative humidity (Bartková-Ščevková, 2003; Puc, 2006; Kasprzyk, 2008). Daily mean global solar flux is also found to be an important predictor of local ragweed pollen levels (Laaidi et al., 2003; Štefanič et al., 2005; Oh, 2009).

Long-range *Ambrosia* pollen transport is described by (1) the real 3D length of the back-trajectories, (2) the straight distance from the arrival point to the starting point (96 h before the arrival) of the 3D back-trajectories, (3) their ratio, (4) the average daily highest and (5) lowest positions of the backward trajectories based on their 4-day, 6-hourly positions. In order to take into account further characteristics of long-range *Ambrosia* pollen transport, stereographic projection of each back-trajectory is considered. The target station is located into the origin of an imaginary frame of reference. Further parameters of the long-range transport are as follows: x coordinates belonging to the (6) easternmost and the (7) westernmost points of the given back-trajectory, as well as the y coordinates belonging to the (8) northernmost and the (9) southernmost points of the same given back-trajectory. The average daily highest and lowest positions of the backward trajectories refer to the vertical transport of pollen in the atmosphere that comes from either turbulent transport dominating the vertical exchange of pollen in the boundary layer or intense convective upwelling, which results in large amounts of airborne pollen being transported from near the surface to high elevations, e.g. 3 km (Rousseau et al., 2003, 2004, 2005, 2008) or sometimes even to heights of 8–12 km (Rantio-Lehtimäki, 1994). The latter four (6–9) characteristics represent the extreme points of a backward trajectory both to west-east and south-north directions on a horizontal plane, representing the west-east and south-north extension of the long-range transport.

Days of the arrival points of the backward trajectories were divided into two groups: non-rainy and rainy days. This reveals the role of rainfall in the quantity of transported pollen (Spieksma and den Tonkelaar, 1986; Galán et al., 2000). Main conclusions of the factor analysis with special transformation performed with the above mentioned variables are as follows. On non-rainy days, medium-range transport is important, while on rainy days the two transport ranges have equal weights (Table 6).

On non-rainy days, global solar flux is in significant positive association with *Ambrosia* pollen concentration. This assumes an anticyclonic weather situation when undisturbed irradiance contributes to pollen production and dispersion, and hence an increase in pollen counts. Significant positive weights of the 3D length of the back-trajectories and those as the crow flies on *Ambrosia* pollen level indicate that longer trajectories have higher chance to uplifting and transporting pollen plumes on their path to the target area. Furthermore, substantial adverse effect of the westernmost and southernmost points of the back-trajectories denotes that these extreme trajectory points are beyond potential source areas. On rainy days, significant negative weight of temperature and substantial positive weight of global solar flux on *Ambrosia* pollen level highlights the importance of days with local shower when repeatedly increasing irradiance following temporary cooling after rainfall facilitates increasing pollen dispersion. Among the influencing variables associated to the backward trajectories the markedly negative weight of the ratio of the real length and the "crow fly" length on the pollen level indicates that the shorter the trajectory length, the smaller the chance of wash-out is. Equally important negative role of average daily highest and lowest positions of the back-trajectories refers to the dilution of transported pollen due to the increased weight of the vertical component. Important weights of easternmost and northernmost points of the back-trajectories influence *Ambrosia* pollen level proportionally and adversely, respectively. This can be interpreted by large source areas in the east and the absence of ragweed habitats in the north (Table 6).

Note that these findings are valid only for variations of daily pollen concentrations accounted for by the twelve explanatory variables and nothing is known about the variance portion not explained by these variables.

4. Discussion and conclusions

The manuscript counts to be unique in aerobiology in several aspects. According to the Web of Science only 21 papers used HYSPLIT in aerobiology; while, a mere 4 of them analysed aerobiological particles with cluster analysis (Makra et al., 2010, *Ambrosia* pollen, Table 1; Efstathiou et al., 2011, *Ambrosia* pollen, Table 1; Fernández-Rodríguez et al., 2015, Alternaria spp. spores; Hernández-Ceballos et al., 2015, Quercus ilex pollen). Table 1 comprises the most complete information in the aerobiological literature on ragweed related studies modelling long-range transport of *Ambrosia* pollen so far. Regarding ragweed pollen related papers (Table 1), surprisingly few (a mere 2 of the 23 (Makra et al., 2010; Efstathiou et al., 2011) used cluster analysis for classifying groups of individuals back-trajectories in order to localise potential source areas. We used a one-step *k*-means clustering algorithm with the Mahalanobis distance (Mahalanobis, 1936) for performing the clustering procedure. Though Mahalanobis metric is better than the generally used Euclidean one (see Section 2.3, paragraph 2); however, besides us no one has been used Mahalanobis distance in aerobiology and, according to the Web of Science, no one used this procedure before us even in papers related with the transport of chemical air pollutants when performing cluster analysis. For representing Figs. 3 and 4, we wrote an own computer programme, which was encoded in Java programming language using JOGL. Figs. 3 and 4 are also unique in representing 3D clusters of the back-trajectories in (1) *different spatial ways* [(1a) stereographic projection, view from a given angle; Fig. 3, top left, top right, down left) and (1b) side view (Fig. 3, down right)], and (2) *a planar way* (stereographic projection, top view; Fig. 3, top middle and Fig. 4). Note that, to our knowledge, no one has yet applied 3D convex hulls besides us for inclosing groups of backward trajectories (Fig. 3). In addition, significance analysis of clusters based on their standardised cluster averages has not yet been performed by others in the aerobiological special literature. We carried out this using Tukey test. Representation of potential source contribution function (*PSCF*) and concentration weighted trajectory (*CWT*) maps of *Ambrosia* pollen for indicating source potential of different back-trajectory clusters is also the first application in aerobiology. Similarly, this is the first occasion using Granger causality in aerobiological studies. Separating long-distance pollen transport and medium-range transport including local pollen release using factor analysis and special transformation is also a unique application in the international special literature including transport processes of not only biological but chemical air pollutants, as well.

After performing cluster analysis for back-trajectories, nine clusters were retained for Szeged. Among them, cluster 1 (orientation: from the Channel area south of Great Britain) and cluster 5 (orientation: from Northern Mediterranean) were selected as the most relevant clusters in terms of *Ambrosia* pollen transport to Szeged. Both cluster 1 and cluster 5 have centroid trajectories of below average length. Accordingly, slow moving back-trajectories of these clusters have higher performance in pollen transport compared to longer-range back-trajectories.

The area of cluster 1 is characterised by low pollen levels, because the temperature sum here only slightly exceeds the threshold necessary for blooming of ragweed (1400 °C, Cunze et al., 2013) (the Channel area south of Great Britain, oceanic climate). Furthermore, pollen dispersion over the area of cluster 5 is also limited, since during the long-lasting and warm summers, in the lack of sufficient precipitation, ragweed pollen production and, hence, dispersion may substantially decline (Northern Mediterranean, Mediterranean climate). However, clusters 1 and 5 play the major role in long-range ragweed pollen transport to Szeged, since when entering the Pannonian Plain within the Carpathian Basin in Central Europe, they uptake substantial amount of pollen along their path that makes them the most significant clusters regarding the transport of pollen to the target area (Figs. 3–5; Tables 3–5).

Both *PSCF* and *CWT* values delimit well the potential source areas. For all back-trajectories, the *CWT* values for the central and eastern part of France, the northern and central part of Italy, as well as the Pannonian Basin itself and its south-western part within the Carpathians, as three main source regions are high, but the *PSCF* values are not high. This can be explained by the fact that besides cluster 1, the relatively clean trajectories of cluster 4 also pass over the above regions except for Italy, thus lowering the *PSCF* scores (Figs. 2–4, Tables 3 and 4). Hence, the *CWT* approach provides a more comprehensive picture of *Ambrosia* pollen source regions than *PSCF* (Fig. 6). However, it should be noted that individual cells with high *PSCF* and *CWT* values over the North Sea, Northern Europe and the British Isles (*PSCF* and *CWT*, all back-trajectories; Fig. 6), as well as over the Adriatic and the Mediterranean (*PSCF* and *CWT* for all back-trajectories and cluster 5, respectively; Fig. 6), cannot be interpreted. In addition, the centroid 3D backward trajectory of cluster 5 comes from Central Italy (Fig. 2), however this area practically without any pollen release (Maira Bonini, personal communication) cannot be a source area of long-range *Ambrosia* pollen transport to Szeged. At the same time, the importance of cluster 5 is due to partly Northern Italy and partly the south-western part of the Pannonian Basin within the Carpathians (Figs. 2 and 4, cluster 1; Tables 3 and 4). Namely, back-trajectories passing over these areas heavily infested with ragweed habitats can transport large amount of ragweed pollen to the target area. Note that the above uncertainties in *PSCF* and CWT values are due to the great day-by-day variability of the pollen concentration and the relatively short 5-year daily data set that is further shortened by the limited pollen season.

For Szeged, the contribution of transport to the annual total *Ambrosia* pollen amount at Szeged was found to be 7.5%. This can be considered as long-range transport due to distances between Szeged on one hand, and Győr, Nyíregyháza, Kecskemét and Sombor on the other. Note that pollen levels of these latter four sites were detected as cause of pollen levels of Szeged. Although findings of previous other studies vary widely, depending on the location of the target area and its distance from the source area(s), they show good similarity with the above estimation. Namely, in Galápagos Islands, a remote archipelago lying around 1000 km from South America, long-distance transported pollen account for approximately 5% of total pollen (van der Knaap et al., 2012). In southern Greenland, 11% of total pollen measured was identified as exotic pollen (Rousseau et al., 2005, 2006). At Kevo, northernmost Finland, in some years, non-local *Betula* pollen can account for more than 20% of the annual total birch pollen amount (Hicks et al., 1994), while "extra-local" pine pollen here for the whole pollen season was found 34% (compared to 11% pre-flowering pine pollen) (Ertl et al., 2012). The only estimate in the literature for the ratio of long-range transported pollen in the measured *Ambrosia* pollen concentration was found to be up to 20% of the total ragweed pollen load over Eastern Germany that came from distant sources in Hungary through slow back-trajectories (Zink et al., 2012). Note, however, that 7.5% of Hungarian pollen levels represent a substantially higher amount than 20% of German pollen levels.

On non-rainy days, medium-range transport is important, while on rainy days the two transport ranges have equal weights (Table 6).

Besides HYSPLIT, other models are also planned to use in defining potential source areas of long-range ragweed pollen transport. Namely, the SILAM model (Sofiev et al., 2006) and the COSMO-ART model (Zink et al., 2012) will also be used in the near future for delimiting source regions and assessing the amount of long-range transported ragweed pollen at Szeged, as target area.

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